

Politehnica University Timișoara

HABILITATION THESIS

TEZĂ DE ABILITARE

From nanoscale effects to macroscopic quantities behavior:
effects of electric and magnetic fields on two-phase media

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Part A

A1. Abstract

This thesis presents the main scientific achievements after defending my PhD thesis, entitled “Contributions regarding the nucleate boiling heat transfer enhancement using high voltage electric fields”, at the Faculty of Mechanical Engineering, Politehnica University of Timisoara on November 13th, 1997 and confirmed by the Ministry of Education Order no.5653 / 22.12.1997.

Part B of this thesis has five chapters, which cover the scientific, professional and academic achievements and the development plan. The first chapter, entitled Milestones and Highlights of the scientific achievements, is presenting, in a time line manner, the contributions to the main research themes that continued and developed the subject of my PhD studies: (i) study of the liquid – vapour phase transition control by electric fields and (ii) magnetic nanofluids: heat transfer control by magnetic fields and thermal properties. The second chapter details the first theme, while the third chapter presents the main results obtained from the second theme.

The first theme emerged as a post-doctoral stage and was approached with the aim of understanding the basics of phase-change phenomena at nanoscale and how manifest at macroscopic scale in change of measurable quantities. To accomplish this, I started learning statistical thermodynamics, molecular simulation and using molecular dynamics simulation codes to obtain, through numerical experiments, answers on the posed problems. Further, I used molecular dynamics method to study the near-critical point phase transition in either two-dimensional or three dimensional systems, of hundreds up to one thousand molecules.

The post-doctoral research was carried out at Toyo University, Faculty of Engineering, Kawagoe Campus (Japan), from February 1999 to February 2000, JSPS Fellow ID P98386 of the Japan Society for the Promotion of Science, and the results were later presented in an ISI article (as first and corresponding author). The obtained results were consistent in part with the macroscopic observations and theory of the critical point phenomena and properties behaviour. Carrying out such numerical experiments after returning from this stage was a task hard to continue considering the available hardware resources but, with the technical support of Toyo University and the post-doc stage host, by granting access via internet to the university’s computing facilities, I proposed and was awarded a CNCSIS grant for Young Scientists, for a period of two years (2001–2002), to study the phase transitions of a fluid under

the influence of external force fields. Thus, the molecular dynamics simulation study was oriented toward obtaining an evaluation of the effects of an external electric field at nanoscale. This subject was also studied for both two-dimensional and three-dimensional systems of molecules. An analogy with the macroscopic observations, regarding the boiling phenomenon in applied electric fields and microgravity conditions, has been carried out in a study entitled “Micro – nanoscale effects of electric fields on boiling – modelling and simulation by molecular dynamics”, during a fellowship granted by a CNR–NATO (Consiglio Nazionale delle Ricerche, Italy – Nord–Atlantic Treaty Organisation) Senior Fellowship program, at the University of Pisa, Italy, between August and October 2004, Bando n.217.35.S del 30/04/2003. The main conclusions of this study were: (a) the increase of the system temperature partially counteracted the effects of the applied electric field; (b) the effect of the electric field on the cluster formation and orientation can be related to the experimental results regarding the “shift” of the boiling curve and change in the critical heat flux value. The results obtained from molecular dynamics simulations matched the macroscopic behaviour observations from microgravity experiments, as pointed out in the end of Chapter II.

The second theme is comprising the main results in terms of my personal scientific contributions to three subsequent research themes, whose common factor is the use of a special class of smart materials in heat transfer related problems, which were studied in a multidisciplinary team, at the Research Center for Engineering of Systems with Complex Fluids from Politehnica University Timisoara (RCESCF–UPT), in framework of five national research projects: CNCSIS A665/2005–2007; Contract UPT 11657/2001–2004 – PNCDI AEROSPATIAL; PN II 21–043/2007–2010 CFEEL; PN II PCCE IDEI 76/2010–2013; PN II 63/2014–2017, and an international research project, FP7 MNT–ERA.NET Project 7–018/2009–2011 MAFINCO. For the underlined projects, I have been the project coordinator for the Politehnica University Timisoara team and, for the international project, I have been the coordinator of the Romanian partners in the project.

This theme gave me also the opportunity to start in 2001, a long-term collaboration with the RCESCF–UPT team, whose results are seen in our common publications, part of it representing 9 (out of the 10) of my relevant publications, as well as in two national patents. My scientific contributions envisaged mainly the experimental design, running the experiments and data analyses, and several experimental set-ups have been built for this researches.

The sections of the third chapter are presenting my scientific contributions to the following research themes: (1) the fundamentals of controlling the nucleate boiling of a

magnetic nanofluid, by an applied magnetic field; (2) natural convection heat transfer in water based magnetic nanofluids; (3) thermal properties and heat transfer control by an applied magnetic field, in transformer oil based magnetic nanofluids.

The first theme, part of the research carried out in the project Contract UPT 11657/2001–2004 – PNCDI AEROSPATIAL, envisaged the possibilities of using magnetic nanofluids as cooling agents in microgravity thermal management systems. A fundamental study was carried out with respect to the dependence of characteristic parameters of nucleate boiling (bubble emission frequency and bubble departure diameter) with the applied magnetic field and the magnetic nanofluid magnetic properties. The main contributions to this research were published in 3 ISI conference papers (2 papers, as first and corresponding author), found in the list of relevant publications.

The second theme envisaged also fundamental aspects, regarding the natural convection in water based magnetic nanofluids, in view of using this type of magnetic nanofluid in cooling applications for automotive engineering. This research was started in the project CNCSIS A665/2005–2007 and continued in the project PN II PCCE IDEI 76/2010–2013, which represented a collaboration with the team from the Romanian Academy – Timisoara Branch, Laboratory of Magnetic Fluids. The main outcomes of this research were presented in 2 ISI articles (both as first and corresponding author), included in the main publication list.

The third theme had both fundamental aspects as well as application-oriented, as the targets were related to the use of transformer oil based magnetic nanofluids in electric transformers, either as cooling and insulating medium in power transformers or as magnetic liquid core in miniature planar transformers.

The first type of application was the research theme in the projects: PN II 21–043/2007–2010 CFEEL and FP7 MNT-ERA.NET Project 7-018/2009–2011 MAFINCO. My contribution to this theme envisaged the study of thermal properties, the cooling capacity and control by an applied magnetic field. The results have been published in 1 ISI article and 3 conference papers (2 papers, as first and corresponding author), all four part of the relevant publication list. Also, a national patent (co-author) has been filed, approved and published.

The second type of application is the theme of project PN II 63/2014–2017, being an ongoing research, my main scientific contribution being related to the study of thermal and electrical properties of highly concentrated magnetic nanofluids, for use as magnetic liquid core. To date, in the framework of this project, I have been studying the thermal conductivity of magnetic nanofluids, the results being part of a patent request, approved and planned for publication in March 2017.

Chapter IV of this thesis summarises the most important achievements of the candidate's academic and professional activities since defending the PhD thesis, covering the period January 1998 – present. Thus, achievements in academic activities with respect to the following aspects are highlighted: academic teaching and new courses for graduate students; published textbooks, lecture notes, teaching materials, in print or online; coordination of diploma projects and dissertations; contributions to the infrastructure for academic activities. The professional achievements during the same period are briefly presented in terms of publications, coordination of research projects, research stages, presentations at international seminars abroad, organisation of seminars and workshops, expert membership, awards, PhD referee activity, expert evaluator for national projects, ISI journal reviewer activities and professional membership.

The Vth chapter is introducing the main directions to follow for future scientific and academic activities. For short and medium term, the scientific activities will be oriented toward the ongoing project PN II 63/2014–2017 regarding the study of thermal conductivity and electrical properties of the highly concentrated magnetic nanofluids, followed by studies regarding the effects of magnetic fields on the thermal conductivity of magnetic nanofluids. As medium term directions of research are planned: (i) a study regarding the potential of using magnetic nanofluid and magnetic composite media, for heat transfer applications and thermal storage; (ii) the use of molecular dynamics to study the effect of the representative volume size on bulk thermal properties. The short term directions of research have the potential to be approached using the existing infrastructure, which would give the opportunity to enrol new PhD students in these directions of research. The medium term directions will need funding support, thus new projects proposals.

The development plan regarding the academic activity in general, the following paths are considered: integration of the research results as part of the teaching subjects, especially for graduate students, as well as integration of undergraduate and graduate students in the candidate's research team along with the new PhD students; development of the course materials and infrastructure, in such a manner to attract students toward the will of learning, academic study and research. In view of pursuing as PhD adviser, the candidate will look for the integration of new PhD students in the well-established interdisciplinary research team working at RCESCF-UPT and the research theme that are carried out or planned for; tutoring of the new PhD students, based on the experience gained as an independent researcher, regarding the way of conduct their research, in an ethical manner, such that, they will be able to accomplish their research plan and PhD thesis.

A2. Rezumat

Această teză de abilitare prezintă principalele realizări științifice obținute după susținerea tezei de doctorat, intitulată "Contribuții privind intensificarea transferului de căldură la vaporizarea globulară utilizând câmpuri electrice de înaltă tensiune", la Facultatea de Mecanică, Universitatea Politehnica Timișoara, în 13 noiembrie 1997, confirmată prin Ordinul Ministrului Învățământului nr. 5653-22.12.1997.

Partea B a acestei teze are cinci capitole, care acoperă realizările științifice, profesionale și academice și planul de dezvoltare al carierei științifice și academice. Primul capitol, intitulat "Puncte de referință și evidențierea realizărilor științifice", prezintă, în ordine cronologică, contribuțiile la principalele teme de cercetare care au continuat și dezvoltat subiectul studiilor doctorale: (i) studiul controlului tranziției de fază lichid - vapori cu ajutorul câmpurilor electrice și (ii) nanofluide magnetice: controlul transferului de căldură cu ajutorul câmpurilor magnetice și proprietăți termice. Al doilea capitol detaliază prima temă, în timp ce al treilea capitol prezintă principalele rezultate obținute din studiul celei de-a doua teme.

Prima temă a apărut în cadrul unei burse post-doctorale și a fost abordată cu scopul de a înțelege bazele fenomenelor de tranziție de fază la scară nano și modul în care acestea se manifestă la scară macroscopică prin modificarea cantităților măsurabile. În vederea realizării sale, am început studiul termodinamicii statistice, al simulării la scară moleculară și utilizarea dinamicii moleculare și a unor programe de calcul dedicate, pentru a obține, prin intermediul unor experimente numerice, răspunsuri la întrebările puse. În continuare, am utilizat metoda dinamicii moleculare pentru a studia tranziția de fază din apropierea punctului critic în sisteme bi-dimensionale și tri-dimensionale, conținând câteva sute până la o mie de molecule.

Cercetarea post-doctorală a fost întreprinsă la Universitatea Toyo, Facultatea de Inginerie, Campusul Kawagoe, Japonia, din februarie 1999 și până în februarie 2000, JSPS Fellow ID P98386, prin intermediul Fundației Japoneze pentru Promovarea Științei (JSPS), iar rezultatele au fost mai apoi prezentate într-un articol cotate ISI indicat în lista de publicații relevante (prim-autor și autor corespondent). Rezultatele numerice obținute au fost, în parte, în concordanță cu teoria fenomenelor de la starea critică privind comportarea unor proprietăți termodinamice. Deși realizarea unor asemenea experimente numerice reprezenta o temă greu de continuat după întoarcerea din acest stagiu, având în vedere dotările existente, cu suportul tehnic al Universității Toyo și al coordonatorului stagiului doctoral, prin accesul via internet la infrastructura de calcul din universitatea japoneză gazdă, am propus și mi-a fost

acordat un grant CNSIS AT, Programul pentru tineri cercetători, pentru o perioadă de doi ani (2001–2002), pentru studiul tranziției de fază a unui fluid sub influența unui câmp extern de forțe. Astfel, studiul de dinamică moleculară a fost orientat către obținerea unei evaluări a efectelor câmpului electric la scară nano. Acest subiect a fost, de asemenea, studiat atât pentru sisteme bi-dimensionale cât și tri-dimensionale de molecule. Pe baza cercetărilor efectuate în cadrul studiului "Efecte la scară micro/nano ale câmpurilor electrice asupra modelării și simulării fierberii prin dinamică moleculară", pe parcursul unei burse acordate de CNR – NATO Senior Fellowship Program (Italia), Bando n.217.35.S del 30/04/2003, la Universitatea din Pisa, între august și octombrie 2004 – a fost realizată o analogie cu observațiile macroscopice privind fenomenul de fierbere în prezența unor câmpuri electrice aplicate în condiții de microgravitație. Principalele concluzii ale acestor studii au fost: (a) creșterea temperaturii sistemului a contracarat parțial efectele aplicării câmpului electric, (b) efectul câmpului electric asupra formării și orientării clusterilor de molecule din simularea moleculară poate fi corelat cu rezultatele experimentale privind "ridicarea" curbei de vaporizare și creșterea valorii fluxului critic. Rezultatele obținute din dinamica moleculară au corespuns cu observațiile mărimilor macroscopice rezultate din experimentele realizate în condiții de microgravitație, așa cum este subliniat în finalul Capitolului 2.

Cea de-a doua temă principală prezentată, cuprinde cele mai importante rezultate cu privire la contribuția științifică personală la trei teme de cercetare conexe, al căror factor comun este utilizarea unei clase speciale de materiale inteligente în probleme de transfer de căldură, care au fost studiate într-o echipă multidisciplinară, la Centrul pentru Ingineria Sistemelor cu Fluide Complexe de la Universitatea Politehnica Timisoara, în cadrul a cinci proiecte de cercetare naționale, după cum urmează: CNCSIS A665/2005–2007; Contract UPT 11657/2001–2004 – PNCDI AEROSPATIAL; PN II 21–043/2007–2010 CFEEL; PN II PCCE IDEI 76/2010–2013; PN II 63/2014–2017, și proiectul internațional, FP7 MNT–ERA.NET Project 7–018/2009–2011 MAFINCO. La toate proiectele subliniate, dintre cele de mai sus, am fost director de proiect (CNCSIS A665), respectiv responsabil de proiect pentru UPT, iar la proiectul internațional am fost director de proiect pentru partea română.

Această temă mi-a dat, de asemenea, oportunitatea de a începe, în anul 2001, o colaborare pe termen lung cu echipa CCISFC–UPT, ale cărei rezultate sunt vizibile în publicațiile comune, parte dintre acestea reprezentând 9 din cele 10 publicații relevante atașate, precum și două brevete naționale. Contribuțiile mele științifice au avut în vedere în principal proiectarea experimentală (inclusiv măsurători) și analiza

datelor, având în vedere cele câteva standuri experimentale construite și testate pentru aceste cercetări.

Astfel, secțiunile celui de-al treilea capitol prezintă contribuțiile mele științifice la următoarele teme de cercetare: (1) probleme fundamentale privind controlul controlul fierberii cu bule în cazul unui nanofluid magnetic, cu ajutorul unui camp magnetic aplicat; (2) transferul de căldură prin convecție naturală în nanofluid magnetice pe bază de apă; (3) proprietăți termice și controlul transferului de căldură printr-un câmp magnetic aplicat, în nanofluid magnetice pe bază de ulei de transformator.

Tema (1), parte dintr-o cercetare realizată în cadrul proiectului Contract UPT nr. 11657/2001-2004 - PNCDI AEROSPATIAL, a avut în vedere posibilitățile de utilizare a nanofluidelor magnetice ca agenți de răcire în sisteme de management termic aflate în condiții de microgravitație. A fost realizat un studiu fundamental cu privire la dependența unor parametri caracteristici ai fierberii cu bule (frecvența de generare a bulelor și diametrul de desprindere al bulelor) cu câmpul magnetic aplicat și proprietățile magnetice ale nanofluidului magnetic. Principalele contribuții la această cercetare au fost publicate în 3 lucrări la conferințe ISI (la două lucrări fiind prim autor și autor corespondent) din lista de publicații relevante.

Tema (2) a avut în vedere aspecte fundamentale, privind convecția natural în nanofluid magnetice pe bază de apă, în vederea utilizării acestui tip de nanofluid magnetic în aplicații de răcire pentru domeniul autovehiculelor. Această cercetare a fost inițiată prin proiectul CNCSIS A665/2005-2007 și a fost continuată în proiectul PN II PCCE IDEI 76/2010-2013, acest al doilea proiect reprezentând o colaborare cu Laboratorul de Fluide Magnetice, Academia Română - Filiala Timișoara. Principalele rezultate ale acestei cercetări au fost prezentate în 2 articole cotate ISI (la ambele fiind atât autor corespondent cât și prim-autor), incluse în lista de publicații relevante.

Tema (3) a avut atât aspecte fundamentale cât și aplicative, deoarece scopurile principale au fost legate de utilizarea nanofluidelor magnetice pe bază de ulei de transformator în transformatoare electrice, fie ca mediu de răcire și izolare a transformatoarelor fie ca miez lichid în mini-transformatoare planare.

Prima dintre cele două aplicații a constituit temă de cercetare în proiectele: PN II 21-043/2007-2010 CFEEL și FP7 MNT-ERA.NET Project 7-018/2009-2011 MAFINCO. Contribuția mea la această temă a avut în vedere studiul proprietăților termice, capacitatea de răcire și controlul câmpului magnetic aplicat. Rezultatele au fost publicate în 1 articol cotate ISI și 3 lucrări la conferințe ISI (la 2 lucrări fiind atât

autor corespondent cât și prim-autor), toate patru fiind parte a listei de publicații relevante.

Cea de-a doua aplicație este tema proiectului de cercetare PN II 63/2014-2017, fiind o cercetare în curs, principala mea contribuție referindu-se la studiul proprietăților termice și electrice ale unor nanofluid magnetice de concentrație ridicată, spre utilizare ca miez magnetic lichid. Până în prezent, în cadrul acestui proiect, am studiat conductivitatea termică a nanofluidelor magnetice, rezultatele fiind parte a unei cereri de brevet, aprobate și în curs de publicare în martie 2017.

Capitolul IV al tezei cuprinde cele mai importante realizări privind activitățile academice și profesionale de la susținerea tezei de doctorat, acoperind perioada ianuarie 1998 – prezent. Astfel, au fost scoase în evidență realizările activității academice cu privire la următoarele aspecte: predare și noi cursuri introduce pentru studenții masteranzi; manuale, note de curs, lucrări de laborator publicate pe support de hârtie și online; coordonarea de proiecte de diploma și disertație, contribuții la infrastructura pentru activități academice. Realizările profesionale de pe parcursul aceleiași perioade de referință au fost prezentate sintetic, cu privire la publicații științifice, coordonarea de proiecte de cercetare, stagii de cercetare, prezentări în plen la seminarii internaționale și ateliere de lucru din străinătate, apartenențe la organisme de lucru ca expert, premii, activitatea de referent la teze de doctorat, expert evaluator al unor programe naționale de cercetare, recenzor la reviste cotate ISI din domeniul de specialitate și apartenența la asociații profesionale.

Capitolul V prezintă succint principalele direcții ale viitoarelor activități științifice și academice. În ceea ce privește activitățile științifice planificate pe termen scurt și mediu, voi avea în vedere continuarea proiectului în curs, PN II 63/2014-2017, privind:

- studiul proprietăților termice și electrice ale nanofluidelor magnetice concentrate pe bază de ulei de transformator (parte din proiectul);
- influența unui câmp magnetic și/sau electric asupra conductivității termice ale nanofluidelor magnetice.

În ceea ce privește planurile pe termen mediu, acestea vor fi materializate prin:

- (i) un studiu privind potențialul de utilizare al nanofluidelor magnetice și al mediilor compozite magnetice pentru a fi aplicate în sisteme termice de stocare și schimb de căldură eficient și
- (ii) utilizarea dinamicii moleculare pentru a studia efectul dimensiunii volumului reprezentativ asupra proprietăților termice de volum.

Planul de dezvoltare pentru activitatea academică de conducător de doctorat cuprinde:

- coordonarea studenților doctoranzi în vederea realizării planului de cercetare și a tezei de doctorat;
- îndrumarea studenților doctoranzi, în vederea dezvoltării de către aceștia a competențelor privind managementul unei teme de cercetare;
- integrarea viitorilor studenți doctoranzi în echipa de cercetare multidisciplinară din cadrul CCISFC, cu care candidata colaborează;

De asemenea, pe planul general al activității didactice academice, voi avea în vedere în continuare să introduc rezultatele cercetărilor în curricula disciplinelor predate, precum și să integrez, în măsura posibilităților, studenți din ciclul licență și master în echipa de cercetare, alături de viitorii studenți doctoranzi.

Acknowledgements

During the past 20 years, I had the great opportunity to work with internationally recognised research groups from abroad, and I start chronologically with the group led by Professor Ichiro TANASAWA† at the Institute of Industrial Science, University of Tokyo (Japan) in 1996, during the preparation of my PhD thesis, followed by the LOTHAR Lab group led by Professor Walter GRASSI, at the Faculty of Engineering, University of Pisa (Italy), where I had two short research stages in 1998 and 2004, and the Heat and Mass Transfer Lab group led by Professor Toru MAEKAWA, at the Faculty of Engineering, Toyo University (Japan), where I had a post-doctoral fellowship (1999–2000) and a follow-up collaboration. Thus, I had the chance to learn using various research techniques (theoretical, experimental and numerical) and up to date research equipments, to learn the proper way to conduct and present the research results and to work in an international team with colleagues coming from different cultures.

In 2001 I joined a well-established and internationally recognised research group in the field of magnetic nanofluids, based at the Research Center for Engineering with Complex Fluids, Politehnica University Timisoara, led at that time by Academician Ioan ANTON and at present by Dr. Ladislau VÉKÁS, corresponding member of the Romanian Academy. I have been collaborating with this research group in many national and international research projects regarding the use of magnetic nanofluids prepared by Dr. Doina BICA†, from the Laboratory of Magnetic Fluids, Romanian Academy – Timisoara Branch, in various engineering applications.

I would like to acknowledge the contribution of my colleagues Sorin Holotescu, Gheorghe Pop, Virgil Stoica, Oana Maria Marinică, Daniela Susan-Resiga and Alina Tăculescu Moacă, to the success of each project we work in together, and to thank each of them for their collaboration.

Also, my gratitude goes to my family, for their unconditioned and heartfelt support.

Part B. Scientific, professional and academic achievements and development plan

I. Milestones and Highlights of the scientific achievements

After I obtained my PhD title in 1997, my research activity developed into two main topics, whose milestones are presented in this section. The research projects and the most relevant publications related to my research activity are also outlined here.

Liquid –vapour phase transition control by electric fields

(1): **1998.** After defending the PhD Thesis, at the end of 1997, I continued studying electrohydrodynamics (EHD). The first post-doctoral project I was involved in aimed to study the control of nucleate boiling process of dielectric liquids in microgravity conditions, being funded by the Italian Space Agency, at the Low Gravity and Thermal Advanced Research Laboratory (LOTHAR), University of Pisa, Italy (Di Marco & Grassi, 2002). As invited researcher, between June and September 1998, I contributed to this project with my expertise in the field of EHD, gained during the PhD studies, and prepared a state of the art review, introducing a set of criteria to classify the analytical and numerical models for the vapour and gas bubbles behaviour in electric field, as follows: type of the applied field (DC or AC), geometry of the applied field (uniform or non-uniform with end effects), temperature dependence of the fluid electrical properties, gravitational field. A synthesis of this work was included later in a monography regarding the electrohydrodynamic enhancement of boiling heat transfer (Stoian, 2000).

(2): **1999 – 2000.** In my quest for understanding the physical phenomena involved in the liquid – vapour phase transition in the presence of an electric field at microscopic level and how these are reflected in the thermodynamic properties of the system and in its structure, I began studying the molecular simulation during a one year post-doctoral stage at Toyo University, Faculty of Engineering, Heat and Mass Transfer Laboratory (Kawagoe Campus, Japan), under the

supervision of Professor Toru Maekawa, with funding awarded by the Japan Society for the Promotion of Science (JSPS Post-doctoral Fellowship Program, February 16, 1999 – February 16, 2000, JSPS Fellow ID P98386). The subjects studied were: molecular modelling by equilibrium molecular dynamics, the first and second order phase transitions of fluids. I carried out a study on the structure and thermodynamic properties of a Lennard – Jones type of fluid near the critical state (Stoian, Morimoto, Sato and Maekawa, 2002– Paper no.1).

(3): 2001–2002. The subjects of the post-doc research were further developed in my home university, together with a team of colleagues and students, through a national research grant CNCSIS AT, entitled “Study of phase transitions of a fluid under the influence of external force fields”, Theme 47/CNCSIS code 147/2001 and Theme 11/CNCSIS code 75/2002 (Stoian, Holotescu, Pop, 2001; Stoian, Holotescu, 2002; Stoian, Holotescu and Maekawa, 2002). To accomplish the simulation runs carried out during the period of this grant (2001 – 2002), the JSPS post-doc host scientist, Professor T. Maekawa, and host university, Toyo University (Japan), offered access to their lab computing infrastructure, via an internet connection.

(4): 2004. Using molecular dynamics simulation, I carried out a study regarding the effect of electric fields on boiling phenomenon at nanoscale. Several two-phase thermodynamic states (corresponding to liquid–vapour mixture states at macroscale) for a dielectric fluid and the effects of an applied external electric field on the internal structure and thermodynamic properties, when the effect of the gravitational field is neglected (‘) were analysed (Stoian, 2004). This research was carried out during a short term CNR – NATO Senior Fellowship (Bando n.217.35.S del 30/04/2003) at the University of Pisa (Italy), from Aug. 25th to October 25th 2004, the host scientist being once again Professor W. Grassi (Head of LOTHAR Lab).

Magnetic nanofluids: heat transfer control by magnetic fields and thermal properties

I began studying and using magnetic nanofluids in 2001, when I joined an interdisciplinary team composed of chemists, physicists and engineers from the Research Center for Engineering of Systems with Complex Fluids (RCESCF – UPT) and the Laboratory of Magnetic Fluids, from the Romanian Academy – Timisoara Branch (RA–TB).

II.1: 2001–2004: I started this research topic in the framework of a national research project entitled: “Studies and researches regarding the behaviour of magnetizable nanofluids with micro– and nano– complex structure in terrestrial and microgravity conditions” (Contract UPT 11657/2001–2004– PNCDI AEROSPATIAL) with the team from RCESCF – UPT and colleagues from my department. Several students joined us for their graduating projects.

One major theme of this research project envisaged researches regarding the heat transfer in magnetisable multiphase multicomponent media. In view of clarifying the mechanism of bubble formation and departure in boiling of magnetic nanofluids (Vekas et al, 2000), an experimental set-up for studying the effect of an applied magnetic field on the bubbles dynamics in magnetic nanofluid was built. Thus, the dependences of bubble dynamics characteristic parameters – bubble departure diameter and bubble frequency, on the values and orientation of the magnetic field gradient with respect to gravity, were determined (**Stoian et al, 2003 – Paper no.2; Stoian et al, 2010 – Paper no.3**).

During this period and in the framework of this project and others that followed, I gained experience regarding the characteristic properties of the magnetic nanofluids, especially the measurement and analysis of magnetic properties of magnetic nanofluids (Bica et al, 2002b; **Bica et al, 2004 – Paper no.4**; Vekas et al, 2005) carried out at the Laboratory of Magnetometry (RCESCF – UPT).

II.2:2004–present.

Starting 2004, my research work was focused on studying the heat transfer in magnetic nanofluids and its control by an applied magnetic field, and the thermal properties of magnetic nanofluids, as follows:

- convective heat transfer in water based magnetic nanofluids, in view of use it as heat transfer fluids;

This topic was studied in the framework of the CNCSIS type A Project Code A665 entitled “Researches regarding the use of magnetisable nanofluids as heat transfer fluid”, 2005–2007, and the Project PN II PCCE IDEI 76, entitled “Surface science: physics, chemistry, biology and applications”, 2010–2013. The influence of the magnetic nanofluids physical properties, the working conditions and set-up configuration on the heat transfer coefficient were analysed. (Stoian et al, 2008– Paper no.5; Stoian & Holotescu, 2012 – Paper no.6).

- thermal properties and heat transfer control by an applied magnetic field, for transformer oil based magnetic nanofluids;

This research was started in the framework of the project PN II 21–043/2007–2010, entitled “CFEEL – Functional compatibility of special electrotechnical equipments with ferrofluids”, with the team of the Magnetic Fluids Laboratory, Romanian Academy – Timisoara Branch, partner in this national project coordinated by ICPE–CA Bucharest¹. My main contribution to this project consisted in the evaluation of the heat transfer characteristics and heat capacity of the magnetic nanofluid used as cooling and insulation fluid in a power transformer (Morega et al, 2008; Morega et al, 2010 – Paper no.7; Patent no. 126613 B1/2014).

The theme was further developed in an international project of the European Research Area, ERA–Net, entitled “Magnetic Fluid – *New Insulated and Cooling Medium for Power Transformers*” (acronym MAFINCO), financed for Romanian partners by UEFISCDI, project no. 7–018/2009–2011. This project had an international team, led by Dr. Milan TIMKO (International Project Coordinator) from the Institute of Experimental Physics, Slovak Academy of Sciences, Slovakia. The Romanian partners were Politehnica University Timisoara, Romanian Academy – Timisoara Branch and S.C. ROSEAL S.A. Odorheiu Secuiesc².

Due to the specific working condition of the power transformer (high voltage, high frequency), special care needed to be taken in adjusting the magnetic, dielectric and thermal properties of the magnetic nanofluid, in order to achieve its cooling and insulation (Timko et al, 2010; Holotescu et al, 2011 – Paper no.8; Timko et al, 2012a; Timko et al, 2012b; Stoian et al, 2013 – Paper no.9; Stoian & Holotescu, 2014 – Paper no.10).

¹ <http://www.icpe-ca.ro/proiecte/proiecte-nationale/pn-2007-2009/cfeel.pdf>

² <http://mafinco.mec.upt.ro>

- thermal properties of magnetic nanofluids used as magnetic liquid core.

The subject of using magnetic nanofluids as liquid core was approached by our research group in 2010 and was funded through a national project entitled “Environmental energy harvesting hybrid system through photovoltaic and piezoelectric conversion, DC/DC transforming with MEMS integration and adaptive storage – ASEMEMS HARVEST”, financed by UEFISCDI in the framework of the Joint Applied Research Projects, PN II PCCA 63/2014–2017, in a collaboration with research groups from ICPE–CA Bucharest (coordinator), POLITEHNICA University of Bucharest and S.C. SYSCOM CONTROL SRL Bucharest³.

My main scientific contribution to this project, which is an ongoing research, is regarding the thermal and electrical properties of highly concentrated magnetic nanofluids. Till present, the main outcomes of this research from our university’s team are a national patent request, filed on October 7, 2016 at OSIM (Patent request no. A/00713/07.10.2016, approved, to be published) and a book chapter (Pislaru–Danescu et al, accepted for publication, Intech Open, 2017).

³ <http://www.icpe-ca.ro/proiecte/proiecte-nationale/pn-2014/ asemems-harvest.pdf>

II. Researches on liquid – vapour phase transition and its control by electric fields

This research area emerged from the subject of my PhD studies – nucleate boiling heat transfer enhancement using high voltage electric fields (Stoian, 1997), and was extended from 1998 until 2004. It represents my post-doctoral stages and is divided into two themes: (i) the study of liquid – vapour phase transition by molecular simulation and (ii) the study of micro/nanoscale effects of external electric fields on the liquid – vapour phase transition, by molecular simulation.

II.1. Analyses on the liquid – vapour phase transition by molecular simulation

This research theme was motivated by the necessity to get an understanding of the fundamental phenomena that take part at microscopic level during liquid – vapour phase change and it started as a post-doctoral stage (1999 – 2000) at Toyo University (Japan), where I studied a molecular simulation technique– the equilibrium molecular dynamics, which I applied for the analysis of the liquid – vapour phase transition near the critical point. I continued this study in my home university, in the framework of a Grant AT – CNCSIS (National Research Grant for Young Scientists) awarded by the National Council of Scientific Research in Higher Education for the period 2001 – 2002, entitled “Study of the fluid phase transition near critical state under the influence of external force fields” (Project codes: AT 147/2001 and AT 75/2002).

II.1.1. Introduction

The phase transition at the critical state of a substance is characterised by a unique set of thermodynamic state parameters (p_c , T_c and v_c), as well as a peculiar behaviour of the thermo–physical properties of the substance and of its inner structure, as this thermodynamic state is approached from the liquid or vapour side (Hall, 1971). Although its first observation was documented almost two hundred years ago, in the experiments of Cagniard de la Tour in 1822, the experiments on the pressure – volume dependence of the liquid – vapour

coexistence line of CO₂ and introduction of the term “critical point” by Andrews in 1869 (Berche et al, 2009), gave the start to the researches aimed at describing theoretically the phenomena at critical point (CP). It is recognised nowadays that the most important contributions regarding the theoretical knowledge on the critical point phenomena belong to van der Waals, Landau, Ising, Onsager, Kadanoff and Wilson (Jany, 1988). In accordance with the developed theory, experimental studies on the thermo–physical properties of simple substances and of mixtures, demonstrated that properties like heat capacities at constant volume and at constant pressure, thermal conductivity, isothermal compressibility, adiabatic compressibility show a peculiar behaviour near CP (Pittman, Cohen and Mayer, 1982; Sengers and Sengers, 1968; Sengers, 1985; Maekawa, 1997). For instance, while the thermal conductivity, k , increases when is approaching CP, the thermal diffusivity, D_T , decreases, due to the increase of the isobaric heat capacity, c_p (Sengers and Sengers, 1968). Another peculiar phenomenon that is associated with the approaching of CP is the adiabatic or piston effect, related to the process of temperature change in the fluid (Smit and Frenkel, 1991; de Bruijn, 1999). Moreover, the internal structure of a fluid near CP shows a different pattern compared to that of other regions of the phase diagram (Okumura et al, 2000), its study being of interest for long time in connection with the applications of supercritical fluids, especially those in chemical engineering (Oshii and Okazaki, 1997; Tassaing et al, 2010; Lei et al, 2016) and, more recently, in connection with experimental evidence of new thermodynamic boundaries in the $p - V$ diagram, in the supercritical region (Bolmatov et al, 2015; Prescher et al, 2016).

The study realised during my post–doc research stage at Toyo University (Japan), envisaged the internal structure of a system of molecules near the critical point and several thermodynamic properties for states near the critical point, using the molecular dynamics method. An in–house made Fortran code was adapted for the use in this study, together with routines to analyse the system behaviour in imposed thermodynamic states. The analyses were carried out for a two–dimensional (2D) system. Let us mention here that the 2D systems are interesting for study in real applications, for instance those regarding adsorption on solid or liquid interfaces. For example, Luo and Dai (Luo and Dai, 2007) studied the self–assembly at water– trichloroethylene interfaces and the effect of hydrocarbon nanoparticles and SDS (sodium dodecyl sulfate) surfactants on such systems, using molecular dynamics in two dimensions. When we compare a 2D system with its 3D analogue, the lower interfacial tensions for the 2D systems make it harder to obtain stable two – phase coexistence in Gibbs ensemble

simulations, especially close to CP. We, therefore, searched for the values of the critical state parameters of the analysed system by calculating its static properties and comparing them with the theory of critical fluids or, when possible, with other molecular simulation results. The study was further extended to 3D molecular systems, in the framework of the project AT Grant CNCSIS mentioned above, in collaboration with colleagues from my home university and benefiting of the hardware support from the Heat and Mass Transfer Lab, Toyo University (Japan).

II.1.2. The molecular dynamics simulation procedure – A summary

II.1.2.1. BASIC THEORY

The simulation method employed – molecular dynamics (MD) – is a deterministic method, based on solving the Newton's equations of motion for each particle (atom or molecule) that belong to a system of N particles, characterised microscopically by positions and momenta:

$\mathbf{r}^N = (r_1, r_2, r_3, \dots, r_N)$ – the complete set of $3N$ coordinates of the particles contained in the system and

$\mathbf{p}^N = (p_1, p_2, p_3, \dots, p_N)$ is the complete set of $3N$ momenta of the particles contained in the system.

The force exerted by the $(N-1)$ particles on particle “ i ” is:

$$\mathbf{F}_i(t) = m \ddot{\mathbf{r}}_i(t) \quad (\text{II.1})$$

which is derived from the potential energy, $U(\mathbf{r}^N)$:

$$\mathbf{F}_i(t) = -\frac{\partial U(\mathbf{r}^N)}{\partial \mathbf{r}_i} \quad (\text{II.2})$$

By integrating the Eq. (II.1) for an enough large number of time steps, one can obtain the particles trajectories and momenta for a definite time interval, from which time averages, $\langle A \rangle_t$, defined as:

$$\langle A \rangle_t = \lim_{t \rightarrow \infty} \frac{1}{t} \int_{t_0}^{t_0+t} A(\tau) d\tau \quad (\text{II.3})$$

can be further obtained. Eq. (II.3) assumes that the system is at equilibrium.

To attain the goal of having relevant data for the system at equilibrium, long enough runs have to be carried out. Because time steps of up to $2 - 3 \cdot 10^{-15}$ s are required for numerical stability, an MD run has a duration that can last at present up to about few milliseconds (Mori et al, 2016; Dror et al, 2013), whilst during the period in which the reported personal research work was carried out, was of less than 100 ns (Haile, 1997). Still, even nowadays, MD simulations are limited by the computational speed and that dedicated supercomputers are built for such numerical experiments (Mori et al, 2016).

According to the molecular theory of matter, macroscopic properties result from the behaviour of collections of individual molecules. Thus, any system property can be interpreted as a function $A(r^N, p^N)$ that is depending on the system position in the phase space $\{r^N, p^N\}$. Therefore, once the analysed system is at equilibrium, the initial moment cannot affect the time average so the ergodic hypothesis can be applied:

$$\langle A \rangle_t = \langle A \rangle_{ens} \quad (II.4)$$

where $\langle A \rangle_{ens}$ is the ensemble average (Haile, 1997). The ensemble from statistical physics, is a collection of points in the phase space satisfying the conditions of a particular thermodynamic state, characterised by a partition function, Q_{ens} , such that $\Psi_{ens} = \ln Q_{ens}$, where Ψ_{ens} is a thermodynamic potential.

Our MD calculations were carried out using two types of statistical ensembles (Frenkel and Smit, 1996; Haile, 1997), as follows:

- the canonical ensemble was used to relax the system and to perform the structure analysis, and
- the microcanonical ensemble was used to determine the static thermodynamic properties of the systems considered for study.

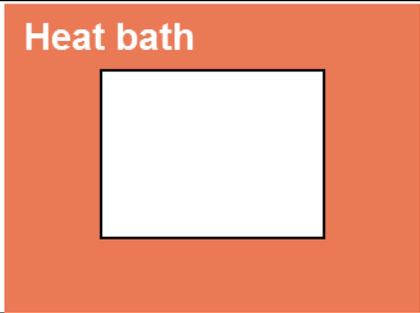
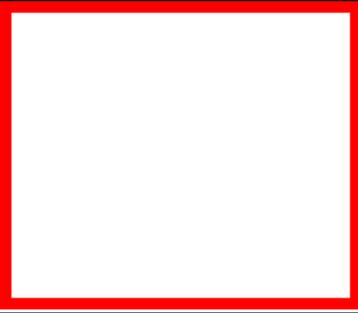
The main characteristics of the statistical ensembles used in this research work are presented in Table II.1.

The velocity distribution at equilibrium is the Maxwell - Boltzmann distribution, such that the fraction of particles that have the velocities between \mathbf{v} and $(\mathbf{v} + d\mathbf{v})$ is:

$$f(\mathbf{v})d\mathbf{v} = \frac{N(\mathbf{v})d\mathbf{v}}{N} = \frac{1}{C} \exp\left(-\frac{mv^2}{2kT}\right)d\mathbf{v} \quad (II.5)$$

where C is a constant that ensures the normalization $\int N(\mathbf{r}, \mathbf{p}) d\mathbf{r} d\mathbf{p} = N$ is satisfied.

Table II.1.

Ensemble type	Canonical ensemble (closed system)	Microcanonical ensemble (isolated system)
Constant parameters	NVT	NVE
Graphical representation		
Partition function	Q_{NVT}	Q_{NVE}
Thermodynamic potential	$\frac{F}{k_B T} = -\ln Q_{NVT}$ F – Helmholtz free energy	$\frac{S}{k_B} = \ln Q_{NVE}$ S – entropy

Eq. (II.5) can be expressed for each velocity component as exemplified below for the x-component:

$$f(v_x) dv_x = \frac{N(v_x) dv_x}{N} = \sqrt{\frac{m}{2\pi kT}} \exp\left(-\frac{mv_x^2}{2kT}\right) dv_x \quad (II.6)$$

where $C_x = \int_{-\infty}^{+\infty} \exp\left(-\frac{mv_x^2}{2kT}\right) dv_x = \sqrt{\frac{2\pi kT}{m}}$. This velocity distribution is Gaussian, with the standard deviation of $\sigma = \sqrt{kT/m}$ about the mean value $\langle v_x \rangle = 0$.

Although the velocity distribution is derived in NVT conditions, it should be Maxwellian in any equilibrium situation (Haile, 1997). For an isolated system (that is NVE conditions), the Maxwell velocity distribution is applied at $\langle T \rangle$. The velocity components have the same distribution and standard deviation, so they have the same mean square value:

$$\langle v_x^2 \rangle = \langle v_y^2 \rangle = \langle v_z^2 \rangle = \sigma^2 = \frac{kT}{m} \quad (II.7)$$

The above relation represents the equipartition of the kinetic energy. Using the velocity distribution, the average speed and its mean square value, the average kinetic energy can be calculated.

Custom tailored energy potentials have been developed for many types of interactions and, consequently, for various types of substances (Sadus, 1999). The model used in our work to describe non-bonded interactions between two particles (atoms or molecules) was the pair energy potential Lennard - Jones 12-6 (Lennard-Jones, 1931):

$$\Phi(r) = 4\varepsilon \left[\left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^6 \right] \quad (\text{II.8})$$

that has two parameters: ε – the well depth, and σ – the atom (or molecule) diameter, r being the distance between two atoms (molecules).

This intermolecular potential is still commonly used in molecular simulations (see, for example: Li, Ji and Zhang, 2016), because it describes well the non-polar molecules interactions. The first term of Eq. (II.8) is repulsive and acting at short distances, while the second is attractive, acting at long distances, as exemplified qualitatively in Fig.II.1. Parameters ε and σ can be determined for each type of molecule whose intermolecular interactions are described using this potential (Verlet, 1967; Oobatake and Ooi, 1972).

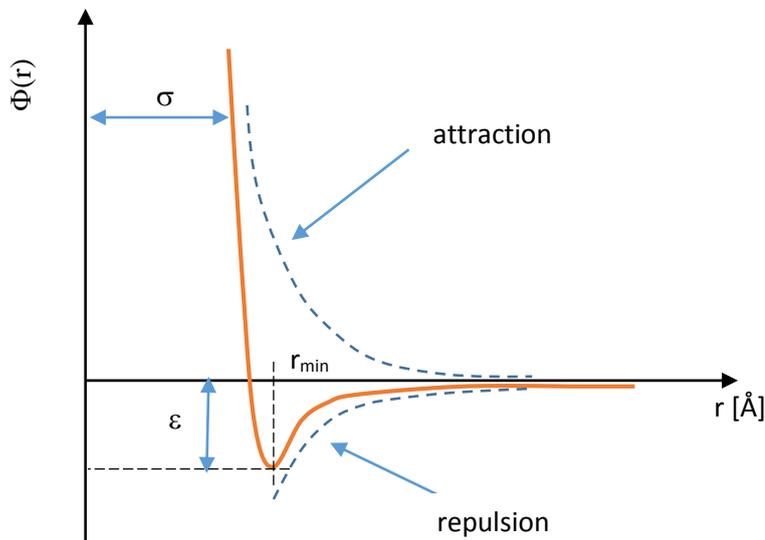


Fig. II.1. The Lennard - Jones intermolecular potential, $\Phi(r)$

The intermolecular potential is usually truncated at a cut-off radius, r_c , which is taken usually equal to $r_c = 2.5\sigma$, a value that we also employed in our

simulation. The simulations were, consequently, performed with the truncated potential, $\Phi'(r)$, expressed as follows:

$$\Phi'(r) = \begin{cases} \Phi(r), & r \leq r_c \\ 0, & r > r_c \end{cases} \quad (\text{II.9})$$

The intermolecular force between a pair of atoms is equal to $-\partial\Phi/\partial r$. Differentiating with respect to r gives a delta function at $r = r_c$. Therefore, in MD simulations the L-J potential is not only truncated, but also shifted (Haile, 1997):

$$\Phi_{\text{shift}}(r) = \begin{cases} \Phi(r) - \Phi(r_c), & r \leq r_c \\ 0 & r > r_c \end{cases} \quad (\text{II.10})$$

II.1.2.2. THE STRUCTURE ANALYSIS

This analysis uses the results of the canonical (NVT) ensemble runs and envisaged to obtain the cluster aggregation mode from the time histories of the cluster size, $s(t)$, and number of clusters in time, $N_s(t)$, for each of the analysed systems of molecules.

The dynamic scaling law (Vicsek and Family, 1984) gives the number of clusters composed of s particles (molecules), $N_s(t)$, expressed as:

$$N_s(t) \sim t^{-w} s^{-\tau} f(s/t^z) \quad (\text{II.11})$$

where w , τ and z are exponents.

Assuming that this dynamic scaling law is correct, the average number of particles in a cluster, $S(t)$, the mean radius of inertia of a cluster, $R(t)$, and the total number of clusters, $N(t)$, change with time as follows:

$$S(t) = \frac{\sum_s N_s(t) s^2}{\sum_s N_s(t) s} \sim t^z \quad (\text{II.12})$$

$$R(t) \sim S(t)^{\frac{1}{D}} \sim t^{\frac{z}{D}} \quad (\text{II.13})$$

$$N(t) = \sum_s N_s(t) \sim \begin{cases} t^{-z}, & \text{for } \tau < 1 \\ t^{-w}, & \text{for } \tau > 1 \end{cases} \quad (\text{II.14})$$

where D is the fractal dimension of the system.

The definition of a cluster necessitates the introduction of the cluster radius, r_{cl} , which represents the maximum distance between two neighbouring particles (molecules) such that they belong to the same cluster. The value of this radius is obtained from the analysis of the radial distribution function of the particles positions, $g(r)$, given by the following relation (expressed taking into consideration the employed numerical method):

$$g(r) = \frac{V}{N_i N_j} \sum_j \frac{n_{ij} \left(r - \frac{\Delta r}{2}; r + \frac{\Delta r}{2} \right)}{4\pi^2 \Delta r} \quad (\text{II.15})$$

where N_i and N_j are, respectively, the number of particles i and j , in a cubic box of volume V , n_{ij} is the number of particles j existing between the radii $r - \Delta r/2$ and $r + \Delta r/2$ from particle i .

II.1.2.3. CALCULATING THERMODYNAMIC PROPERTIES IN THE MICROCANONICAL ENSEMBLE

The analysed systems were 2D and 3D type. Therefore, the equations used for calculating the thermodynamic properties in the microcanonical ensemble are given for both type of systems in the followings.

THERMODYNAMIC PROPERTIES FORMULAE IN 3D SYSTEMS

As in the case of the microcanonical ensemble (NVE) temperature is not constant, a “kinetic temperature” is defined $T = T_Q$, such as:

$$T = T_Q = \frac{\langle Q \rangle}{n_Q k} \quad (\text{II.16})$$

where Q is the total kinetic energy per molecule, with its two components, the translational and the rotational part, and k is the Boltzmann constant. We considered only the translational component of the kinetic energy, so $n_Q = \frac{3}{2}$.

Also, the pressure is defined in the microcanonical ensemble:

$$P_m = \frac{\rho Q}{n_Q} - \frac{1}{3} \sum_{i < j} r_{ij} \frac{\partial \Phi_{ij}(r_{ij})}{\partial r_{ij}} \quad (\text{II.17})$$

where $\rho = \frac{N}{V}$, is the number density and

$\Phi(r_{ij})$ is the intermolecular energy potential.

Using the fluctuation expressions obtained for the microcanonical ensemble, the following properties can be determined:

- the volumetric heat capacity:

$$C_v = \frac{3Nk}{2} \left[1 - \frac{2}{3} \cdot \frac{N \langle (\delta Q)^2 \rangle_m}{(kT_Q)^2} \right]^{-1} \quad (\text{II.18})$$

- the adiabatic compressibility:

$$\kappa_{Sm} = \left[\frac{2P_m + \rho k T_Q}{3} + \frac{\rho k T_Q}{n_Q} + \langle \Theta \rangle_c - \frac{N \langle (\delta P_m)^2 \rangle_m}{\rho k T_Q} \right]^{-1} \quad (\text{II.19})$$

- the thermal pressure coefficient:

$$\gamma_v = \frac{k C_v^*}{n_Q} \left[\rho - \frac{N \langle \delta Q \delta P_m \rangle_m}{(k T_Q)^2} \right] \quad (\text{II.20})$$

- the isothermal compressibility:

$$\begin{aligned} \kappa_{Tm}^{-1} &= \kappa_{Sm}^{-1} - \frac{T_Q}{\rho} \frac{N \gamma_v^2}{C_v} = \\ &= \left[\frac{2P_m + \rho k T_Q}{3} + \frac{\rho k T_Q}{n_Q} + \langle \Theta \rangle_m - \frac{N \langle (\delta P_m)^2 \rangle_m}{\rho k T_Q} \right] - \\ &\quad - \frac{k T_Q C_v^*}{\rho n_Q^2} \left[\rho - \frac{N \langle \delta Q \delta P_m \rangle_m}{(k T_Q)^2} \right]^2 \end{aligned} \quad (\text{II.21})$$

- the thermal expansion coefficient:

$$\alpha_p == \kappa_{Tm} \gamma_V \quad (II.22)$$

where:

$\delta P_m = P_m - \langle P_m \rangle$ is the fluctuation of the instantaneous value of pressure from the time average value;

$\delta Q = Q - \langle Q \rangle$ - fluctuation of the instantaneous value of kinetic energy from the time average value;

$\langle (\delta P_m)^2 \rangle$, $\langle (\delta Q)^2 \rangle$ - mean square of the fluctuations;

$$\langle \Theta \rangle_m = \langle \Theta \rangle_c, \text{ where } \Theta = \frac{1}{9V} \sum_{i<j} \sum r_{ij}^2 \frac{d^2 \phi(r_{ij})}{dr_{ij}^2}$$

The non-dimensional formulae for the above listed thermodynamic properties are:

$$T^* = \frac{kT}{\varepsilon}; \rho^* = \rho \sigma^3; P^* = \frac{p \sigma^3}{\varepsilon}$$

$$C_V^* = \frac{C_V}{Nk}; \kappa_{Sm}^* = \frac{\kappa_{Sm} \sigma^3}{\varepsilon}; \gamma_V^* = \frac{\gamma_V k}{\sigma^3}; \kappa_{Tm}^* = \frac{\kappa_{Tm} \sigma^3}{\varepsilon}; \alpha_p^* = \frac{\alpha_p k}{\varepsilon}$$

THERMODYNAMIC PROPERTIES IN 2D SYSTEMS

In the case of a 2D microcanonical system, the same kind of definition of the “kinetic temperature” is employed. Also, only the translational motion of the molecules was taken into consideration, so in this type of system $n_Q = 2/2$.

The pressure function for the 2D system is expressed by:

$$P_m = \frac{\rho Q}{n_Q} - \frac{1}{2} \sum_{i<j} r_{ij} \frac{\partial \Phi_{ij}(r_{ij})}{\partial r_{ij}} \quad (II.23)$$

Again, using the fluctuations expressions obtained for the microcanonical ensemble, the thermodynamic properties of interest can be determined as follows:

- the isometric heat capacity:

$$C_V = k \left[1 - \frac{N \langle (\delta Q)^2 \rangle_m}{(kT_Q)^2} \right]^{-1} \quad (II.24)$$

- the adiabatic compressibility:

$$\kappa_{Sm} = \left[\frac{P_m}{2} + \rho k T_Q + \langle \Theta \rangle_m - \frac{N \langle (\delta P_m)^2 \rangle_m}{\rho k T_Q} \right]^{-1} \quad (II.25)$$

- the isothermal compressibility:

$$\begin{aligned} \kappa_{Tm}^{-1} = & \frac{P_m + 3\rho k T_Q}{2} + \langle \Theta \rangle_m - \\ & - \frac{k T_Q}{\rho} \cdot \left[1 - \frac{N \langle (\delta Q)^2 \rangle_m}{(k T_Q)^2} \right]^{-1} \cdot \left[\rho - \frac{N \langle \delta Q \delta P_m \rangle_m}{(k T_Q)^2} \right]^2 - \frac{N \langle (\delta P)^2 \rangle_m}{\rho k T_Q} \end{aligned} \quad (II.26)$$

We accept that $\langle \Theta \rangle_m = \langle \Theta \rangle_c$, where $\Theta = \frac{1}{4V} \sum_{i<j} \sum r_{ij}^2 \frac{d^2 \Phi(r_{ij})}{dr_{ij}^2}$.

The adimensionalisation formulae for the thermodynamic properties employed in the 2D microcanonical system are listed below:

$$P^* = \frac{P \sigma^2}{48\varepsilon}; \quad \Theta^* = \frac{\Theta \sigma^2}{48\varepsilon}; \quad v^* = \frac{1}{\rho^*} = \sqrt{\frac{m}{48\varepsilon}} \cdot v$$

$$\theta^* = \frac{kT}{48\varepsilon}; \quad U^* = \frac{\Phi}{48N\varepsilon}; \quad \Phi^* = \frac{\Phi}{48\varepsilon}$$

$$\kappa_{Sm}^* = \frac{\kappa_{Sm} \varepsilon}{\sigma^2}; \quad \kappa_{Tm}^* = \frac{\kappa_{Tm} \varepsilon}{\sigma^2}; \quad C_V^* = \frac{C_V}{Nk}; \quad t^* = \sqrt{\frac{48\varepsilon}{m\sigma^2}} \cdot t$$

II.1.2.4. THE NUMERICAL METHOD

The employed numerical method is based on the Verlet “leap-frog” algorithm (Hockey, 1970). This algorithm is characterised by a good energy conservation for short times and small deviation for long runs, uses small time steps because is not accurate for large time steps, it is fast and needs a small

memory compared to other algorithms. In this algorithm, the velocities are not defined at the same time step as the positions, so the total energy cannot be calculated directly. The velocities are calculated first and then the positions, as presented in the following relations:

$$v_i\left(t + \frac{\Delta t}{2}\right) = v_i\left(t - \frac{\Delta t}{2}\right) + \frac{\Delta t}{m_i} F_i(t) \quad (\text{II.27})$$

$$r_i(t + \Delta t) = r_i(t) + \Delta t v_i\left(t + \frac{\Delta t}{2}\right) \quad (\text{II.28})$$

$$v_i(t) = \frac{v_i\left(t + \frac{\Delta t}{2}\right) + v_i\left(t - \frac{\Delta t}{2}\right)}{2} \quad (\text{II.29})$$

The coordinates and velocities values are characterised by errors of the order $(\Delta t)^4$ and $(\Delta t)^2$, respectively.

For every analysed system, the run had three stages: initialisation, equilibration and production. The first two stages were carried out in a canonical ensemble simulation (NVT), the first representing the first 100,000 time steps (if the Lennard – Jones parameters for Argon are taken, it would correspond to ~ 30 ps), while the second continued until 1,000,000 time steps were calculated. The third stage is the production of trajectories needed for calculating the thermodynamic properties. It was carried out in a microcanonical ensemble simulation (NVE), for a duration of 3,000,000 time steps (similarly, if the Lennard – Jones parameters for Ar are taken, it would correspond to ~ 9 ns). Additionally, the calculation of the thermodynamic properties needs the average of the $\langle \Theta \rangle_c$ function, as $\langle \Theta \rangle_m = \langle \Theta \rangle_c$ was accepted. Therefore, an additional canonical ensemble simulation was performed, for a duration equal to all three stages, that is $5 \cdot 10^6$ time steps.

The structural analyses were carried out based on canonical ensemble simulations, of a duration of 100000 time steps.

Both the 3D and the 2D system were initialised from a uniform grid (as shown in Fig.II.2 and Fig.II.3).

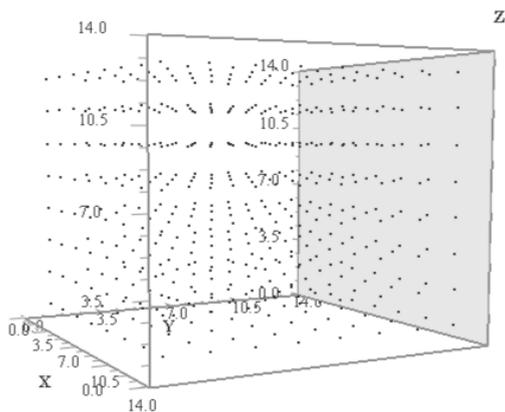


Fig.II.2. Initial positions grid in 3D

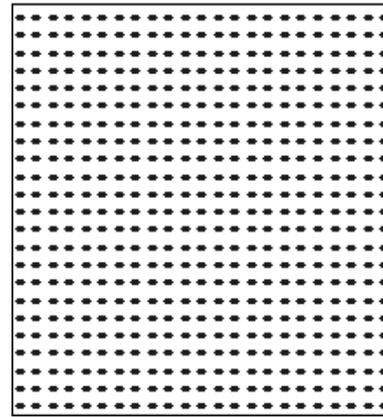


Fig. II.3. Initial position grid in 2D

Periodic boundary conditions were employed, to overcome surface problems between the fluid molecules and the walls of the container. This hypothesis implies that the primary cell that contains the N molecules that form the analysed system is surrounded by identical cells (called imaginary cells), with the same number of molecules, called images of the molecules in the primary cell. Also, both the positions and momenta of the imaginary cells are characterised by periodicity (Haile, 1997).

II.1.3. Results on the structure analysis and thermodynamic properties near the critical point of a Lennard - Jones fluid

II.1.3.1. 2D SYSTEM ANALYSES

This analysis was carried out during the post-doctoral stage, the results being presented in (Stoian et al, 2002). The 2D system consisted of $N = 529$ molecules, interacting via the Lennard - Jones 12-6 intermolecular potential (L-J 12-6). The influence of the tail of the potential was estimated analytically, assuming the radial distribution function of the molecules in the system (Eq. II.15), is equal to 1, for a distance r greater than r_c (where r_c is the cut-off radius).

We considered the cut-off radius $r_c^* = 2.5$ and a non-dimensional time step $\Delta t^* = 0.001$, that is 0.30216 fs, taking the parameters of Argon: $\epsilon = 1.67 \cdot 10^{-21}$ J and $\sigma = 3.4 \cdot 10^{-10}$ m, and its mass $m = 6.6 \cdot 10^{-26}$ kg/molecule, where:

$$t^* = \sqrt{\frac{48\varepsilon}{m\sigma^2}} \cdot t \quad [-]$$

The thermodynamic parameters for the CP for a Lennard – Jones fluid are different if modelled with one of the equations (II.8) to (II.10). Smit and Frenkel (Smit and Frenkel, 1991), showed that the interfacial tension is the driving force which causes the liquid and vapour phase separation. Moreover, the 2D system has a very low interfacial tension of the liquid – vapour interface. Also, even there are studies regarding the determination of the critical state parameters for a Lennard – Jones fluid, both in a 2D and 3D system, the results concerning the 2D system differ from author to author – see, for example, (Rovere, Heermann and Binder, 1990), (Panagiotopoulos,1994), (Frenkel & Smit,1996). In Table II.2 are presented several reference data for the critical point (expressed in non-dimensional units), as given by listed references.

Table II.2

$r_c^* [-]$	$T_c^* [-]$	$\rho_c^* [-]$	Reference
–	0.515 ± 0.002	0.355 ± 0.003	Smit & Frenkel, 1991
–	0.472	0.33 ± 0.02	Singh et al, 1990
5	0.497 ± 0.003	0.38 ± 0.01	Panagiotopoulos, 1992
2.5	0.472 ± 0.010	0.35 ± 0.01	Rovere et al., 1990
2.5	0.477 ± 0.003	0.35 ± 0.02	Panagiotopoulos, 1992
2	0.44 ± 0.005	0.368 ± 0.003	Wilding and Bruce, citat in Panagiotopoulos, 1992

where $r^* = \frac{r}{\sigma}$; $T^* = \frac{kT}{\varepsilon}$; $\rho^* = \rho\sigma^2$.

Taking into consideration the reference data for the non-dimensional critical state thermodynamic parameters of the 2D L–J fluid, we chose a set of parameters that spanned the critical region, as presented in Table II.3. States B, C, E and F are in the critical region, D is in the subcritical region and A is in the supercritical region. In Fig.II.4 are presented the chosen states together with data for the critical state given in references, which used the same cut-off radius for the 2D L–J potential.

Table II.3

state	A	B	C	D	E	F
$T^* [-]$	0.672	0.470	0.456	0.432	0.456	0.456
$\rho^* [-]$	0.338	0.338	0.338	0.330	0.330	0.350

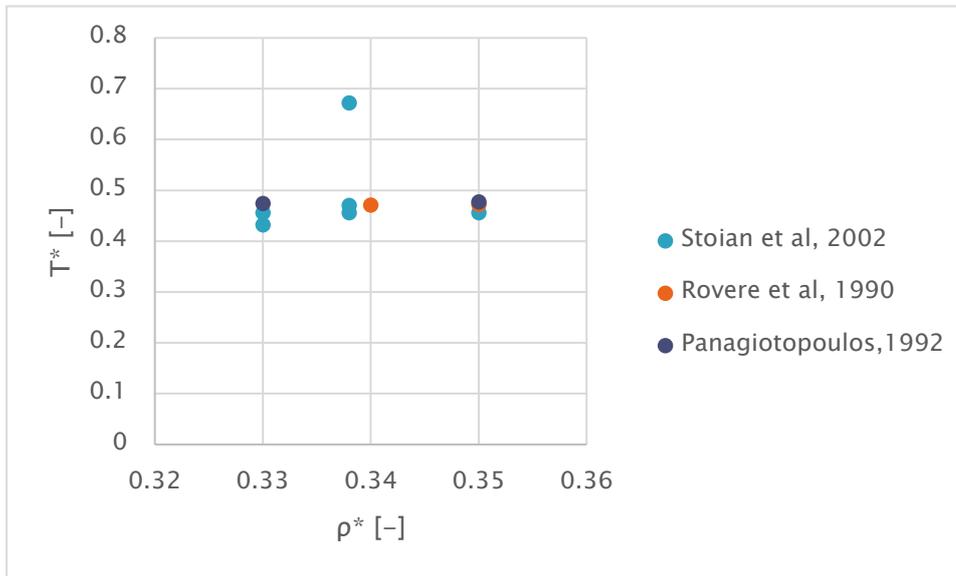


Fig.II.4. Representation of the analysed thermodynamic states (A to F), compared to the reference data for the critical state parameters of 2D L-J, using the same cut-off radius for the potential ($r_c^* = 2.5$)

CLUSTER GROWTH ANALYSIS

This analysis used the initialisation stage of the NVT simulation, that is the first 100,000 non-dimensional time steps. Then we calculated the pair correlation function of molecules, $g(r)$, expressed in Eq. (II.15). The resulting function for case C is presented in Fig.II.5.

The cluster radius is the maximum distance between two neighbouring molecules such that they belong to the same cluster. We defined the cluster formation, considering that the position of the mean point between the first and the second peak positions (1.1d and 1.7d, respectively) was at 1.4d. After the system reached the steady state, the cluster structures were investigated.

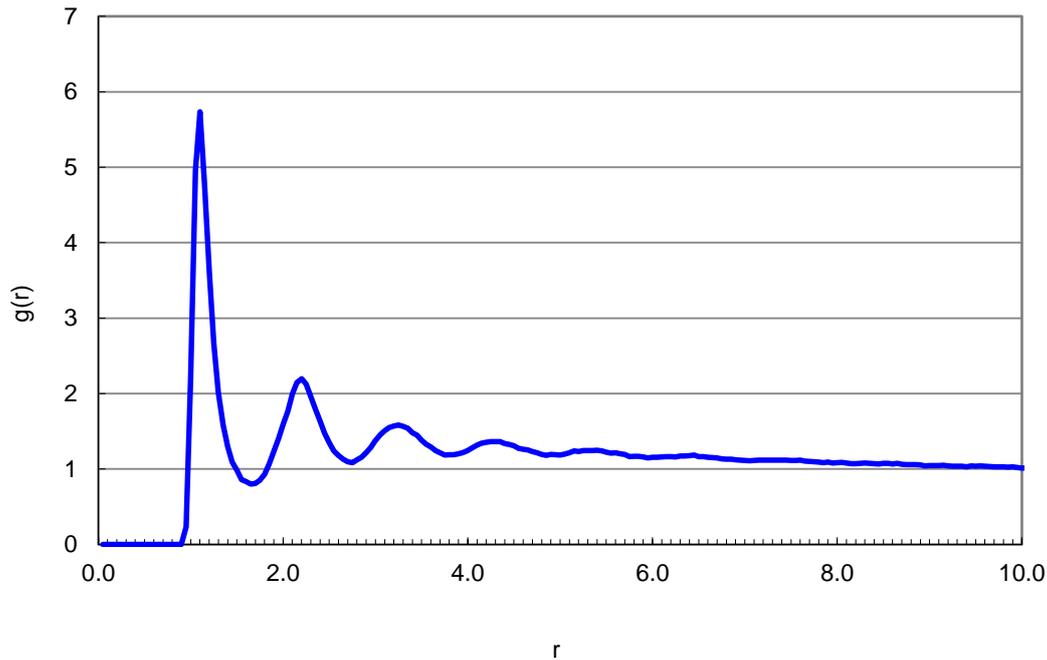
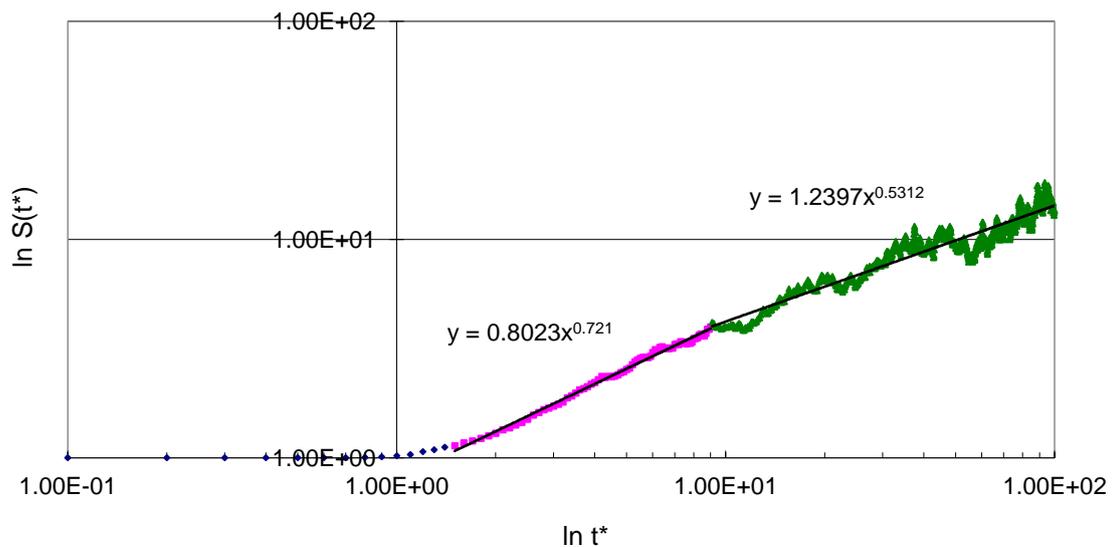


Fig.II.5 The pair correlation function for state C

We present here several results for state C, who resulted to be the closest to the actual critical point of the 2D L-J fluid. The time variations of $S(t)$ and $N(t)$, for $r_{cl} = 1.4d$, are shown in Figures II.6 and II.7, while in Fig. II.8 are presented snapshots of the positions of molecules at four time steps (the last is at the 100,000 time step). We observe a power law-like behaviour for both functions for this state, with the same exponent in absolute value.

Fig. II.6. Time variation of the average number of particles in a cluster near the critical point (state C), for the mean cluster radius $r_{cl} = 1.4d$

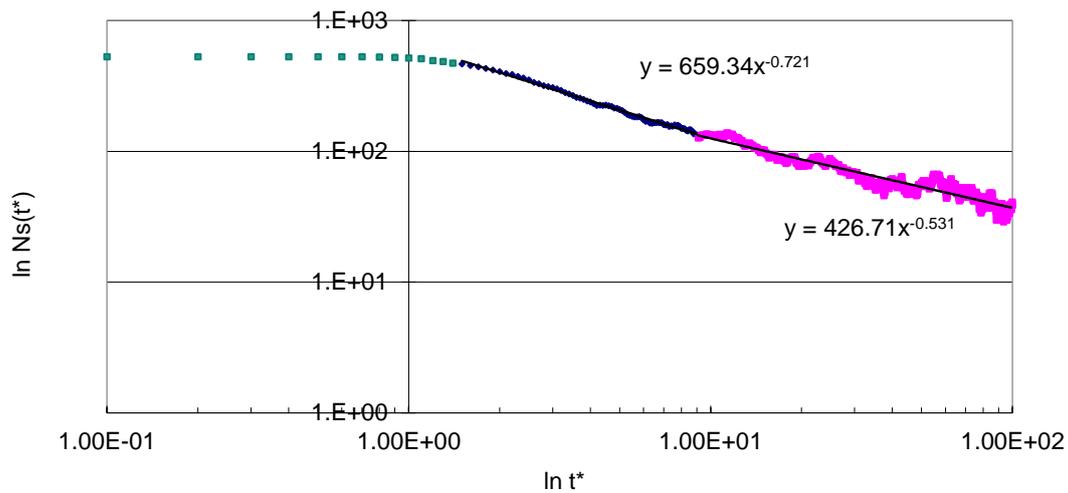


Fig. II.7. Time variation of the number of clusters in the system near the critical point (state C), for $r_{cl} = 1.4d$

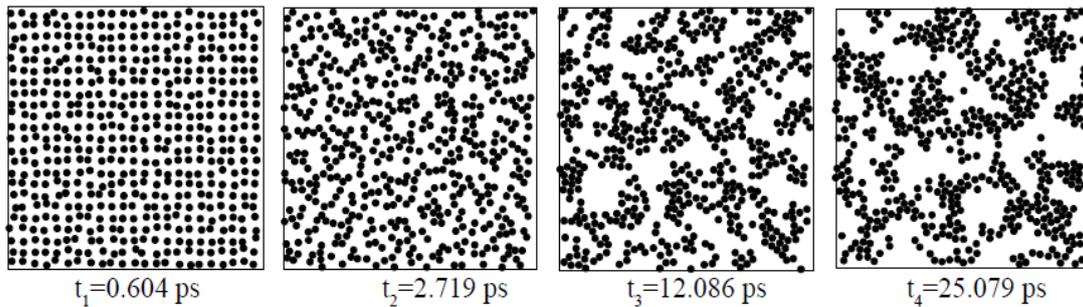


Fig. II.8. Snapshots of the positions of the $N = 529$ molecules of L-J fluid near the critical point (state C), $r_{cl} = 1.4d$

To check the influence of the cluster radius definition on the cluster growth process, these simulations were carried out for three values of the distance $r_{cl} = 1.2d$, $1.3d$, and $1.4d$. The cluster growth process did not change even when the definition of r_{cl} was changed, for the interval $1.1d < r_{cl} < 1.7d$. For the other two cases (E and F) the power law-like behaviour changes to a fluctuating behaviour much more quickly for the same time interval observed (Stoian et al, 2002).

The time variation of $R(t)$, the mean radius of inertia of the cluster, for state C is presented in Fig. II.9. We can observe a power law behaviour, very close to $3/2$ (that is 1.419). The dependence of $S(t)$ with $R(t)$ is shown in Fig.II.10. We can distinguish here two regions, described in the followings.

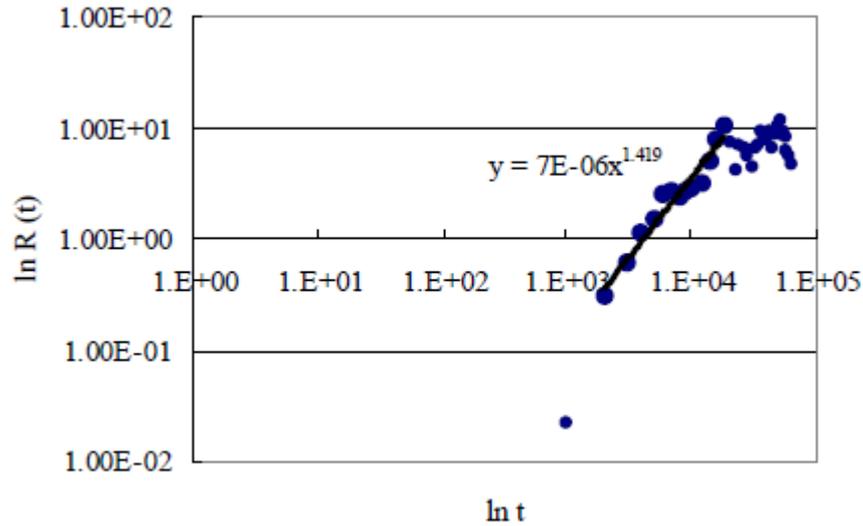


Fig. II.9. Time variation of the mean radius of inertia, $R(t)$, near the critical point (state C), for the mean cluster radius definition, $r_{cl} = 1.4d$

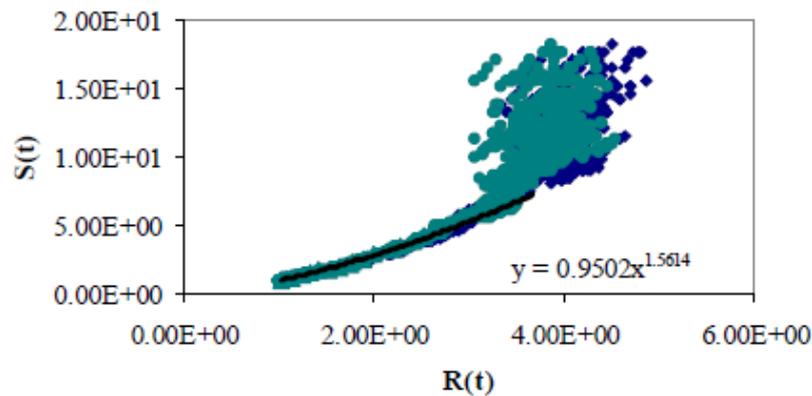


Fig. II.10. Dependence of $S(t)$ versus $R(t)$, near the critical point (state C), for the mean cluster radius definition, $r_{cl}=1.4d$ (circle symbols/green – the x component of R , rhomb symbols/blue – the y component of R)

The first region, lasting from 1,400 to about 27,000 time steps, is characterised by a power law, of approximately $3/2$ (that is 1.5614), which is equal to the fractal dimension of the system, D (see Eq. II.13). This value of D is close to that given for the diffusion-limited cluster – cluster aggregation (CCA) process in 2D systems (Meakin et al, 1985; Meakin, 1998; Vicsek & Family, 1984; Vicsek, 1992). Also, the obtained value is close to that which characterises the aggregation of ferromagnetic clusters in 2D systems (Morimoto & Maekawa, 2000; Byrom & Biswal, 2013). For instance, in the study carried out by Morimoto & Maekawa (Morimoto & Maekawa, 2000) on the cluster structure in a ferromagnetic

2D system, the fractal dimension of the system resulted 1.3, in the absence of magnetic field, irrespective of the ratio between the magnetic dipole energy of a particle and its thermal energy. More recently, Byrom and Biswal (Byrom & Biswal, 2013) analysed the aggregation of 2D system composed of a mixture of paramagnetic and diamagnetic colloids in a ferrofluid, in the presence of the magnetic field. Depending on the ratio of paramagnetic to diamagnetic nanoparticles, the fractal dimension of their system varied from 0.94 to 1.54 (± 0.3), with a parabolic shape.

The second region in Fig. II.10, that is lasting up to the end of the simulation (the interval from ~ 27000 to 100000 time steps), is characterised by scattered data. As for the other cases chosen near the critical point, that is B, E and F, the variation of $N(t)$ and $S(t)$ have a power law trend for approximately 30000 time steps, for the case B, and $40\,000$ time steps, for cases E and F. After these initial periods of simulation, appear fluctuations which are too large to be approximated by a power law (Stoian et al, 2002).

In Fig.II.11 and Fig.II.12 are presented the functions $S(t)$ and $N(t)$ for the thermodynamic states A and D, corresponding to supercritical and subcritical regions, respectively. As both two states are far from the critical region, we can observe that the power law applies only to a very limited duration from the initiation of the MD simulation (hundreds of time steps).

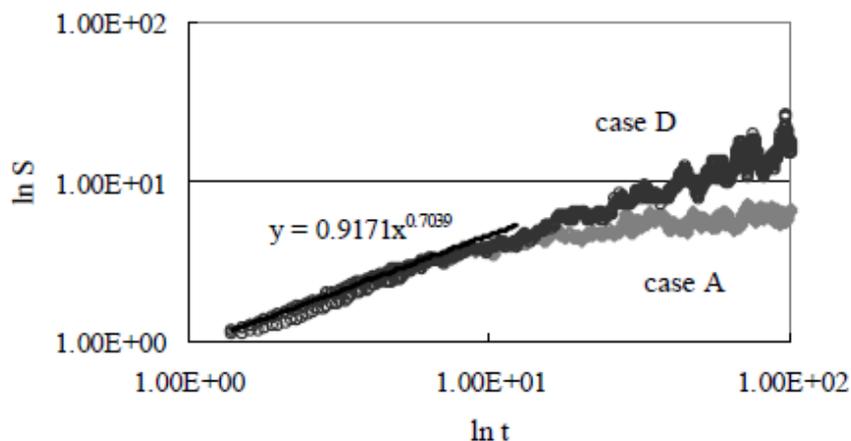


Fig.II.11. Dependence of number of clusters versus time ($r_{cl} = 1.4d$), for the subcritical region (state D) and the supercritical region (state A)

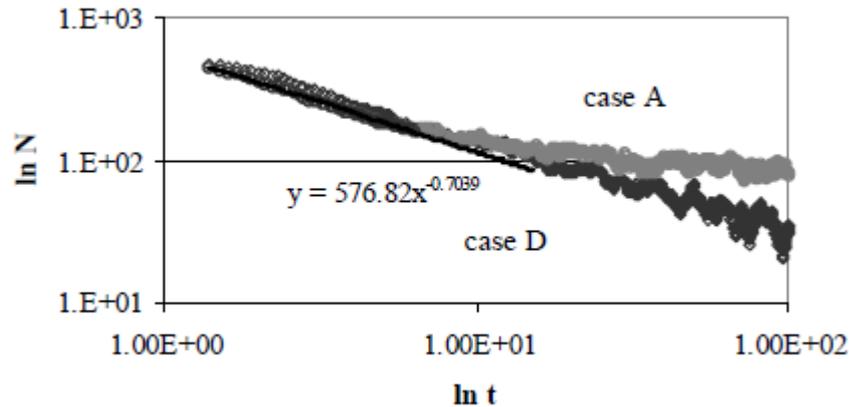
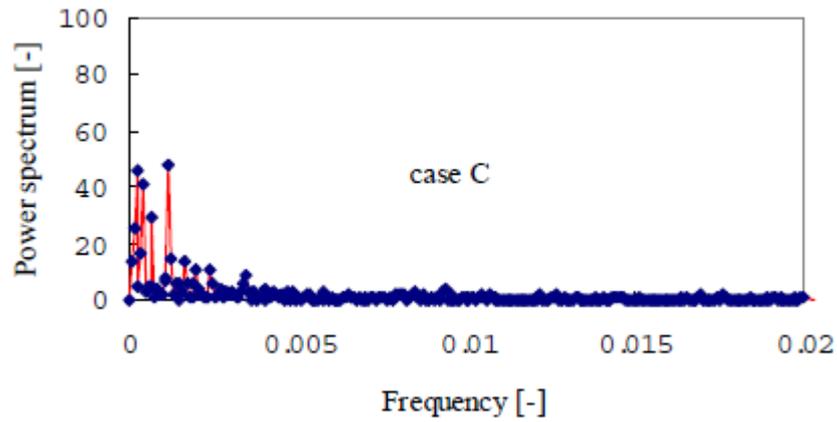


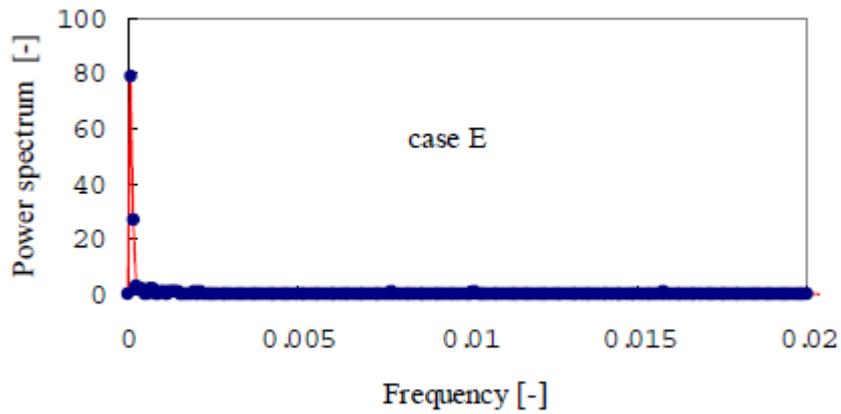
Fig. II.12. Dependence of the average number of particles in a cluster versus time ($r_{cl} = 1.4d$), for the subcritical region (state D) and the supercritical region (state A)

The cluster analysis for the six chosen thermodynamic states (A to F), indicates that state C is the closest to the critical point of the 2D L-J fluid, as the resulting fractal dimension is very close to that predicted by the theory of the cluster-cluster aggregation model in 2D particle systems.

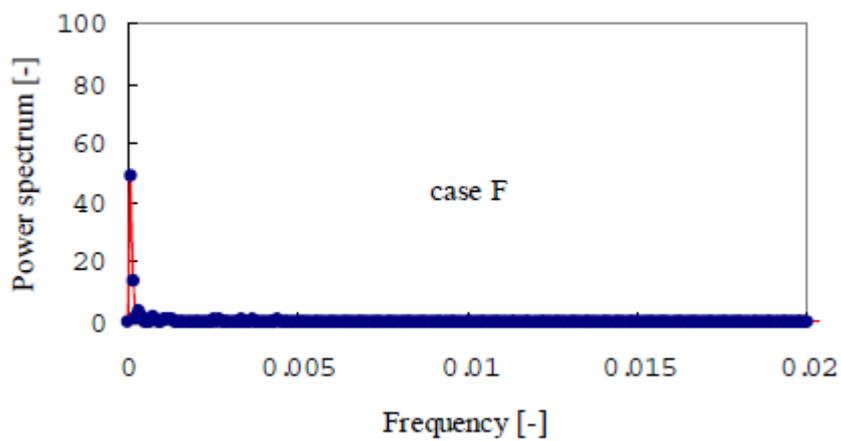
To further verify the validity of the chosen states near critical region of the 2D L-J fluid, a Fourier analysis was carried out for the states C, E and F. The power spectrum of the total energy fluctuations obtained for each of these states is presented in Fig. II.13.a – c. We can observe that this analysis confirms the assumption that state C is the closest to the critical point of the 2D L-J fluid, as even all three cases are characterised by a similar maximum frequency, state C has the largest spectrum fluctuations for a larger frequency range (Fig. II.13.a).



(a)



(b)



(c)

Fig. II.13. The power spectrum of the total energy fluctuations for the states near the critical point (a – state C; b – state E; c – state F)

*CALCULATION RESULTS FOR NEAR-CRITICAL POINT THERMODYNAMIC
PROPERTIES OF 2D L-J FLUID*

The calculation of C_V , κ_T and κ_S used the same assumptions as the cluster analysis:

- the same time step as in the cluster analysis, that is: $\Delta t^* = 0.001$, equal to 0.30216 fs, taking the parameters of Argon: $\epsilon = 1.67 \cdot 10^{-21}$ J and $\sigma = 3.4 \cdot 10^{-10}$ m, and its mass $m = 6.6 \cdot 10^{-26}$ kg/molecule;
- the same cut-off radius for the L-J potential, $r_c = 2.5$.

A steady-state analysis was carried out for the near-critical state (state C) and the supercritical state (state A). The MD simulation was started with a NVT ensemble simulation for 5,000,000 time steps (about 150 ps, using the same Ar parameters), then it was continued with a NVE ensemble simulation, for another 10 million steps (about 300 ps).

For the system of 529 molecules, we carried out a steady - state analysis for cases A and C. We started the molecular dynamics simulation in the NVT ensemble and after about 150 ps (that is about 5 million steps), then changed to the NVE ensemble for another approximately 300 ps. Moreover, to obtain the average function $\langle \Theta \rangle_m = \langle \Theta \rangle_c$, a NVT ensemble simulation was carried out for the entire time interval (10 million steps) of the NVE ensemble simulation.

The results are presented in Table II.4.

Table II.4. Non-dimensional values of thermodynamic properties of a L-J fluid

State	C_V^* [-]	κ_{Tm}^* [-]	κ_{Sm}^* [-]
A	1.82039	8.930796	1.998998
C	4.89306	8.086022	29.37179

As predicted by the thermodynamic macroscopic theory (Jany, 1998), the adiabatic compressibility has a large increase at the critical point. However, the isometric heat capacity has increased only about three times, while the isothermal compressibility has only less than 100 % increase. The conclusion of our study was that, although state C has the theoretical characteristics of the critical point in what it concerns the cluster growth, only one thermodynamic property showed a discontinuity of value, while the other two had only small variations. Thus, it may be referred as a “near- critical state”.

STUDY OF THE INFLUENCE OF THE SYSTEM SIZE

This study was continued in the framework of the research grant CNCSIS AT (2001 - 2002). The influence of the 2D system size on the cluster growth process for the 2D L-J fluid was studied, by comparing the L-J fluid system of $N_1 = 529$ molecules with an L-J fluid system of $N_2 = 1024$ molecules.

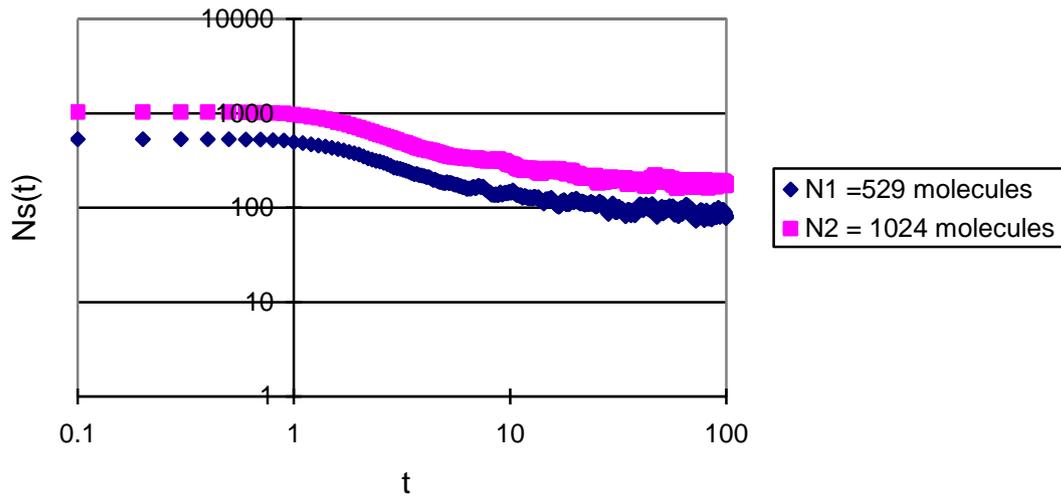


Fig.II.14. Comparison of the average number of clusters vs. time for the two systems, for state A ($T^*=0.672$, $\rho^*=0.338$, $r^*_{cl} = 1.4$)

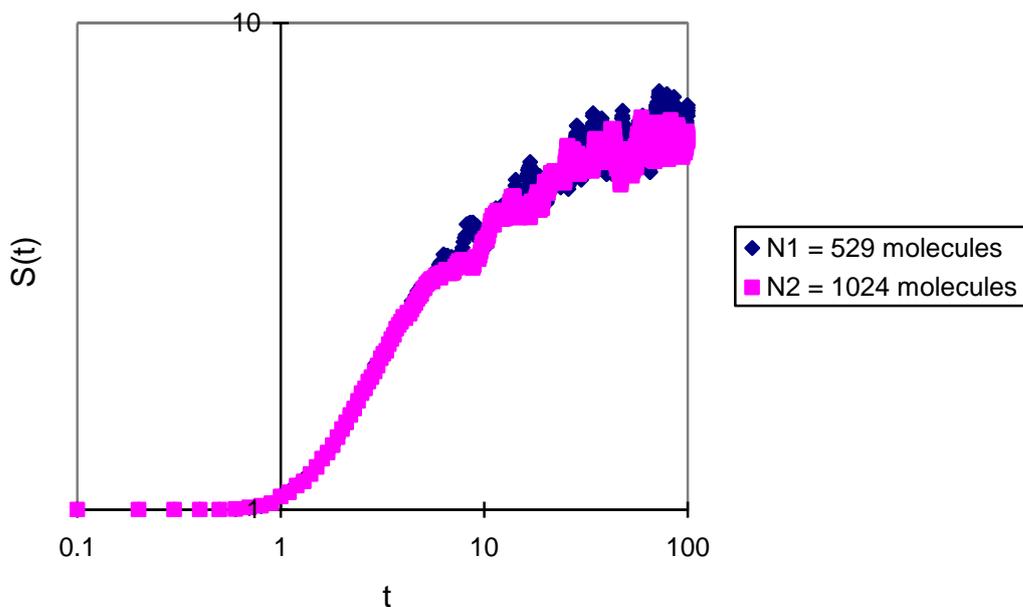


Fig. II.15. Comparison of the average number of particles in a cluster vs. time for the two systems, for the state A ($T^*=0.672$, $\rho^*=0.338$), using $r^*_{cl} = 1.4$

The comparison of the cluster growth characteristic functions, $S(t)$ and $N(t)$, for these two systems are presented in Fig. II.14 and Fig. II.15. We observe that, even the number of clusters increases with time proportionally with the system size, the average number of particles in a cluster is similar for both systems.

STUDY OF THE EFFECT OF THE INITIAL POSITIONS ON THE CLUSTER GROWTH

Another study carried out for the 2D L-J fluid system envisaged the effect of the initial positions of the particles on the cluster growth process (Stoian, Holotescu & Pop, 2001). This analysis showed that, although for the uniform initial configuration the number of clusters is equal with the number of particles in the system, while in the case of random initial configuration, the system contains initially less clusters, the process of cluster growth becomes obvious after the same period of time and tends to stabilise close to the end of the calculation period, irrespective of the initial configuration. An example is given in Fig.II.16, for the same thermodynamic state A (the supercritical region) of the 2D L-J fluid phase diagram.

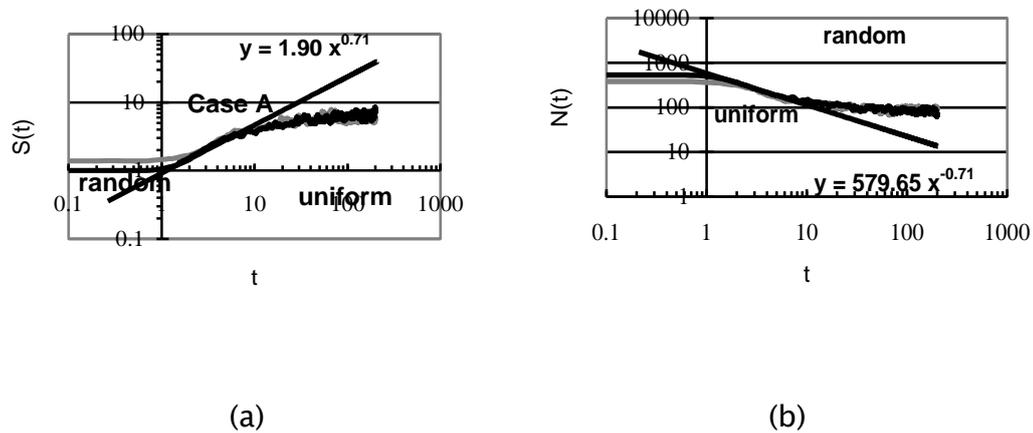


Fig.II.16. Comparison of the cluster growth process, starting from a uniform grid (dark line) and a random grid (light line), for the thermodynamic state A ($T^*=0.672$, $\rho^*=0.338$), using $r_{cl}^*=1.4$

II.1.3.2. 3D SYSTEM ANALYSES

The study for 3D molecular systems was also carried out in the framework of the research grant CNCSIS AT (2001 - 2002). In what it follows, there will be presented the main results, some of which were presented in several national conferences (Stoian, Holotescu & Maekawa, 2002). As in the case of the 2D system, the phase diagram of the Lennard - Jones fluid in 3D is depending on the truncation radius of the intermolecular potential. Several critical point data, as given by reference literature, are presented in Table II.5.

Table II.5. Data regarding the critical point of a Lennard - Jones fluid (3D system)

r_c^*	T_c^*	ρ_c^*	Reference
2,5	1,119	0,409	Powles, 1984 (shifted potential)
2,5	$1,085 \pm 0,005$	$0,317 \pm 0,006$	Smit, 1992 (reduced and shifted potential)
-	$1,36 \pm 0,03$	$0,36 \pm 0,03$	Ree, 1980
-	$1,321 \pm 0,004$	$0,306 \pm 0,001$	Kofke, 1992
-	$1,316 \pm 0,006$	$0,304 \pm 0,006$	Smit, 1992
-	1,350	0,349	Nicolas, 1979

L-J FLUID SYSTEM STRUCTURE AND THERMODYNAMIC PROPERTIES

The analysis regarding the behaviour of the near-critical thermodynamic states for the 3D L-J fluid envisaged the thermodynamic properties of interest. The 3D system consisted of 512 molecules interacting via the L-J intermolecular potential. The same assumption regarding the truncation of the potential at $r_c^* = 2.5$ was used as well as the value for the average cluster radius, $r_{cl}^* = 1.4$, and the

time step, $\Delta t^* = 0.001$. The software program used was adapted for the 3D system simulation. Several thermodynamic states, with parameters close to the critical points listed in the reference literature, were analysed. Here are presented results concerning three states, whose characteristics are given in Table II.6. These were chosen such that to characterise a subcritical state (state A), a near-critical state (state B) and a supercritical state (state C).

The MD simulations were conducted similarly, using the NVT and NVE ensemble simulations. The calculation of thermodynamic properties was accomplished using the following scheme: 1,000,000 time steps of NVT ensemble simulation to equilibrate the system, followed by 2,000,000 time steps of NVE ensemble simulation to calculate the properties of interest.

Table II.6

state	A	B	C
$T^* [-]$	1.2	1.23	1.25
$\rho^* [-]$	0,284	0,284	0,284

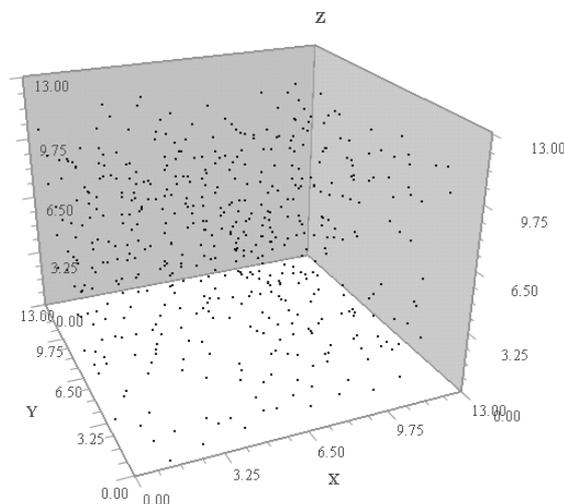


Fig. II.17. a. Snapshot of the 3D L-J system of $N=512$ molecules at state C, after 10,000 time steps

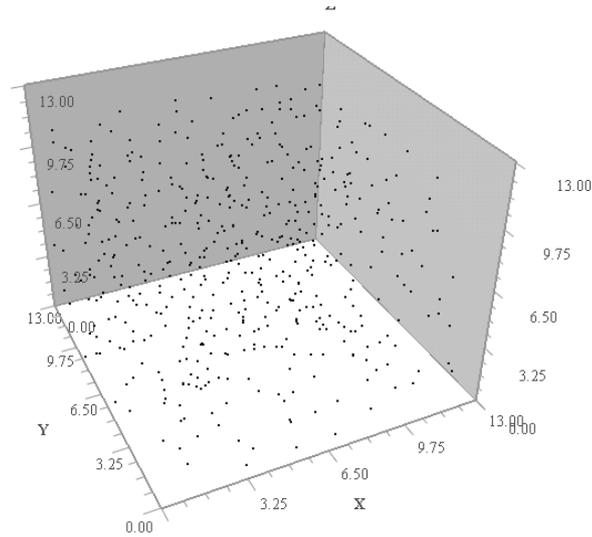


Fig. II.17.b. Snapshot of the 3D L-J system of $N=512$ molecules at state C, after 50,000 time steps

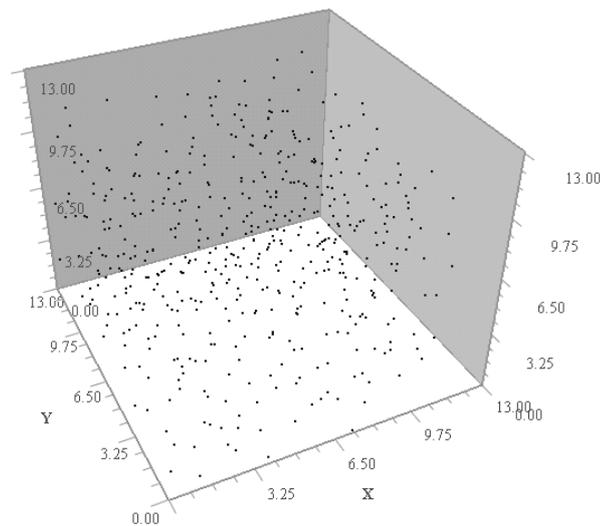


Fig. II.17.c. Snapshot of the 3D L-J system of $N=512$ molecules at state C, after 100,000 time steps

The cluster - cluster aggregation process of the L-J fluid is presented at three instant moments in Fig. II.17.a -c, for a supercritical state (state C, Table II.6). As expected, short clusters can be observed, as the system evolves in time. The pair correlation function, $g(r^*)$, is presented in Fig. II.18, for a near critical state (state B, Table II.6).

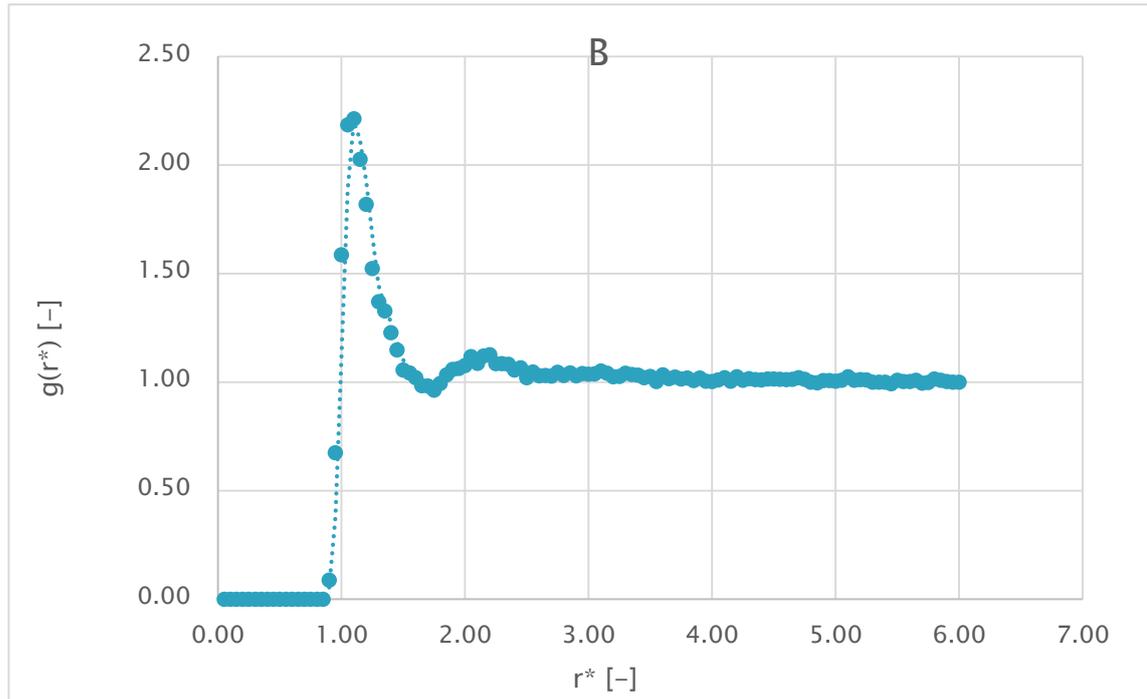


Fig.II.18. The pair correlation function for state B (Table II.6)

Based on the NVE ensemble simulations, the isothermal compressibility, κ_T , and adiabatic compressibility, κ_S , were calculated using the equations (II.19) and (II.21). The results obtained for two thermal properties of interest are presented in Table II.7, in non-dimensional values. We observe the same type of behaviour as in the case of the 2D L-J analysed system: the isothermal compressibility shows an increase from the sub-critical to the supercritical state, while the adiabatic compressibility is almost constant.

Table II.7

State	κ_{Tm}^* [-]	κ_{Sm}^* [-]
A	9,7165	1,7251
B	11,1263	1,6330
C	16,0634	1,8196

EFFECT OF SYSTEM SIZE ON THE THERMODYNAMIC PROPERTIES OF A NEAR-CRITICAL LENNARD - JONES 12-6 FLUID

One of the objectives of the CNCSIS AT/2001 - 2002 grant, was to apply the molecular simulation to certain refrigerants and calculate their bulk thermodynamic properties, in the phase transition region, using the MD simulation. We used the 12-6 L-J intermolecular potential to model the interactions in 3D molecular systems consisting of $N_1 = 512$ molecules and $N_2 = 1000$ molecules and several thermodynamic properties of these systems were calculated. The analyzed thermodynamic states belonged to the two-phase region and the near critical region. The employed simulation scheme was the same combination of NVT - NVE ensemble simulations. In this series of simulations, we used the NVT ensemble to equilibrate the system for a duration of 1,000,000 steps, followed by a NVE ensemble simulation, with a duration of 1,000,000 steps. The time step is $\Delta t^* = 4 \cdot 10^{-3}$. The potential was truncated and the cut-off radius was $r_c^* = 2.5$. To calculate the intermolecular forces, the Verlet list of neighbouring molecules was used, with the maximum radius of interaction $r_f^* = 3.5$. For each 3D system, the isometric heat capacity, isothermal compressibility and adiabatic compressibility for 22 thermodynamic states were calculated. A selection of these results (for the same thermodynamic states A, B and C from Table II.6) is presented in Table II.8 (Stoian, Holotescu & Maekawa, 2002).

Table II.8

state	$N_1 = 512$			$N_2 = 1000$		
	A	B	C	A	B	C
$T^* [-]$	1,21	1,23	1,25	1,21	1,23	1,25
$\rho^* [-]$	0,284	0,284	0,284	0,284	0,284	0,284
$P^* [-]$	0.1718	0.1814	0.1788	0.1563	0.1761	0.1754
$E^*_{total} [-]$	-0.095	-0.045	-0.064	-0.080	-0.082	-0.056
$C^*_V [-]$	2.3642	2.1807	2.2781	2.3975	2.2550	2.3822
$\kappa^*_{Sm} [-]$	1.6145	1.6331	1.8196	1.5265	1.6058	1.5730
$\kappa^*_{Tm} [-]$	8.558	11.002	15.774	5.914	10.396	6.915

We can observe that the results for the isometric heat capacity and adiabatic compressibility were not influenced by the system size, while the isothermal compressibility values show differences. The latter behaviour is similar to that observed in the previous analysis, being an indication that the states are near the critical point.

If the L-J potential parameters for R134a refrigerant, $\epsilon = 383.461 \cdot 10^{-23}$ J/molecule and $\sigma = 0.5067$ nm, are used to calculate the dimensional values of temperature and density for state B, for example, we obtain the following values: $T = 341.62$ K and $p = 5.346 \cdot 10^6$ Pa. If compared to the experimental data for the critical point of R134a, that is $T = 374.179$ K and $p = 4.056 \cdot 10^6$ Pa (Huber and McLinden, 1992), we observe that the state is corresponding to the liquid region of the R134a phase diagram. Thus, the approximation of the intermolecular interactions in the R134a (CH_2FCF_3) through only the repulsion – attraction (or dispersion) interactions that L-J potential include, is not enough. More recent developments in the field showed that, by combining the L-J potential with point charges for the coulombic interactions, can describe accurately the bulk and interfacial properties of R134a (Penguin et al, 2009; Do, Wheatley & Hirst, 2010).

II.1.4. Conclusions

The MD simulation was used to analyse the phase transition near the critical point and the subcritical region, by studying the cluster formation and the effect of the system size and its initial conditions.

A calculation method for the thermodynamic properties: K_s , K_t and C_v , for canonical and microcanonical 2D and 3D molecular systems, was set up and verified for several systems in the critical region of the liquid–vapour phase transition curve and for the liquid region. The results were compared to the macroscopic theory of critical fluids.

Reference publication no.1: Stoian, F.D., Morimoto, H., Sato, T., and Maekawa, T. (2002), MICROSCALE THERMOPHYSICAL ENGINEERING (from 2006 renamed as: NANOSC MICROSC THERM). Vol 6(1), pp. 15–30, ISSN: 1556–7265, DOI:10.1080/108939502753428211 (ISI Article, FI 2015: 2.39)

II.2. Effects of the external electric fields on the liquid – vapour phase transition

This topic is a continuation of the studies in the field of my PhD, that can be separated into a study of the vapour/gas bubble behaviour under micro-g conditions and an applied electric field, followed by a study of the effects of external electric fields at nanoscale, by molecular simulation. The former envisaged the study of the existing models for gas/vapour bubble behaviour and was carried out in 1998, in the framework of a collaboration with the Heat and Mass Transfer Laboratory, University of Pisa (Italy), while the latter was started in 1999, during my post-doctoral stage at Toyo University (Japan), and continued in the framework of the same CNCSIS AT/research grant (2001–2002) and the CNR–NATO Senior fellowship (August – October 2004).

II.2.1. Introduction

As well-known, boiling heat transfer is generally associated with the highest heat transfer coefficients and well suited for high heat flux density cooling applications. When the vapour bubble detachment and the motion of the superheated vapour layer formed on the heated surface are obstructed by the reduction of the gravitational field and, consequently, of the gravitational force, the liquid – vapour phase separation and the heat removal from the surface are affected (Warrier, Dhir, & Chao, 2015). Therefore, the thermal management applications using boiling heat transfer and designed to work in micro-gravity conditions need to achieve the control of the boiling process. The subject is currently under investigation both experimentally and theoretically (Zhang, Mudawar, & Hasan, 2009; Konishi & Mudawar, 2015; Ellis & Kurwitz, 2016; Colin et al, 2016). Also, it is included in experimental programs carried out on board the International Space Station (Raj, Kim, & McQuillen, 2012; Toth, 2012; Warrier, Dhir & Gao, 2015).

A way to overcome the absence of gravity is to use an external field to replace the gravitational one. In the case of boiling, the forces of the external field act both in the bulk and at the vapour – liquid interface (Stoian, 2000; Vekas et al, 2000). The choice of the external field is determined by the working fluid that is

envisaged for a cooling application, such that the thermo–physical properties, the fluid flow and the heat transfer can be manipulated by the applied external field.

The effects of electric fields on boiling heat transfer are characterized by several parameters, which, based on the involved phenomena, can be divided into three main groups:

- physical quantities that influence the phase transition, like: heat of vaporization, saturation pressure and temperature, vapor density, surface tension at the interface between the vapor bubble and the surrounding liquid, microstructural characteristics of the heating surface, the work of adhesion between the heating surface and the liquid etc;
- forces acting on the growing vapor bubble: gravitational forces, resistance forces due to molecular and eddy viscosity, forces due to the presence of pressure gradients about the bubble and along the wall, virtual body forces, forces causing circulation within the bubble;
- forces exerted by the applied field: electrostrictive, polarizing and electroconductive forces (if it is the case of boiling of solutions – both diffusion – electric and thermoelectric effects arise).

Thus, the system of equations that governs the electrohydrodynamic (EHD) heat transfer in the continuum medium hypothesis, in the presence of the gravitational force field, is given below (Jones, 1978; Melcher, 1981):

$$\operatorname{div} \vec{u} = 0 \quad (\text{II.30})$$

$$\rho \left[\frac{\partial \vec{u}}{\partial t} + (\vec{u} \operatorname{grad}) \vec{u} \right] = -\operatorname{grad} p + f_E + \rho \vec{g} - \mu \nabla^2 \vec{u} \quad (\text{II.31})$$

$$\frac{\partial T}{\partial t} + \vec{u} \operatorname{grad} T = a \nabla^2 T + \frac{\sigma_e E^2}{c_p \rho} \quad (\text{II.32})$$

$$\vec{j} = \sigma_e \vec{E} + \rho_F \vec{u} + \frac{\partial (\varepsilon_0 \varepsilon \vec{E})}{\partial t} \quad (\text{II.33})$$

$$\operatorname{div} \vec{j} = 0 \quad (\text{II.34})$$

$$\operatorname{div} (\varepsilon_0 \varepsilon \vec{E}) = \rho_F \quad (\text{II.35})$$

The electric surface force, f_E in Eq. (II.31), is most commonly expressed in the form given by Helmholtz (Melcher, 1981):

$$\vec{f}_E = \rho_F \vec{E} - \frac{1}{2} \varepsilon_0 E^2 \text{grad } \varepsilon + \frac{1}{2} \text{grad} \left[\varepsilon_0 E^2 \rho \left(\frac{\partial \varepsilon}{\partial \rho} \right)_T \right] \quad (\text{II.36})$$

where terms in this equation are the force components:

- electrophoretic force, $\rho_F \vec{E}$, that is caused by the electric field acting on the free charges in the fluid,
- dielectrophoretic force, $-\frac{1}{2} \varepsilon_0 E^2 \text{grad } \varepsilon$, that is caused by polarization of the fluid and particles in the fluid and it manifests any time the electric field must pass through two containing media of different permittivities, like are the vapor – liquid interfaces.
- electrostrictive force, $\frac{1}{2} \text{grad} \left[\varepsilon_0 E^2 \rho \left(\frac{\partial \varepsilon}{\partial \rho} \right)_T \right]$, is a distortive force associated with fluid compression and shear, being present in high–pressure gradient flows, compressible flows and flows with a non–uniform applied electric field.

The experimental researches on the use of external electric fields to control boiling in micro–gravity conditions were started about two decades ago, by several research groups in US and Europe (Shaw and Stahl, 1996; Di Marco and Grassi, 1997; Herman et al, 2002). The working fluids tested in such experiments were refrigerants FC–72 (C6F14) and PF–5052 (C5F11NO), of very high dielectric strength –that is, dielectric liquids (Di Marco, 2006; Herman & Iacona, 2011). During these micro–g experiments, the effect of the applied electric field on the boiling heat transfer and the effect on the bubble dynamics in the nucleate boiling regime were subjects of investigation. The experimental and numerical results have shown that lowering gravity increases the heat transfer – mainly the critical heat flux (Di Marco and Grassi, 1997a, b), while increasing gravity decreases the heat transfer (Merte, 1990). Also, based on experiments and simulations of boiling heat transfer in microgravity conditions, Pachosa and Chung (1990, 1993) showed that the values of electric field and thus, of the electric forces required, are much less than in the ground based experiments.

The behavior of the vapor bubbles was analyzed from two aspects: the electrostrictive (electroconvection) effects and the interfacial effects of the applied electric field (Di Marco and Grassi, 1995; Karayannis and Allen, 1995; Zaghdoudi et al. – 1995, 1996; Daba and Stoian, 1997; Karayannis et al., 1998, etc).

The molecular simulation, by MD or Monte Carlo, of the effects of an external electric field on a thermodynamic system, ranging from a group of tens, hundreds of atoms/ molecules (Gao, Oh & Zeng, 1999; Vegiri, 2002; Vegiri, 2004; Zangi and Mark, 2004) up to tens of thousands of molecules, recently (Samin, Tsori & Holm, 2013), developed as a research oriented mainly on the structural effects determined by the applied field. Also, the effects of an external electric field on the bulk and interfacial properties (thermodynamic and mechanical surface tension) were analysed. For instance, a theoretical study of a weakly dipolar fluid confined in a certain geometry was presented in (Warshavsky & Zeng, 2003), using the density-functional approach. The results showed that the applied electric field increased the critical temperature of the system, enlarged the vapour-liquid coexistence curve and decreased the surface tension, its reduction being dependent on the electric field orientation. Recently, in (Futera & English, 2016), was analysed the effect of an external electric field on the structural behaviour of TiO₂/water interfaces, using non-equilibrium MD techniques.

In the followings, the results obtained from the macroscopic and micro-/nanoscale analyses on the electric field effects on the liquid - vapour phase transition are presented.

II.2.2. A study of the models for bubble behaviour in electric field

Let us observe that, in both film boiling and nucleate boiling regimes, the heat transfer rates are enhanced (though the enhancement is depending on the geometry of the surface - like wire/rod or flat plate type). Also, the electric field effects become prevailing when a threshold value of the electric field strength is reached, a result observed during on-earth experiments (Di Marco & Grassi, 2002; Di Marco, 2006). Macroscopically, when the bubble dynamics in electric field is analysed, this is explained by the force balance at the gas - liquid interface (Di Marco et al, 2013; Herman & Iacona, 2004; Daba & Stoian, 1997).

Following the study that I carried out in 1998 at University of Pisa (Italy), on the boiling of dielectric liquids on terrestrial and in microgravity conditions, a set of criteria to characterize the models for the bubble behavior in electric field was proposed (Stoian, 1999). It resulted from the analyses of the existing analytical and numerical models to date, each taking into consideration all or part

of these criteria, which are presented schematically in Fig. II.19 and described as follows.

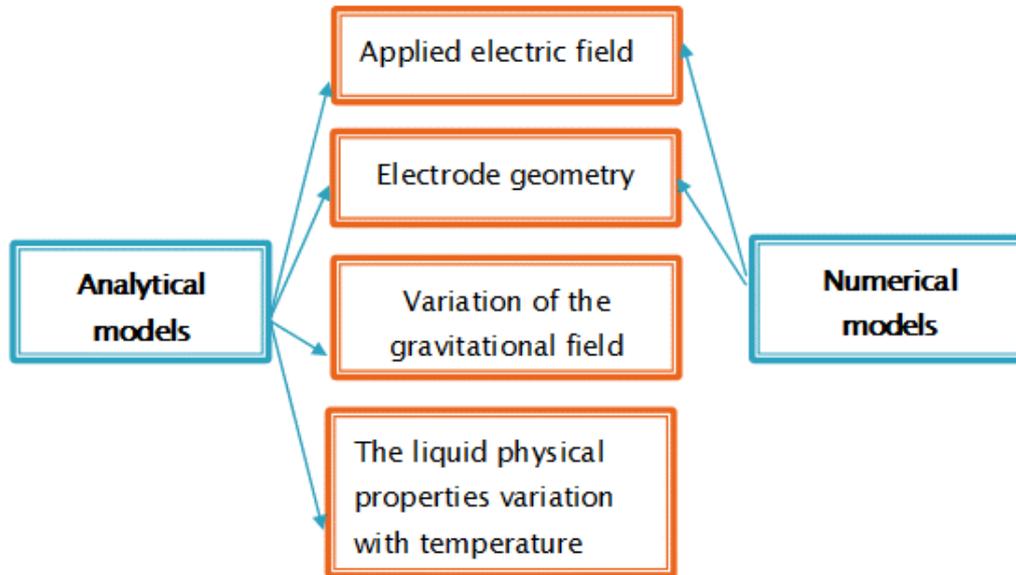


Fig.II.19. Factors that are influencing the bubble behaviour in electric field (Stoian,1999)

The *type of applied voltage* (DC or AC) is a first criterion that differentiate between models. It can be further detailed by the type of polarity in the case of DC field, while in the case of AC fields by frequency.

The *electrode geometry* has the role of ensuring an electric field distribution within the working fluid such that the electrical forces enhance the hydrodynamic processes of forced and/or natural convection. The EHD boiling enhancement is determined by the electric field non-uniformity near the heat transfer surface. The EHD convection is induced due to the forced removal of bubbles from the heat transfer surface (Cooper, 1992). Consequently, the cylindrical geometry was proposed for the heat transfer surfaces in the EHD heat exchangers.

Studies on boiling of dielectric liquids, both theoretical and experimental, showed that, irrespective of the field geometry, the bubble volume decreases as the electric field strength increases (Ogata & Yabe, 1993; Stoian et al, 1997). Thus, comparison of several experimental results showed that the electrode geometry has no significant effect on the decrease of the bubble volume, either the case of vapor or that of gas bubbles is concerned (Kweon et al; 1996, Stoian, 1997).

Although the electrode gap itself was proved to influence the performance of boiling heat transfer, experiments performed for a single gas bubble, for plane geometry electrode and uniform temperature field, showed that the reduction of bubble volume and the bubble elongation are ranging in the same domain, for the same electric field strength obtained with different electrode gaps (Stoian, 1997).

The *effect of gravity* expressed in the value of gravity, g , and of the residence time, t_{res} , was first proposed by Shaw and Stahl (Shaw and Stahl, 1996), who introduced a comparison between the characteristic times of the EHD phenomena: residence time in the electric field (t_{res}), characteristic bubble generation time (t_b) and electric charge relaxation time (t_c). Their conclusion is that electric field effects should be important if $t_c \ll t_{res}$, regardless of whether $t_c \gg t_b$ or $t_c \ll t_b$, that is, so long as the residence time is sufficient for electric effects to occur. Reduced gravity experiments (which greatly increase t_{res}) with liquids that have $t_c \gg t_b$ or $t_c \ll t_b$ should thus delineate whether t_b or t_{res} is more important in determining the effects of electric field.

The most important *physical properties* in this process are *the electrical permittivity*, ϵ , and *electrical conductivity* of the liquid, σ_e , which are both temperature dependent. To decide whether the liquid behaves like an insulating fluid or not, the characteristic frequency of the occurring phenomena, f , must be compared with $1/t_c$ (Jones, 1978). For the case of nucleate boiling this frequency is the bubble departure frequency, f_b . If $1/t_c < f$, then the electric field can affect the bubbles motion by forcing them down, to move on the heat transfer surface (Ogata and Yabe, 1993). If the opposite case is occurring, $1/t_c > f$, then the bubbles float to the free surface, the effects being the decrease of the bubbles volume and the increase of the bubble departure frequency.

This field of research developed in the last two decades taking use of experiments, including those on single bubble growth, and developing models that took into account several of the above criteria (Di Marco et al, 2013). Also, an analysis regarding the fluids for use in terrestrial and microgravity electrohydrodynamic experiments, reviewed the most important physical properties of several such fluids, including safety issues (Herman and Iacona, 2011). Still, according to a recent review (Collin et al, 2017), although the role of the electric forces on the bubble growth and boiling phenomenon was clarified using experimental studies, further studies are needed to investigate the influence of fluid properties and that of the electrode geometry, in view of optimising the electric field configuration with the target of reducing the applied voltage.

The experience that I gained in this field was later used for the analyses of the results obtained on the bubble growth and departure in a magnetic nanofluid, in an applied magnetic field, that are to be presented in the Chapter III.

II.2.3. Micro-/Nanoscale effects of an applied external field on the boiling phenomenon

The subject was initiated during the post-doctoral stage and, later on, continued in the framework of the CNSIS AT grant (Theme 11, code 75/2002) and of a research stage at the University of Pisa (Italy), with a NATO-CNR Senior Fellowship (Bando n.217.35.S del 30/04/2003).

The goal of this study – the effects of electric fields at nanoscale on boiling phenomenon, was accomplished by using the equilibrium molecular dynamics simulation to analyse several two-phase thermodynamic states (corresponding to liquid-vapor mixture states at macroscale) for a dielectric fluid and the effects of an applied external electric field on the internal structure and thermodynamic properties, when the effect of the gravitational field is neglected ($\vec{g} = 0$).

II.2.3.1. THE MOLECULAR DYNAMICS SIMULATION PROCEDURE

The Lennard-Jones 12-6 full potential was used, as described by Eq. (II.8). The electrostatic interaction was modelled considering a linear dielectric system. The definition of the electric field was made, as is acting on an individual molecule of the dielectric (Panofski & Phillips, 1955).

THE ELECTROSTATIC INTERACTION WITH AN EXTERNAL ELECTRIC FIELD

Let us consider the method of molecular fields for a particular molecule located at a non-random position in the material (a dielectric placed between the plates of a parallel plate capacitor), as in Figure II.20. The dielectric and the

capacitor are sufficiently large in the directions parallel to the plate so that end effects may be neglected. The electric field acting on this molecule which is itself one of the molecules of the polarized medium can be approximated as follows. Let us draw a sphere of radius a about this particular molecule, intended to represent schematically the boundary between the microscopic and the macroscopic range of phenomena concerning the molecule. This molecule is thus influenced by the fields arising from the following charges:

- the charges on the surfaces of the capacitor plates;
- the surface charge on the dielectric facing the capacitor plates;
- the surface charge on the interior of the spherical boundary of radius a ;
- the charges of the individual molecules, others than the molecule under consideration, contained within the sphere of radius a .

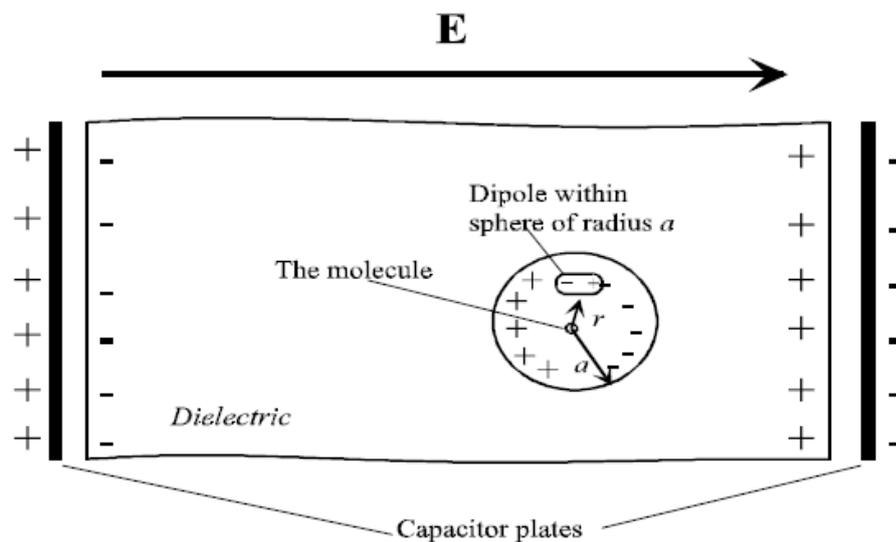


Fig.II.20. The contributions of a dielectric material to the electric field exerted on one of its molecules

The fields due to these sources are, correspondingly:

(a) the charge on the capacitor plates produces a field at the molecule in question equal to:

$$\frac{D}{\varepsilon_0} = E + \frac{P}{\varepsilon_0} \quad (\text{II.37})$$

where D – electric induction, E – electric field strength, P – electric dipole moment and ε_0 – electrical permittivity of free-space.

(b) the polarization charge on the surface of the dielectric facing the capacitor plates, $\sigma_p = P_n$, produces a field on the molecule equal to:

$$E = -\frac{P}{\varepsilon_0} \quad (II.38)$$

(c) the polarization charge from the inside of the sphere, if assumed that the polarization is not affected by the presence of the cavity, produces a field equal to:

$$E_p = \frac{P}{3\varepsilon_0} \quad (II.39)$$

(d) the field due to individual molecules within the sphere, must be obtained by summing over the fields due to the dipoles (of moment p) within the sphere. The field at distance r from a dipole is:

$$E = -\nabla\phi = -\frac{1}{4\pi\varepsilon_0} \left[\frac{p}{r^3} - \frac{3(p \cdot r)r}{r^5} \right] \quad (II.40)$$

Considering that the dielectric is isotropic, the sum over all the dipoles within the sphere gives a null field inside.

The sum of all field contributions gives the total macroscopic field acting on a molecule:

$$E_{eff} = E + \frac{P}{\varepsilon_0} - \frac{P}{\varepsilon_0} + \frac{P}{3\varepsilon_0} = E + \frac{P}{3\varepsilon_0} \quad (II.41)$$

Thus, a term was added to the L - J intermolecular potential to describe the interactions of a system of L - J fluid molecules with an external electric field oriented upon z axis, as presented in Fig. II.21. The resulting interaction is thus given by:

$$\Phi_{ij} = \Phi_{LJ}(r_{ij}) + \Phi_e(r_{ij}) \quad (II.42)$$

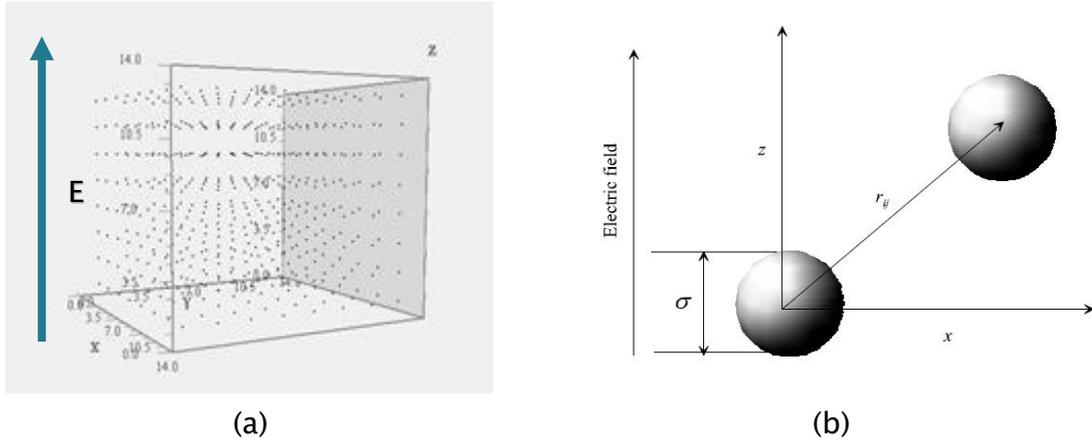


Fig. II.21. Description of Lennard – Jones molecules interaction in electric field: (a) a 3D L – J system; (b) detail for two L – J molecules

The second term in Eq.(II.42) is representing the dipole moment interaction between two molecules/atoms in an external electric field oriented upon z axis is given by:

$$\Phi_e(r_{ij}) = \frac{|P|^2}{4\pi\epsilon_e} \left(\frac{1}{r_{ij}^3} - \frac{3z^2}{r_{ij}^5} \right) \quad (\text{II.43})$$

where ϵ_e is the electrical permittivity and

r_{ij} is the distance between two molecules.

The momentum equation, equipartition theorem and virial equation are then expressed as follows:

$$m \frac{dv_i}{dt} = - \sum_j (\nabla \Phi_{ij}) \quad (\text{II.44})$$

$$NkT = \frac{1}{2} \sum_i m_i v_i^2 \quad (\text{II.45})$$

$$pV = \frac{1}{2} \sum_i m_i v_i^2 + \frac{1}{4} \sum_2 \sum_{j>i} r_{ij} \cdot (-\nabla \Phi_{ij}) \quad (\text{II.46})$$

where p - pressure and v_i - velocity of molecule i .

To simplify the equations of motion and to allow the possibility of scaling the results for a class of systems described by the same model, the formulae are nondimensionalised, using the same formulae as in Chapter II.1. Additionally, for the electric dipole moment nondimensionalisation was used the following formula:

$$e^* = \frac{|P|^2}{64\pi\epsilon_e\epsilon\sigma^3} \quad (II.47)$$

The effect of the value of the applied electric field was studied for three thermodynamic states, as shown in Table II.9:

- states A and B in the two - phase region (vapor-liquid mixture) and
- state C in the liquid region (superheated liquid) of the Lennard - Jones phase diagram (Smit, 1992).

To study the system size-effect, two systems were used for the case A: one with $N_1 = 256$ molecules and the other with $N_2 = 512$ molecules.

Table II.9

Case	A		B	C
	$N_1=256$	$N_2=512$	$N=512$	$N=512$
Temperature $T^*[-]$	0.96	0.96	1.2	0.96
Number density $\rho^*[-]$	0.45	0.45	0.45	0.75

THE NUMERICAL PROCEDURE

The simulation used the same type of simulation scheme as previously: the canonical ensemble simulation, to relax the system and to perform the structure analysis, and the microcanonical ensemble, to determine the static thermodynamic properties of the systems considered for study.

Several methods can be used to consider the effect of electrostatic interaction given by the applied electric field (Norlund, 2003; Rapaport, 1995):

- cut-off methods: simple cut-off at $r=r_c$ or shifted potential and shifted force (to avoid discontinuities at $r=r_c$);
- reaction field: a cut-off method + a continuum medium with constant dielectric constant ϵ ;

- Lattice summation: conditionally convergent in periodic boundary conditions for which a convergence factor is introduced;
- Ewald summation: the electrostatic interaction is divided into faster convergent terms, so periodic boundary conditions can be applied.

As outlined by Bergdorf et al. (Bergdorf, 2003), the cut-off truncation reduces the computational costs and the effect of artificial periodicity in simulations. However, the straight truncation represents a very severe approximation. A countermeasure to this approximation is to increase the system size and, consequently, the cut-off radius (Allen & Tildesley, 1991). Smooth truncation methods may be applied, to reduce heating caused by the application of cut-off, but retain several undesirable effects of abrupt truncation. Nevertheless, some dependencies of simulated observables on the cut-off distance or system size have also been observed for the lattice sum and reaction field methods.

In the followings, several results obtained for 2D and 3D systems of molecules are presented (Stoian & Holotescu, 2002; Stoian, 2004). In these studies, the simple cut-off method was employed but with a larger cut-off radius than the usual one used for Lennard - Jones interactions, usually of $r_c = 2.5\sigma$. Thus, for the system of $N_1 = 216$ molecules, the cut-off radius was taken $r_{c1} = 3.0\sigma$, while for the system of $N_2 = 512$ molecules was taken $r_{c2} = 4.0\sigma$ (for cases A and B), and for case C (liquid) was taken $r_{c2} = 3.5\sigma$. The influence of the tail of the potential was estimated analytically, by assuming the radial distribution function $g(r)$ equal to 1, for a distance $r > r_c$. The Verlet half - step "leap-frog" scheme was used to integrate the equations of motion, same as in the previous analyses (Section II.1).

The simulation box was a simple cubic lattice, with particles distributed uniformly at the initial moment (6x6x6 and 8x8x8, respectively) and periodic boundary conditions. The non-dimensional time step was chosen $\Delta t^* = 1 \cdot 10^{-3}$. In order to obtain uncorrelated data, the simulation was started in the canonical ensemble until the system reached a steady state - the dependence of the potential energy versus time being monitored for 200 000 time steps. These data were used to calculate the radial distribution function, $g(r)$, for the structure analysis. Then, the simulation ensemble was changed to microcanonical for the next 50 000 time steps. During this second part of the simulation, the necessary data for calculating thermo-physical properties (the kinetic and potential

energies, as well as the system temperature and pressure, at every time step) were produced and saved.

INFLUENCE OF THE EXTERNAL ELECTRIC FIELD ON THE STRUCTURE OF THE ANALYSED SYSTEMS

The radial distribution function, $g(r)$, was determined using Eq. (II.15). The results obtained for cases A to C, for several values of the field parameter, e^* , are presented in Figures II.22 – II.25. An overall comparison of the radial distribution functions for cases A(N₁) to C shows that:

- as the electric field parameter e^* is increased, the first maximum peak increases while the first minimum peak decreases, indicating an enhancement of the first neighbour shell in both two-phase states as well as the liquid state;
- the positions of the first maximum peaks are not changed with the field;

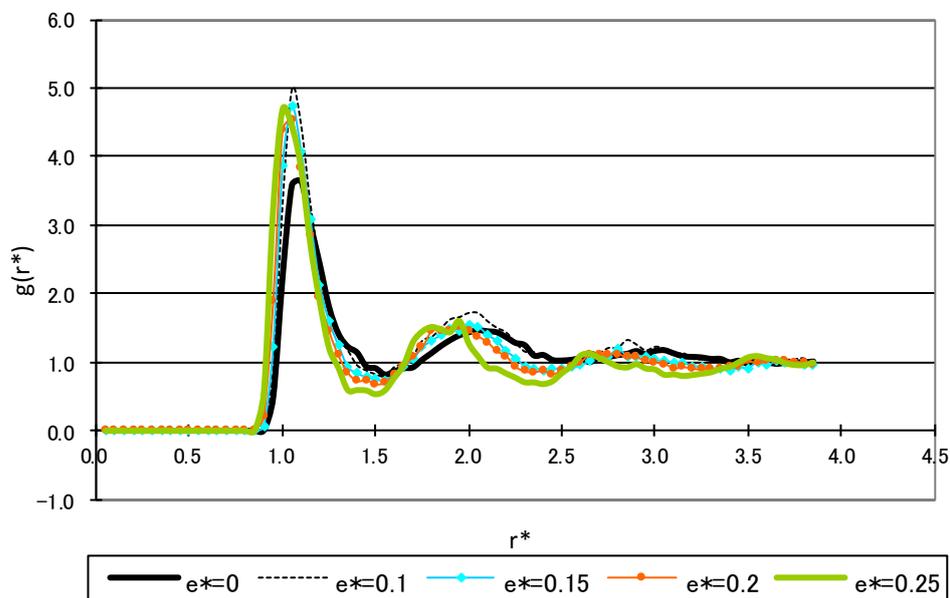


Fig. II.22. The radial distribution function for case A (N₁)

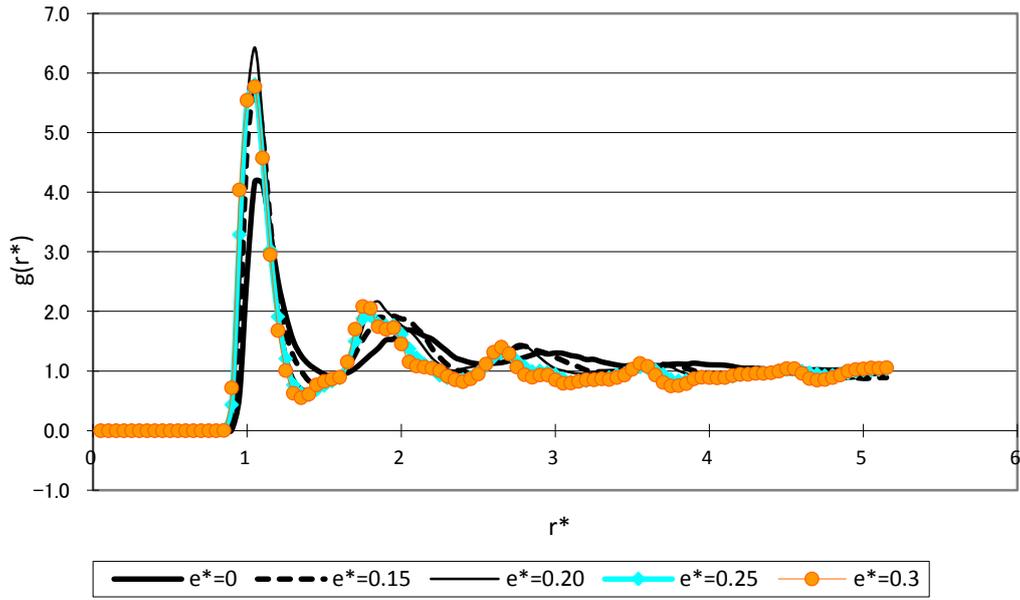


Fig. II.23. The radial distribution function for case A (N₂)

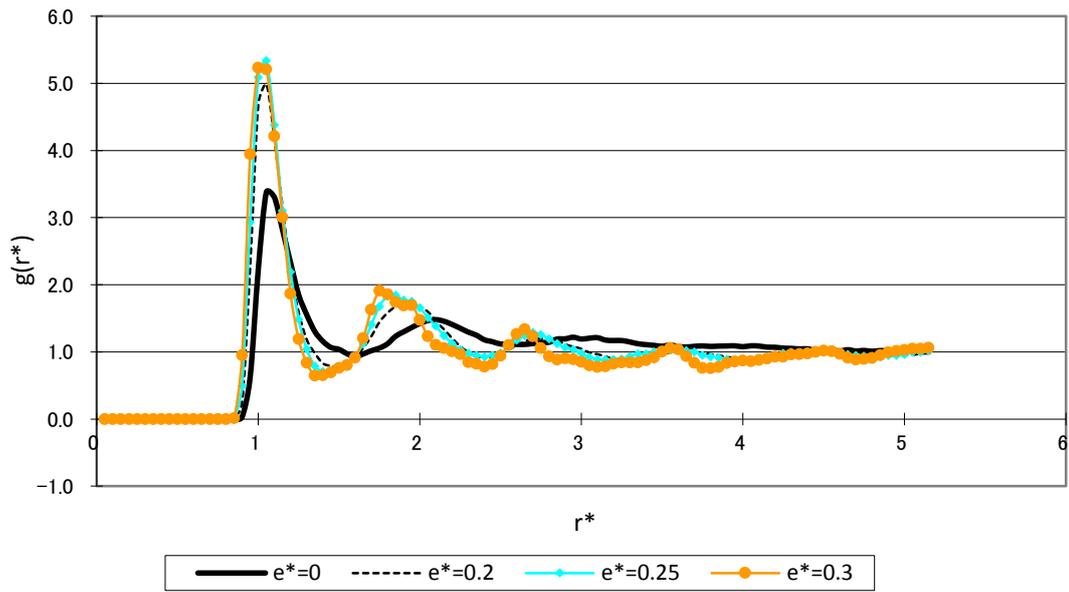


Fig. II.24. The radial distribution function for case B

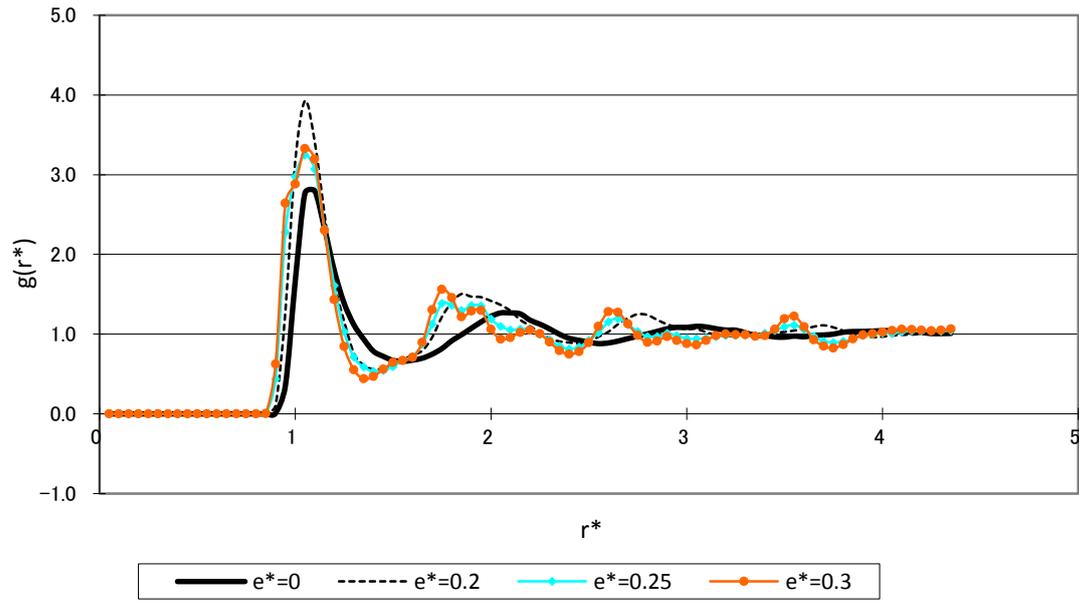


Fig. II.25. The radial distribution function for case C

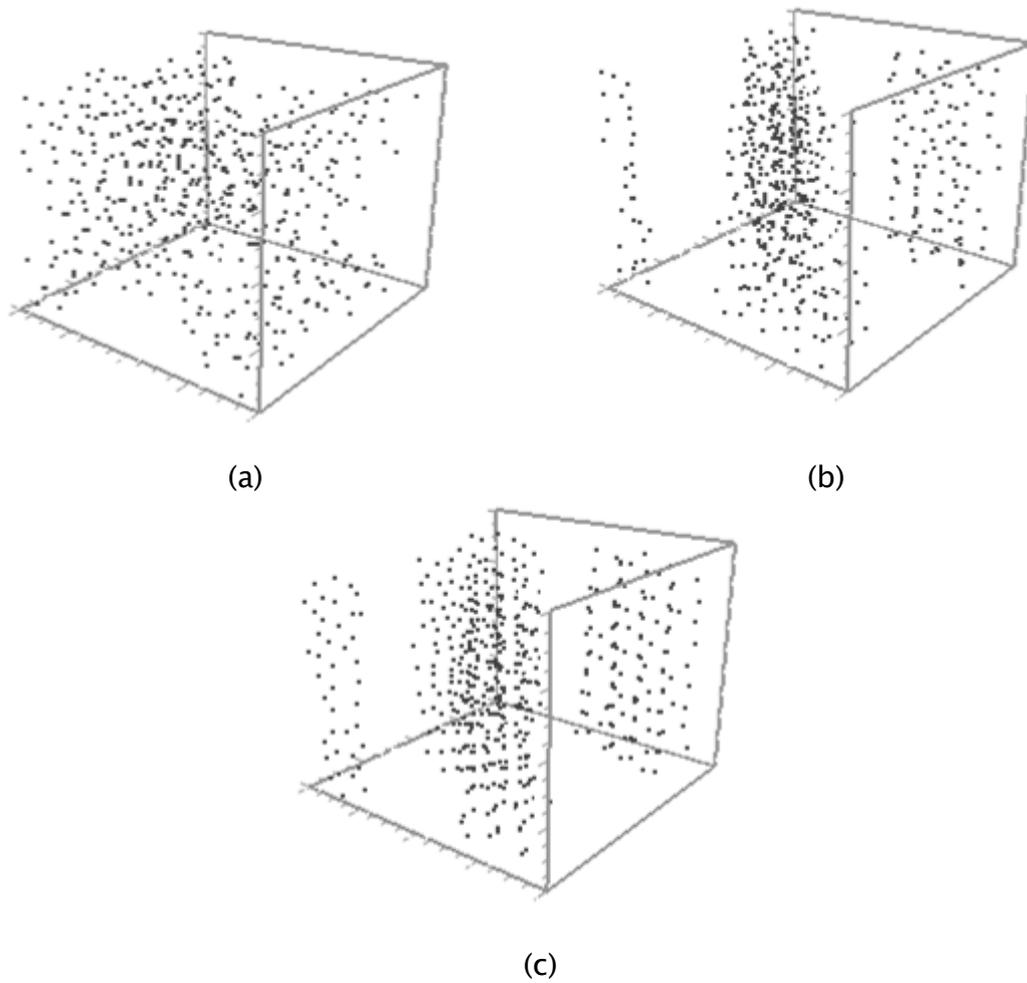


Fig. II.26. Snapshots of the A(N₂), for e* = 0 (a), e* = 0.15 (b), e* = 0.2

- the second neighbour shell is significantly affected by the application of the external field (this is the range which is very important for the dielectric properties of the fluid); the height of the second maximum peak increases and the second minimum peak decreases (less than the second maximum peak), thus the distance between fluid molecules increases. This can also be observed, for instance, from Fig. II.26 (a–c), where the instantaneous positions of molecules at the end of the simulation, for three values of the electric field parameter, are plotted.
- the first minimum and second maximum peaks change positions with the increase of the field parameter;
- from a certain value of the electric field parameter, the second maximum peak starts to exhibit a “secondary” peak – indicating that a different correlation of positions may appear – for both two–phase and liquid systems.

Also, comparing the results shown in Fig.II.22 and Fig.II.23, a size effect can be observed in the values of $g(r^*)$ – the first peak maximum is about 20% higher for case A(N₂), when the system size is almost double compared to case A(N₁). Also, the second peak maximum of case A(N₁) is within the limits of calculation error, for values of the field parameter below $e^*=0.2$. The difference might be due also to the employed method of simple cut–off.

The effect of the temperature increase on the radial distribution function can be observed by comparing Figures II.23 and II.24. The first and second maximum peaks are smaller for a higher temperature of the two–phase system, at the same value of the electric field parameter, indicating that the increase of distance between molecules due to the applied field may be counteracted by the increase of temperature.

THE INFLUENCE OF THE EXTERNAL ELECTRIC FIELD ON THE VALUES OF THERMODYNAMIC PROPERTIES

The non–dimensional values of the calculated thermodynamic properties for each analysed state, for several values of the electric field parameter e^* , were determined. The most relevant results are presented as follows.

The comparison of the isometric heat capacity data shows that C_V increases until a certain value of e^* is reached, and after that is slightly fluctuating. For

states $A(N_1)$, $A(N_2)$ and B, the threshold value of e^* is influenced by the system size. Figure II.27 exemplifies the results obtained for state $A(N_2)$.

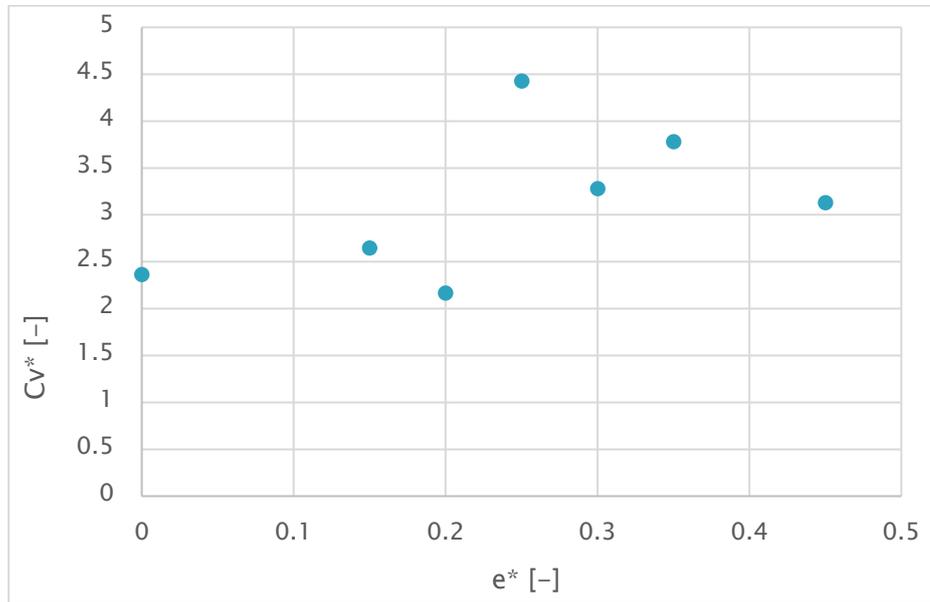


Fig. II.27. The isometric heat capacity behaviour for state $A(N_2)$, as a function of the electric field parameter

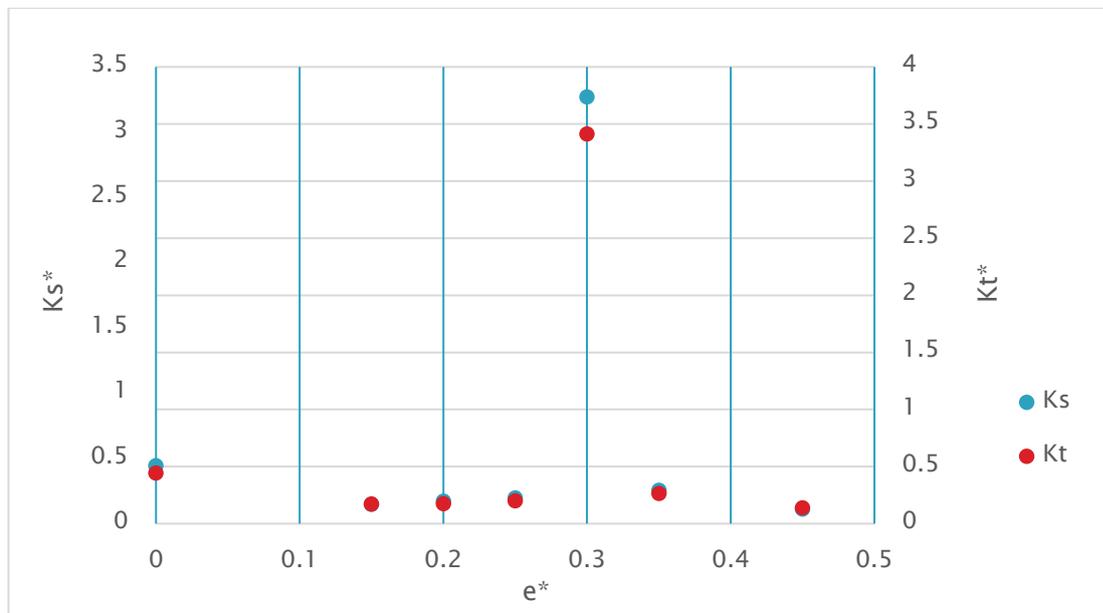


Fig.II.28. Electric field effect on the adiabatic and isothermal compressibility, for case $A(N_2)$

The most important effect of the electric field on the adiabatic and isothermal compressibilities is evidenced by the case $A(N_2)$, when an abrupt increase occurs at $e^*=0.3$. To further investigate this behaviour, additional calculations were carried out, the results being shown in figure II.28, where the jump of the adiabatic compressibility indicates that a “phase-transition” – like phenomenon may occur at $e^*=0.3$.

The results showed also that the internal energy of the system (kinetic plus potential) has a multipeak dependence with the electric field parameter, that is reflected also by the values of the isometric heat capacity. As the temperature of the system is increased the multipeak trend flattens. Moreover, as the system is kept at constant volume, the pressure increases with the applied field, the increase being more pronounced with the increase of the temperature of the system ($T_B > T_A$).

II.2.4. Conclusions

Several conclusions have been drawn from the molecular dynamics simulation study, with respect to the structure behaviour and the thermodynamic properties of a L–J system in the presence of an external electric field:

- the structural analysis carried out using the radial distribution function showed that, at constant temperature, the distance between molecules in the second shell increases and the molecules are grouping in long clusters oriented along the field lines, as the applied electric field is increased;
- the pressure of the system is increased by the applied field, as the volume is kept constant;
- the isometric heat capacity exhibits a multipeak behaviour, like that of the internal energy of the system, as the applied field is increased;
- for the liquid state considered for study (having the same temperature as one of the two-phase states), it was shown that the compressibilities (adiabatic and isothermal) as well as the thermal expansion coefficient decrease when increasing the electric field parameter.

In what it concerns the extrapolation of the MD results to the macroscopic scale results, let us observe that:

- similar to the observations made in boiling experiments in microgravity (Di Marco & Grassi, 1997a), the increase of the system temperature may partially counteract the effects of the applied electric field;

- the effect of the electric field on the cluster formation and orientation can be related to the experimental results regarding the “shift” of the boiling curve and change in the critical heat flux, in microgravity conditions (Di Marco & Grassi, 1997a).

As pointed out previously, some of the drawbacks to the date are the simulation length and the number of molecules that can be taken into consideration. However, if these are comparable to the representative volumes and timescales of a problem, MD can be a powerful tool to analyse the system dynamics and characteristic thermodynamic properties.

III. Magnetic nanofluids: researches on the heat transfer control by magnetic fields and their thermal properties

III.1. A fundamental study regarding the control of nucleate boiling in a magnetic nanofluid, by an applied DC magnetic field

This research was carried out in the framework of the national research project no. 37/2001 (UPT contract no. 11657/2001–2004), entitled “Studies and researches regarding the behaviour of magnetisable fluids with complex micro and nano structure in terrestrial and microgravity conditions”, in the AEROSPATIAL Program (National Research, Development and Innovation Plan – PNCDI I). The Project coordinator was Dr. Doina BICA†, from the Romanian Academy – Timisoara Branch, Laboratory of Magnetic Liquids. This project represented also the start of a long-term research collaboration, that is lasting till present days, with the multidisciplinary research team working at the Research Center for Engineering of Systems with Complex Fluids (RCESCF), Laboratory of Magnetometry and Laboratory of Rheology.

III.1.1. Introduction

Magnetic nanofluids have found various applications, in the decades since were first introduced, in the fields of like engineering, medicine and plants biology (Odenbach, 2002; Vekas et al, 2009; Kitanovski et al, 2014), some of the long-term internationally recognised being developed by the interdisciplinary team from the Politehnica University Timisoara and the Romanian Academy – Timisoara Division (Vekas et al, 1987; Anton, De Sabata & Vekas, 1990; Bica 1995; Bica et al, 2004; Vekas et al, 2009).

First called ferrofluids, then magnetic liquids (magnetic fluids) and, since S.U.S. Choi (Choi & Eastman, 1995) introduced the term “nanofluid”, as “magnetic nanofluids”(symbolised further in this thesis as MNFs), they are a category of

smart materials consisting of magnetic nanoparticles (such as Fe_3O_4 , CoFe_2O_4 , Co, Fe), coated by surfactants to avoid agglomeration, and stable dispersed in a carrier liquid, giving rise to either non-polar magnetic fluids (hydrocarbon based) or polar magnetic fluids (water, alcohols, diesters based). The type of application is imposing the type of nanoparticles, surfactant and carrier liquid, beside the preparation method and procedure.

The first reports on studies of boiling of MNFs, in the presence of an applied magnetic field, for potential use in engineering applications date back to 1993 (Takahashi et al, 1993; Gogosov & Simonovskii, 1993), followed by others during that decade (Kamiyama & Ishimoto, 1995; Nakastuka et al, 1999), for a potential use of magnetic nanofluids as working fluid for heat pipes. The subject continues to be a topic of research both in experimental and theoretical studies on flow boiling (Ishimoto, 2007), critical heat flux (Lee & Jeong, 2011), nucleate boiling mechanism (Yanovskii et al, 2014).

The first international report on the potential use of boiling magnetic nanofluids for cooling applications in microgravity was presented by Vékás et al. (Vékás et al., 2000). The authors presented experimental results regarding the effects of magnetic field on the bubble dynamics, in an experiment that simulated the bubble nucleation, which showed that depending on the value, uniformity and orientation of the magnetic field versus the gravitational field, the bubble frequency can be controlled in both directions (either increased or diminished) or even stopped. One of the main goals of the national project contract no. 11657/2001–2004/UPT envisaged to develop this work and gain knowledge on the bubble growth and departure. This was carried out in a multidisciplinary team, in collaboration with colleagues from my department, as well as undergraduate, graduate and PhD students.

My scientific contribution to the study of control of nucleate boiling in a magnetic nanofluid concerned the design and run of the experiments, as well as the results analyses (Stoian et al, 2003; Stoian et al, 2004; Stoian et al, 2010). Additionally, I contributed with the magnetic properties measurements for the MNF samples used in this project, the results being presented in (Bica et al, 2004).

III.1.2. Theoretical background

The equation of ferro-hydrostatics that governs the evolution of a bubble of volume V_b in a MNF of magnetisation $M(H)$, in the absence of the fluid motion, is given by (Berkovsky et al, 1993):

$$\nabla p = \rho \vec{g} + \mu_0 M \nabla H = \rho \left(\vec{g} + \frac{\mu_0}{\rho} M \nabla H \right) \quad (\text{III.1})$$

where p is the sum of the hydrostatic pressure and the fluid-magnetic pressure, ρ - density, \vec{g} - gravity vector, μ_0 - permeability constant, M - magnetisation of MNF, H - magnetic field intensity.

By applying the magnetic field, a supplementary term arises in the Laplace equation for curved liquid - vapor/gas interfaces, due to the jump of the normal components of the electromagnetic part of the overall stress tensor at the interface. Thus, the magnetic excess pressure, expressed by:

$$\Delta p_M = \mu_0 (\vec{M} \vec{n})^2 / 2 \quad (\text{III.2})$$

influences the process of bubble nucleation and departure.

If the bubble is in a non-uniform magnetic field, that is $|\nabla H| \neq 0$, the additional buoyancy force that arises is given by [Berkovski et al, 1993; Vekas et al, 2000, Drenckhan et al, 2003]:

$$\vec{F}_b^M = -\mu_0 V_b M(H) \nabla H \quad (\text{III.3})$$

The value and orientation of this force plays an important role on the parameters that are characterising the bubble growth and departure from a nucleation site: the emission frequency, f_b , the volume of departing bubble, V_b , and its equivalent diameter, D_{ech} . Therefore, if a non-uniform magnetic field is applied to a boiling MNF, in the nucleate boiling regime, this magnetic buoyancy force can trigger important effects on the bubbles dynamics and the boiling regime and, consequently, the heat transferred.

If the inertia and drag forces are neglected, the force balance for the case when the buoyancy and magnetic forces are acting in the same direction, results in (Blums et al, 1997):

$$\vec{F}_b + \vec{F}_b^M = \vec{F}_s \quad (\text{III.4})$$

The bubble departure diameter is then given by:

$$D_{r,m} = f(\beta) \sqrt{\sigma / (\rho_L - \rho_G) g \pm \mu_0 M(H) \nabla H} \quad (\text{III.5})$$

where: $f(\beta)$ is the liquid to surface contact angle,

σ – the surface tension,

ρ_L – the liquid density, ρ_G – the vapor density,

g – gravitational acceleration;

The sign under the square root is depending on the orientation of the buoyancy force with respect to the magnetic force.

Surface tension at the liquid – vapour interface is also affected by the applied magnetic field. Thus, if a sphere of magnetic permeability μ_i immersed in a fluid medium of magnetic permeability μ_e is considered, when a uniform magnetic field of intensity \vec{H} is applied to the fluid, the normal and tangential components of surface tension vary as follows (Raikher & Shliomis, 1994):

$$\sigma - \sigma_0 = \begin{cases} -lH^2 \left(\mu_e + \frac{\mu_e^2}{\mu_i} \right) \\ \frac{lH^2}{2} (\mu_e + \mu_i) \end{cases} \quad (\text{III.6})$$

where l – characteristic length, H – magnetic field intensity.

One can observe that the normal component of the magnetic field reduces the surface tension, while the tangential component of the magnetic field increases the surface tension. In the absence of the gravitational field, the bubble formation, growth and departure is controlled only by capillary and magnetic forces, the latter being preminent, except for the early nucleation stage, when the curvature of the vapor – liquid interface is very small.

Further, a comparison between the characteristic times for the bubble growth in a MNF, in the presence of an applied magnetic field and absence of gravitational field can be made:

- the residence time in magnetic field, τ_{res}^m ,
- the bubble generation period, τ_b ,
- the magnetic relaxation time.

In what it concerns the last characteristic time, let us note that when a magnetic field is applied, two relaxation mechanisms can occur in MNFs (Raikher and Shliomis, 1994):

- the rotational Brownian diffusion, characterised by the relaxation time, $\tau_B = 3V^{-1}\eta/kT$;
- the Néel relaxation, characterised by the relaxation time, $\tau_N = f_0^{-1}(KV/kT)^{-1/2} \exp(KV/kT)$

where V – volume of dispersed nanoparticles, η – viscosity, k – Boltzmann constant, T – absolute temperature, f – body force, K – anisotropy constant.

Because the magnetic nanoparticles dispersed in the carrier liquid have a characteristic size distribution, both Brownian and Neel relaxation can contribute to the value of MNF sample magnetisation, the effective magnetic relaxation time being expressed by:

$$\tau_{eff} = \tau_N \tau_B / (\tau_N + \tau_B) \quad (III.7)$$

Therefore, both the MNFs characteristic properties as well as the experimental conditions (e.g. bubble emission frequency, applied magnetic field) should be chosen considering the correspondence between these characteristic times, in order to obtain the desired effect for the bubble dynamics.

III.1.3. Results regarding the effects of an applied magnetic field on the bubble growth and departure in a magnetic nanofluid

III.1.3.1. THE EXPERIMENTAL BENCH

Using the experience gained during my PhD studies, I contributed to the design of an experimental bench which allowed for a complex study of the phenomena related to effects of uniform or non-uniform DC magnetic fields on the growth and departure of gas bubbles, injected through a hole made in a flat surface, in a magnetic nanofluid. The experiment aimed to mimic the growth and departure of a vapour bubble in magnetic nanofluid, to study the effect of applied magnetic field on the bubble dynamics. Using this experimental bench, the influence of several characteristic parameters was analysed (Stoian et al, 2003):

- magnetic field parameters: field intensity, field gradient and orientation with respect to the gravitational field;

- bubble growth and departure parameters: bubble emission frequency, bubble volume and equivalent diameter;
- magnetic nanofluid magnetic properties.

The experimental bench employed to this study was built and tested at the RCESCF, together with the team involved in the 11657/2001–2004/UPT project. Figure III.1 shows the bench, as built in the laboratory.

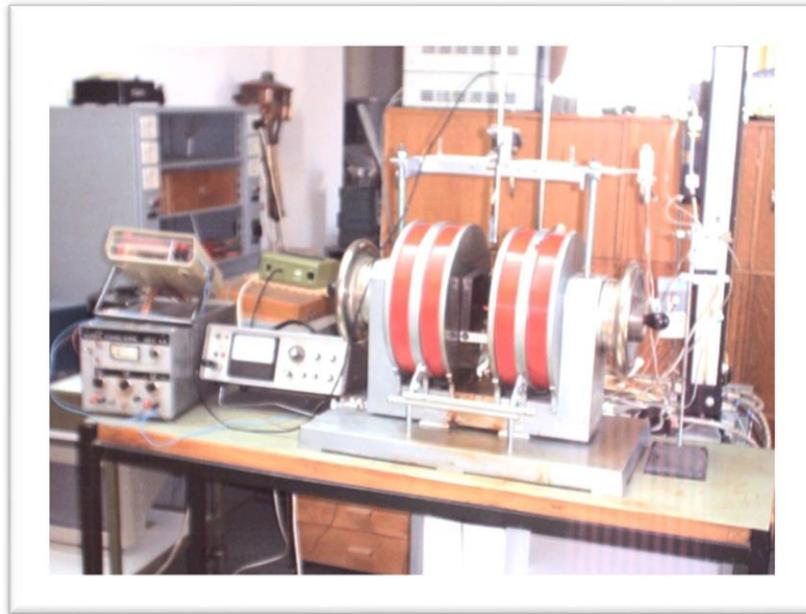


Fig. III.1. A general view of the experimental bench

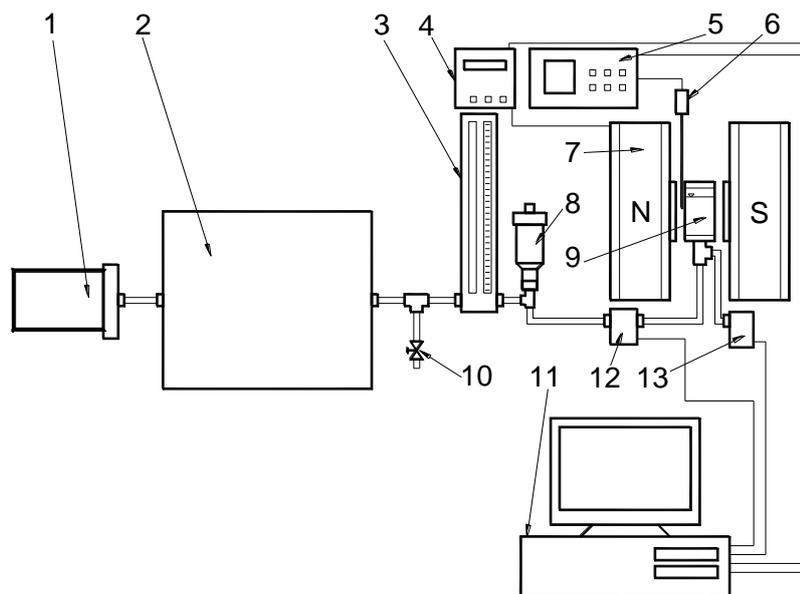


Fig.III.2. Scheme of the experimental bench

Figure III.2 shows the scheme and its main components: 1 – compressor; 2 – gas reservoir; 3 – manometer; 4 – magnets power supply; 5 – gaussmeter; 6 – hall probe; 7 – magnetic poles; 8 – micrometer valve; 9 – experimental cell; 10 – flow rate valve; 11 – PC computer with a National Instruments data acquisition system; 12 – flow rate transducer; 13 – pressure transducer.

The main part of the experimental bench is the experimental apparatus that consists of a cylindrical cell (9), in which a known volume of MNF is introduced. The cell has glass walls of external diameter 38 mm and thickness 3 mm. In the center of its Plexiglas bottom cap a gas injection hole was drilled. The cell is open to air, so at the free surface of MNF acts the atmospheric pressure. Three cells, each with a different central hole diameter, were tested: 0.3 mm, 0.7 mm and 1.5 mm.

The experimental bench was equipped with a compressor (1), to supply the air from a gas reservoir (2). The pressure inside the air circuit was measured using a manometer (3). The air flow rate is finely changed using the micrometer valve (8) and is measured using an air flow transducer (12), with the measurement range of (0 – 20) ml/min, accuracy $\pm 1\%$. The relative pressure close to the entrance in the hole was measured using a pressure transducer (13), with a measurement range of 0 – 12.5 mm H₂O and accuracy of $\pm 1\%$.

The experimental apparatus was placed between the poles of an electromagnet (7), powered by the power supply (4). The magnetic field induction was measured using a Bell Gaussmeter (5) and a Hall probe (6). Two types of magnetic poles were used:

- straight plane, which generated a uniform magnetic field (as in Fig. III.2),
- inclined plane, which generated a non-uniform magnetic field, of gradient parallel to the gravitational field, $\nabla B \parallel \vec{g}$, as shown in Fig. III.3. When these poles were reversed, the field gradient became anti-parallel to gravity, $\nabla B \parallel -\vec{g}$.

Prior to the experiments, the magnetic flux density was measured along the symmetry axis of the air gap, with a 10 mm step. From the magnetic field induction measurements for the inclined plane poles, $\nabla B \parallel \vec{g}$, a field gradient of 79.6 kA/m² was obtained at the hole site, positioned 50 mm above the bottom of the poles (as in Fig. III.3).

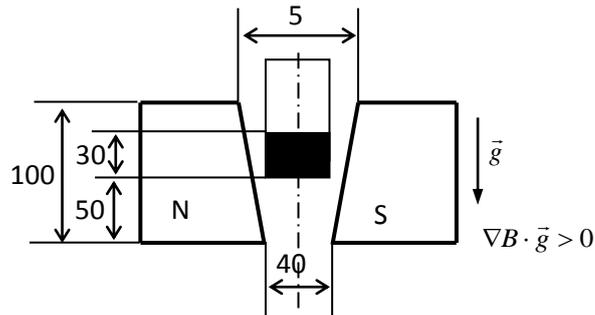


Fig. III.3. The positioning of experimental cell

The measurements were carried out using a National Instruments DAQ system (Lab 1200/AI, 8 channels) and a PC computer (11), to collect the data for processing and analysis. The LabView software was used to produce the data acquisition, storage and the primary analysis program.

III.1.3.2. THE CHARACTERISTIC PROPERTIES OF THE MAGNETIC NANOFUID SAMPLES

The MNF samples were prepared at the Laboratory of Magnetic Fluids, RA-TB. The magnetic measurements were carried out at the Laboratory of Magnetometry of RCESCF -UPT. The samples were prepared using the chemical co-precipitation and monolayer sterical stabilization procedures (Bica, 1995; Bica et al, 2004; Vékás et al, 2004). Two MNF samples were selected for these experiments: one of medium (LM_1) and respectively, one of high saturation magnetization (LM_2). Sample LM_2 has a saturation magnetisation approximately 40% higher than that of sample LM_1. Their composition, density and saturation magnetization are given in Table III.1.

Table III.1. Characteristic properties of MNF samples

Sample	LM_1	LM_2
Carrier liquid	Transformer oil	Transformer oil
Magnetic nanoparticles	Fe ₃ O ₄	Fe ₃ O ₄
Density, ρ [kg/m ³]	1230	1470
Saturation magnetization, M_s [kA/m]	29.46	47.77

III.1.3.3. THE MEASUREMENTS PROCEDURE

A set of parameters were monitored and recorded using the DAQ system and PC computer:

- the relative pressure, measured near the injection hole;
- the injected air flow rate;
- the electromagnet power supply voltage;
- the ambient pressure and temperature, at the beginning of each measurement.

Each series of measurements was run at first without an applied magnetic field, to obtain the reference data for comparison: the bubble emission frequency f_0 and bubble volume V_{b0} .

A number of 1 000 samples/s were recorded for time periods of (2 – 10) s, depending on the periodicity of the relative pressure fluctuations. Each measurement was repeated several times before recording, to avoid any peculiar behaviour to affect the data analyses. Examples of raw data for the relative pressure and gas flow rate plot are presented in Figure III.4.

All measurements were started from the same initial value of the inlet gas flow rate: 5 ml/min, irrespective of magnetic liquid sample.

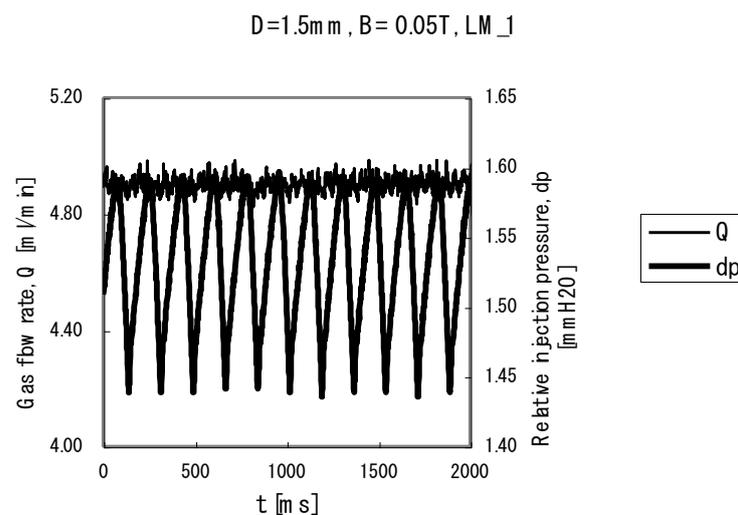


Fig. III.4. Example of a set of recorded data for the relative pressure and the gas flow rate

III.1.3.4. DATA ANALYSIS PROCEDURE

For each series of measurements, the following average parameters were determined:

- average bubble frequency;
- average bubble volume.

To obtain these averages, the recorded data of the relative injection pressure, dp , were used. As observed from Fig. III.4, the variation of dp with time corresponds to the formation and departure of gas bubbles from the hole. By measuring the time between two consecutive peaks, we can obtain the bubble emission period, τ_b , and calculate the bubble frequency from:

$$f = \frac{1}{\tau_b} \quad [s^{-1}] \quad (III.8)$$

By averaging the values of the measured flow rate over the acquisition period, for a certain value and gradient orientation of magnetic field, we get the average gas flow rate. Then, the average bubble volume can be obtained:

$$V_b = \frac{\dot{Q}}{f} \quad [mm^3] \quad (III.9)$$

The results of these average values were further normalised by the corresponding values for of the reference case ($B = 0$; $H = 0$), denoted by: f_0 and V_{b0} .

Assuming an equivalent sphere of volume V_b , an equivalent bubble diameter can be determined:

$$D_{ech} = \sqrt[3]{\frac{6V}{\pi}} \quad (III.10)$$

III.1.3.5. RESULTS AND DISCUSSION

The analysis regarding the influence of the magnetic field parameters on the bubble growth and departure, for each sample of MNF used, showed that:

- a) if the **sample LM_1** is used (Stoian et al, 2004):
- the average bubble emission frequency increases, by increasing the applied magnetic field when $\nabla B \parallel \vec{g}$, for different injection hole diameters, as shown in Figure III.5. The average bubbles frequency is increased up to 20–25 % at the maximum applied field of $B = 0.17$ T, while the average bubble volume decreases with the same ratio. Considering the Eq. (III.7), the average bubble volume behaves opposite to the bubble frequency.
 - if the field gradient is oriented opposite to the gravitational field, that is $\nabla B \parallel -\vec{g}$, the variation of bubbles frequency is different depending on the injection hole diameter, as observed from Fig. III. 6. The bubbles frequency decreases for all injection holes if the applied magnetic field is increased over a certain value, the most effective are the injection holes of $D = 0.3$ and $D = 1.5$ mm.

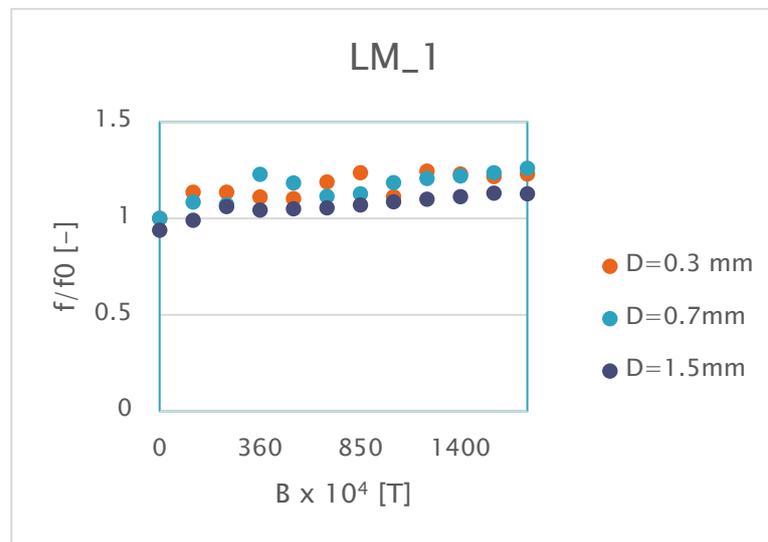


Fig. III.5. The normalised average bubbles frequency versus the applied magnetic field, for sample LM_1 and $\nabla B \parallel \vec{g}$

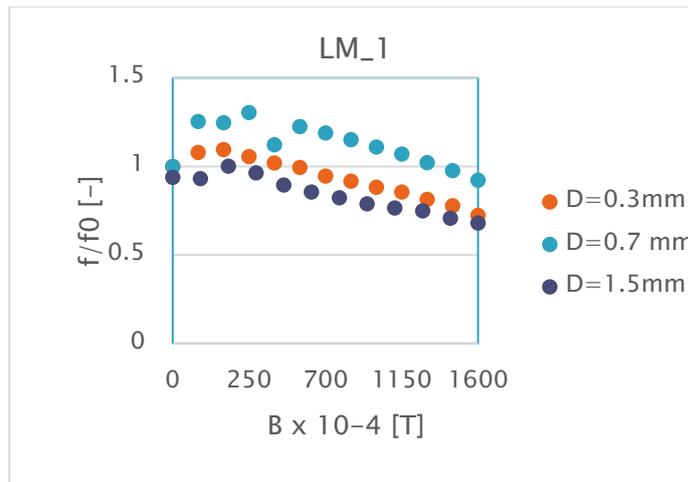


Fig. III.6. The normalised average bubbles frequency versus the applied magnetic field, for sample LM_1 and $\nabla B \parallel \vec{g}$

b) if the **sample LM_2** is used (Pop et al, 2004; Stoian et al, 2004; **Stoian et al, 2010**):

- the behaviour of the average bubble frequency when the applied field has $\nabla B \parallel \vec{g}$, is similar to that exhibited for LM_1 for the largest hole injection diameter ($D = 1.5$ mm) only, while in the case of the other two the dependencies change after a certain applied field is reached, as shown in Fig. III.7.

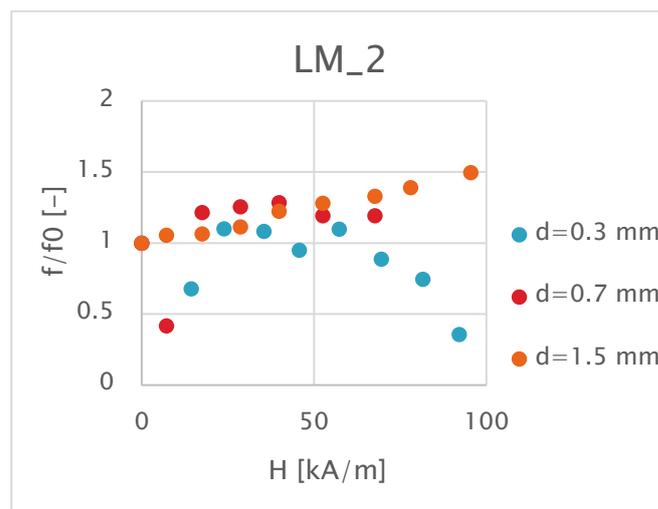


Fig. III.7. The normalised average bubbles frequency versus the applied magnetic field, for sample LM_2 and $\nabla B \parallel \vec{g}$

Thus, as the bubble departure diameter decreases, from a certain value of the applied field, inertia and drag forces should be accounted for. This behaviour is similar to that observed in actual nucleate boiling experiments of a kerosene based magnetic fluid in a uniform field: increasing the fluid boiling temperature and the applied magnetic field, the bubble generation frequency at first increases and then decreases (Kobozev & Simonovskii, 2007).

- a comparison of the influence of the field gradient orientation with respect to the gravitational field, $\nabla B \parallel \vec{g}$ versus $\nabla B \parallel -\vec{g}$, on the absolute value of bubble frequency, f , for the largest injection hole ($d = 1.5$ mm), indicated an interval of variation of f from approximately $(-25\%$ to $+ 55\%) f_0$, taking as reference the bubble generation frequency at zero magnetic field (Fig. III.8). The corresponding values of bubbles equivalent diameter, D_{ech} , calculated using Eq. (III.10), varied from $(-16\%$ to $+ 12\%) f_0$. Results reported in (Bashtovoi et al, 2005), using a magnetic nanofluid of similar composition and saturation magnetisation, injection capillary tubes of 0.8 to 1.8 mm diameter, showed that by applying a much higher field gradient up to about 10 times higher than in this experiment, a corresponding much higher variation of the bubble volume (and consequently, of the bubble frequency) could be obtained.

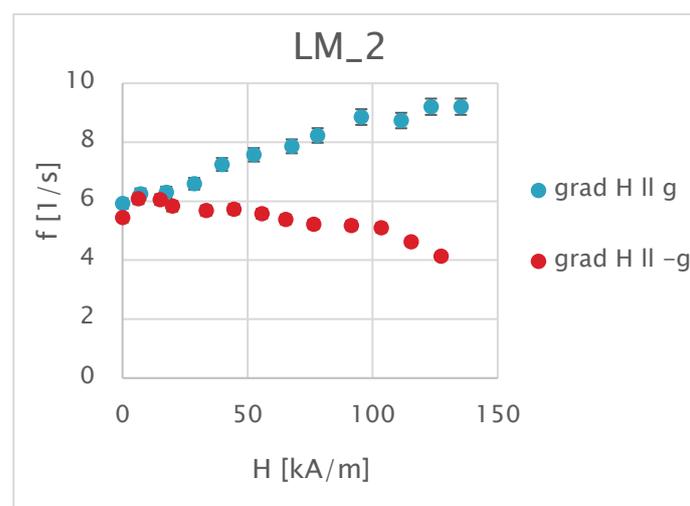


Fig. III.8. Average bubble generation frequency as function of the applied magnetic field intensity, H , and the field gradient orientation vs gravity

Through this experimental study, it was proven that the control of the bubble growth by the applied magnetic field can be achieved in both directions: either increase of the bubble frequency or the opposite.

A comparison of the results obtained with both MNF samples was presented in Fig. III.9 and Fig.III.10, for the injection hole ($d = 1.5$ mm). When the applied magnetic field has a gradient $\nabla B \parallel \vec{g}$, the increase of the average bubble frequency is proportional to that in saturation magnetisation of the MNF samples. If the field gradient changes direction with respect to gravity, the influence of the sample saturation magnetization becomes less important as the magnetic field induction is increased, as shown in Fig. III.10.

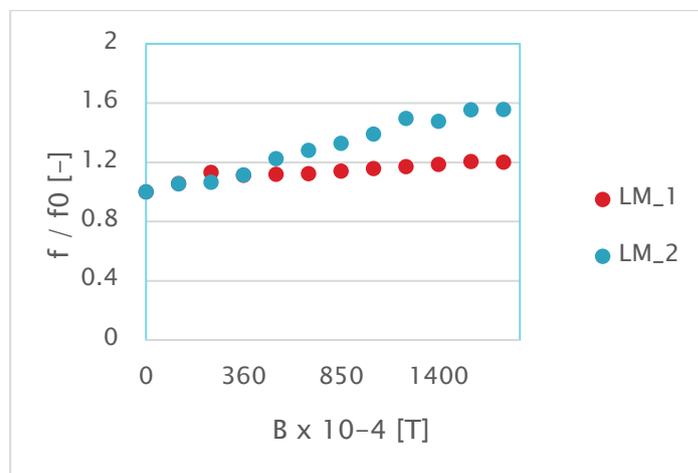


Fig. III.9. Effect of the MNF samples characteristics on the normalised average bubbles frequency, for $\nabla B \parallel \vec{g}$ and $d=1.5$ mm

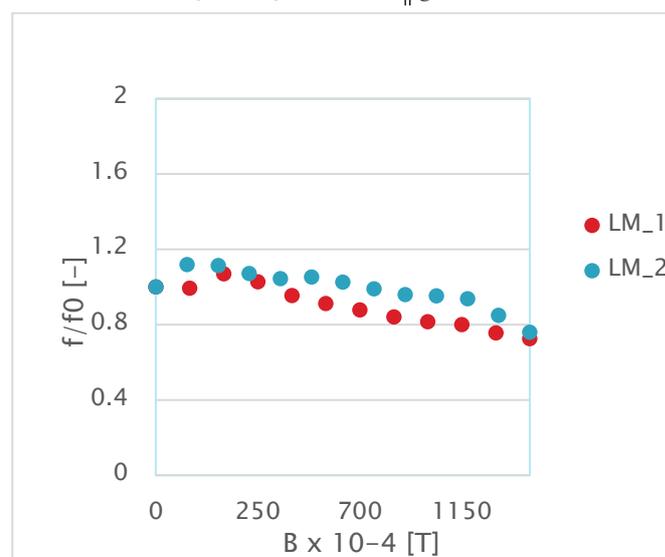


Fig. III.10. Effect of the MNF samples characteristics on the normalised average bubbles frequency, for $\nabla B \parallel -\vec{g}$ and $d = 1.5$ mm

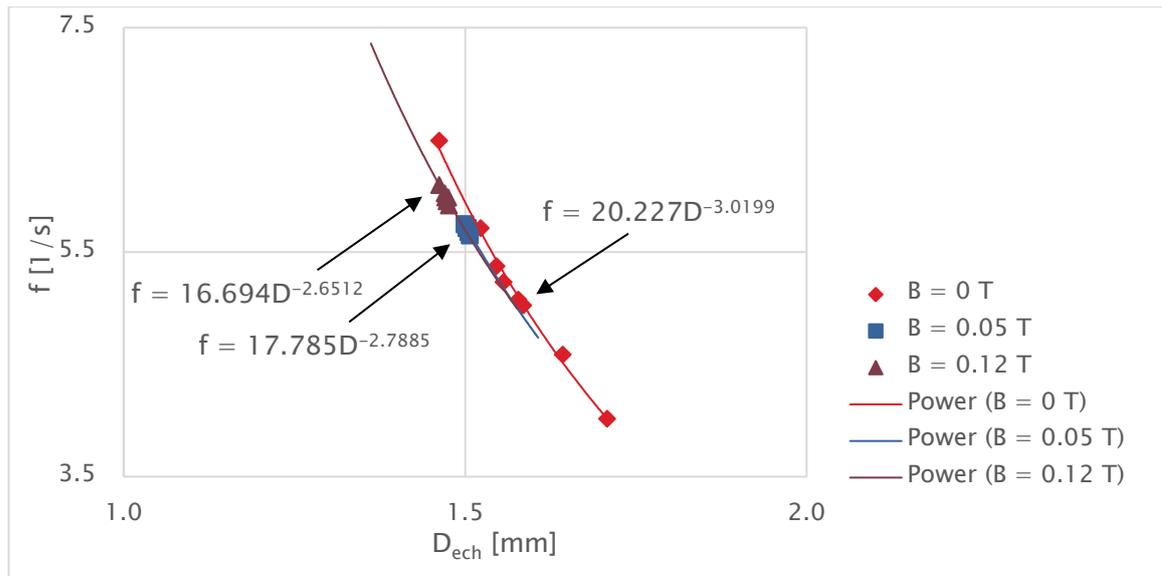


Fig. III.11. Comparison of bubble frequency as a function of bubble equivalent diameter, for different values of the applied magnetic field, for different values of the applied magnetic field, (MNF sample LM_1, $D = 1.5$ mm)

Usually, nucleate boiling correlations are given in the form of dependencies between the vapour bubble emission frequency, f , and its diameter, D (Straub, 1995). Using the experimental data, a dependence between these two parameters was obtained in the form of a power law:

$$f = aD_{ech}^{-n} \quad (III.11)$$

where a and n are constants, as shown in Fig. III.11.

Let us observe that in the absence of the magnetic field ($B = 0$), the dependence has the form $f \propto D^{-3}$ which corresponds to the transition region in the boiling theory, when the bubble dynamics is assumed to depend on buoyancy, drag and surface tension (Dhir, 1998; Kreith, 2011). The expressions for the data sets with $B \neq 0$ have similar constants for the power of D .

III.1.4. Conclusions

This study envisaged the fundamentals of the nucleate boiling of magnetic nanofluids in an applied magnetic field. The main conclusions are:

- the orientation of the magnetic field gradient with respect to gravity determines the increase (for $\nabla B \parallel \vec{g}$) or decrease (for $\nabla B \parallel -\vec{g}$) of the bubble emission frequency;
- the level of these effects is dependent on the magnetic field intensity and the magnetic properties (saturation magnetisation) of the MNF samples, being more prominent for the case of $\nabla B \parallel \vec{g}$.
- the dependence of the bubble frequency with the bubble volume and its equivalent diameter, respectively, are power laws of type $f \propto V_b^{-m}$ and $f \propto D_{eq}^{-n}$, where m and n are dependent on the magnetic field intensity.

Our project developed a pioneer work in this field started in our country. These results proved the potential of using the boiling of magnetic nanofluids and the nucleate boiling regime for microgravity cooling applications but, a validation through an actual microgravity experiment is still a pending research topic.

Reference publication 2: Stoian F.D., Pop G, Bica D, Stoica V, Marinica O, Vekas L. (2003), SPACE TECHNOLOGY AND APPLICATIONS INTERNATIONAL FORUM – STAIF 2003 – AIP Conference Proceedings, ISSN 0094–243X, 2003, Vol.654, pp. 149–156, DOI: 10.1063/1.1541289 (ISI Conference Paper)

Reference publication 3: Stoian F.D., Holotescu, S., Vékás, L. (2010), 12th International Conference on Magnetic Fluids, August 1–5, 2010, Sendai, Japan, Physics Procedia, Vol. 9, pp. 216–220 (ISI Conference Paper).

Reference publication 4: Bica, D., Vékás, L., Avdeev, M.V., Bălăşoiu, M., Marinică, O., Stoian, F.D., Susan–Resiga, D., Török, G., Rosta, L. (2004), Progr. Colloid Polym. Sci., Vol. 125, pp. 1–9, (ISI Conference Paper).

III.2. Natural convection heat transfer in water based magnetic nanofluids

This research was started in the framework of the national project CNCSIS A665/2005 – 2007. It aimed to take advantage of the improved thermal conductivity and heat capacity of magnetic nanofluids, compared to that of the carrier liquids (water, hydrocarbons), of the possibility to control the heat transfer process by applying a magnetic field and envisaged their potential use in the vehicles thermal management systems. The subject was continued in the framework of the project PN II PCCE IDEI 76/2010–2013. This research benefited from the cooperation with the team of the Laboratory of Magnetic Liquids, Romanian Academy – Timisoara Branch, who prepared the MNF samples, as well as the teams of Laboratory of Magnetometry and of Laboratory of Rheology, RCESCF (UPT), where were determined their magnetic, rheological and magneto-rheological properties.

III.2.1. Introduction

Unlike the medium and high magnetisation magnetic nanofluids used in the previous research, and the magnetic fluids that are prepared for their traditional applications in sensors or seals, those intended for cooling systems should have a low volume fraction, to make them competitive with non-magnetic nanofluids that contain solid nanoparticles of higher thermal conductivity and were developed as a new class of thermal fluids during the last two decades (Choi, Eastman, 1995; Das et al, 2007). Moreover, the use of an applied magnetic field offers the possibility to manipulate their thermal conductivity, viscosity, heat capacity and, thus, a control of the heat transferred (Shima et al, 2009; Nkurikiyimfura et al, 2013; Bahiaei, Hangi, 2015).

As mentioned in Ch, III.1, usually MNF is device-oriented designed and prepared, in terms of type of nanoparticles and concentration, surfactant and carrier liquid. The amount of MNF used in established engineering applications, like magnetic seals or magnetic actuators and sensors (Anton et al, 1990; Bashtovoi, Berkovski, 1996; Popa et al, 1997; Borbáth et al, 2011), or in medical applications (Vekas et al, 2009; Laurent et al, 2011), is of several cm³ or less. Moreover, the complex procedures needed to obtain the nanoparticles, their coating and colloidal dispersion in the carrier liquid, limits the amount of MNF produced at once. Therefore, their main use was and still is targeted to

applications where the removal of heat or, inversely, generation of heat, is additional to the main problem to be solved, like drag reduction by magneto-fluidic coatings (Bashtovoi et al, 1991; Mai & Früh;2003) or, the opposite, generation of heat in hyperthermia for cancer treatment (Jordan et al, 1999; Laurent et al, 2011). Also, a special class of magnetic nanofluids are those characterised by the magnetocaloric effect (low Curie Temperature), which can be used for dedicated heat transfer applications like magnetic refrigeration and heat pump cycles (Kitanovski et al, 2014, Chaudhary et al, 2017) or cooling of electrical equipments (Raj, Moskowitz, 1995; Patel et al, 2016).

Our studies regarding the use of magnetic nanofluids in convective heat transfer included the following stages:

- preparation of water based magnetic nanofluids, with Fe_3O_4 nanoparticles coated with a double layer of Dodecylbenzenesulfonic acid (DBS) and transformer oil based magnetic nanofluids, with Fe_3O_4 nanoparticles coated a single layer of oleic acid (OA);
- characterisation of magnetic, rheological and magneto-rheological properties;
- natural convection heat transfer characteristics for a magnetizable nanofluid of low volume fraction, for a certain configuration of cavity that may be suitable to accomplish our target.

Our studies used two types of experimental set-ups, both aimed to evaluate the natural convection heat transfer coefficient for a water based magnetic nanofluid, with respect to that obtained using the carrier liquid. My scientific contribution to this research envisaged the experiment designs, choice of an appropriate MNF sample and data analysis, the main results being reported in (Stoian et al, 2008; Stoian, Holotescu, 2012).

III.2.2. The methodology employed to calculate the heat transfer coefficient

The inverse problem method allows to make the calculation of an unknown quantity using measurable data. We used it to obtain the wall surface temperature and the heat transfer coefficient, as follows.

The heat flux was estimated from the Fourier law, based on the temperature gradient between two thermocouples. For our first experimental set-up shown in Fig.III.12.a (Stoian et al, 2008), we used T_{p1} and T_{p3} , as in the detail from Fig. III.12.b:

$$\dot{Q} = \frac{\lambda_{Al}}{x} (T_{p1} - T_{p3}) \quad [W] \quad (III.12)$$

where λ_p is the hot wall thermal conductivity, x is the distance between the positions of the thermocouples, in the axis of the wall. The wall material is aluminium, with thermal conductivity at room temperature $\lambda_{Al} = 235 \text{ W/m}\cdot\text{K}$, and the distance $x = 10 \text{ mm}$.

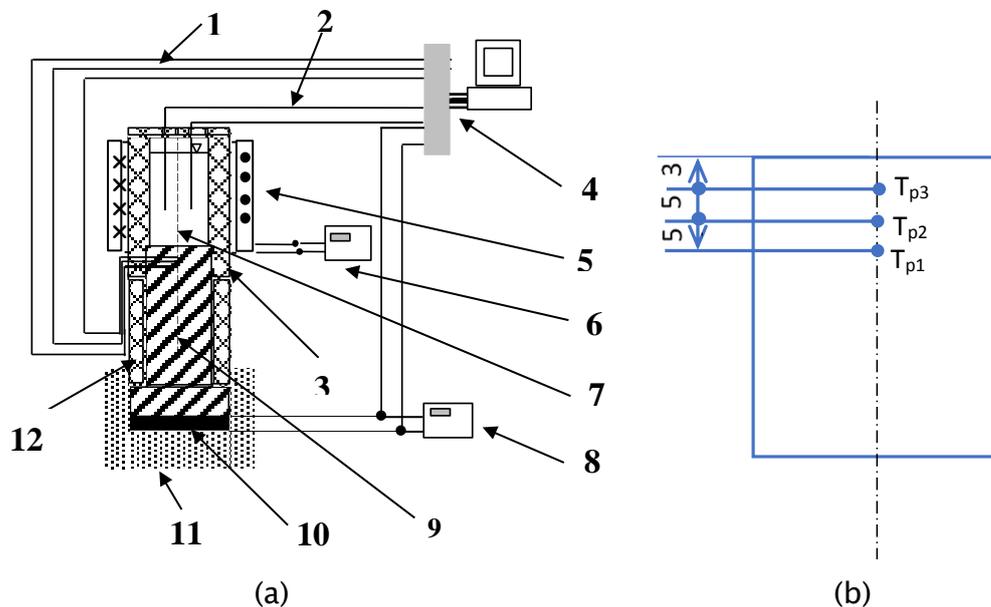


Fig. III.12. The convective heat transfer experiment: a – Experimental set-up; b – Detail of the wall temperature measurement

The wall surface temperature was obtained using the temperature given by the near-surface thermocouple, T_{p3} , and the distance $\delta = 3$ mm, using \dot{q} , the heat flux density, $\dot{q} = \dot{Q}/A$, where $A = \pi d_{\text{int}}^2/4$:

$$T_p = T_{p3} - \dot{q} \frac{\delta}{\lambda_{Al}} \quad [K] \quad (\text{III.13})$$

The average MNF temperature at stationary conditions, far from the wall surface (denoted by T_∞), was calculated using the readings of thermocouples that measured the MNF temperature away from the hot wall surface (labelled 2 in Fig.III.12.a, denoted by T_{l1} and T_{l2}):

$$T_l = (T_{l1} + T_{l2})/2 \quad (\text{III.14})$$

The heat transfer coefficient, h , was determined based on the Newton's law of cooling:

$$h = \frac{\dot{q}}{(T_p - T_l)} \quad [W / m^2 K] \quad (\text{III.15})$$

III.2.3. A Comparative Study of Convective Heat transfer in Water and Water Based Magnetic Nanofluid

III.2.3.1. THE EXPERIMENTAL CELL AND SET-UP

The schematic of the first experimental set-up is presented in Fig. III.12.a and its set up in the laboratory is in Fig. III.13. The experimental cell (3) has a cylindrical shape, with lateral walls made of acrylic resin with the characteristic dimensions: $d_{\text{int}} = 16$ mm, $H = 50$ mm ($H/d_{\text{int}} = 3.125$). The heat transfer surface (9) is represented by the top of an aluminium bar attached firmly to the bottom of the cell lateral walls. The bottom of the bar is in contact with a plane heater (10), of resistance $R = 0.14 \Omega$, placed in a ceramic insulating frame (11), powered by the power supply (8). The cell has a resin cap, with two holes to introduce the thermocouples that measured the temperature of MNF sample (7), away from the heat transfer surface. The external walls of the experimental cell and the aluminium bar were insulated by acrylic resin (12). The ambient temperature in the proximity of the experimental set-up was also monitored using an electronic

thermometer. To study the effect of an applied magnetic field, was used a coil (5), powered by a DC power supply (6). T

The aluminium bar temperature is monitored using a set of three thermocouples (labelled by 1 in Fig. III.12.a and detailed in Fig. III.12.b). The thermocouples are inserted in holes drilled in the aluminium bar and thermal conductive grease being used to ensure a very good contact between the thermocouples and the wall. The thermocouples were K-type, 0.5 mm diameter, 0.35 s time response, calibrated.

The main characteristics of the MNF sample are: density, $\rho = 1088 \text{ kg/m}^3$ ($\pm 1.5\%$ accuracy), dynamic viscosity $\eta = 2.4 \cdot 10^{-4} \text{ Pa}\cdot\text{s}$, saturation magnetisation $M_s = 50 \text{ G}$ ($\pm 2\%$ accuracy), volume fraction of nanoparticles 0.012.

The experimental set-up used a National Instruments DAQ NI 6042E (4), 200 kS/s, 16 channels, 12-bit, embedded in a PC and with a dedicated LabView program, a SCXI 1000 amplifier to obtain high accuracy temperature measurements. The heat transfer surface temperature was determined using the methodology presented in Section III.2.2.



Fig.III.13. The experimental set-up

The experiment was carried initially using distilled water (the carrier liquid of the MNF sample), to test the cell, compare the experimental results with known empirical correlations and gather reference data for the MNF carrier liquid. Each run was recorded for data analyses after the system reached steady-state.

III.2.3.2. RESULTS AND DISCUSSION

The distilled water data were compared with a known correlation, recommended in (Ștefănescu et al, 1983):

$$Nu = 0.54Ra^{1/4} \quad (III.16)$$

where Nu is the Nusselt number:

$$Nu = hl/\lambda \quad (III.17)$$

Ra is the Rayleigh number:

$$Ra = Gr \cdot Pr = \frac{g\beta\Delta T l^3}{\nu^2} \cdot \frac{\nu}{a} \quad (III.18)$$

where g is the gravitational constant, β is the volume expansion coefficient, l is the characteristic length, ν is the kinematic viscosity, a is the thermal diffusivity. The results of this comparison are presented in Fig. III.13, where it can be observed that the water data fit this correlation.

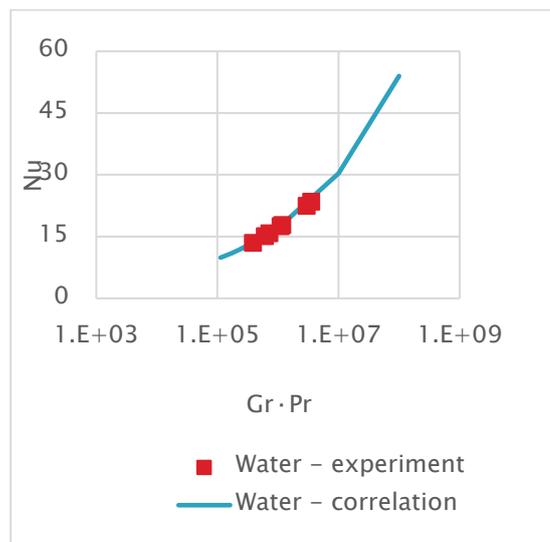


Fig. III.13. Comparison of experimental results for distilled water with correlation Eq. (III.16)

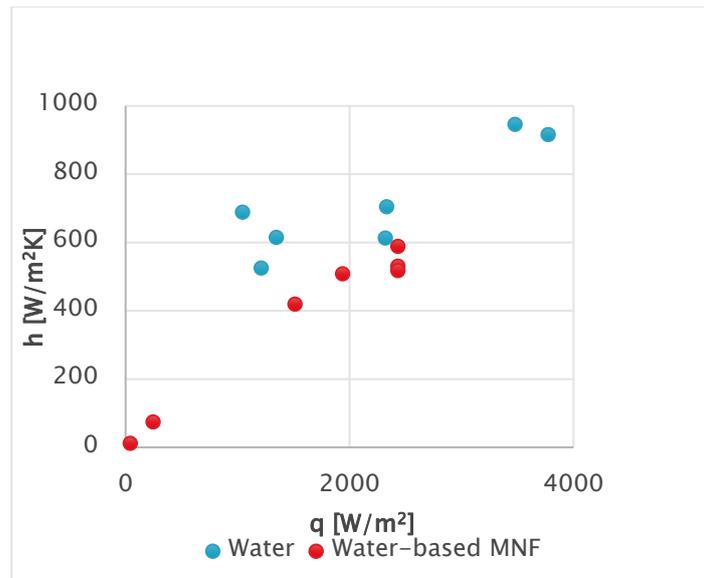


Fig. III.14. Comparison of experimental results for water and MNF sample

A comparison of the heat transfer coefficient versus the heat flux density for water and the MNF sample is shown in Figure III.14. We observe that, for the same heat flux density input, the MNF sample gives lower heat transfer coefficients. The result was similar to those obtained at that date in similar experiments with other type of nanofluids (non-magnetic), like Al_2O_3 , TiO_2 and CuO nanofluids (Wen, Ding, 2005; Li, Peterson, 2006).

The results of this study led to a second experimental set-up, which is going to be presented in the next section.

III.2.4. Natural Convection Enhancement Using a Fe_3O_4 -Water Based Magnetic Nanofluid

Let us observe that, since some of the firsts reported numerical simulations on natural convection of a nanofluid, the reported results showed that, for various working conditions and nanofluid characteristic parameters, an improvement in the heat transfer coefficient is obtained, if the carrier liquid is replaced by a nanofluid (Putra, Roetzel, 2003; Wen, Ding, 2006; Xiao et al., 2009; Aminossadati, Ghasemi, 2009; Jahanshahi et al, 2010). We built a new experimental set-up, with a heat sink, to increase the temperature difference between the hot and cold walls.

III.2.4.1. THE EXPERIMENTAL SET-UP

Figure III.15 shows the new experimental set-up. Compared to the previous experimental cell, this one was cooled at the top wall by a constant flow of water, such that a constant temperature difference between the top and bottom walls was obtained at steady-state.

The new experimental cell (1) is a cylinder with acrylic resin lateral walls, of characteristic dimensions: $d_{int} = 17$ mm and $H = 28$ mm ($L/D = 1.64$).

Similarly, the heat transfer surface is the top of a profiled aluminium bar, firmly attached to the walls, while the base (3) is in contact with the heater (5), of resistance $R = 0.87 \Omega$, connected to a DC power supply (4) of $U_{max} = 32$ V, $I_{max} = 10$ A. The heater is insulated by a ceramic support (6). The cell and aluminium bar are insulated from the ambient with an insulating layer (7). The heat sink is a chamber lid (9) through which is circulated water, in a cooling loop that consists of a pump and a water bath. The water inlet and outlet temperatures were monitored. To fill the experimental cell with the MNF sample, the lid has a central hole through which a plastic tube (8) is passed. After filling, the tube was sealed.

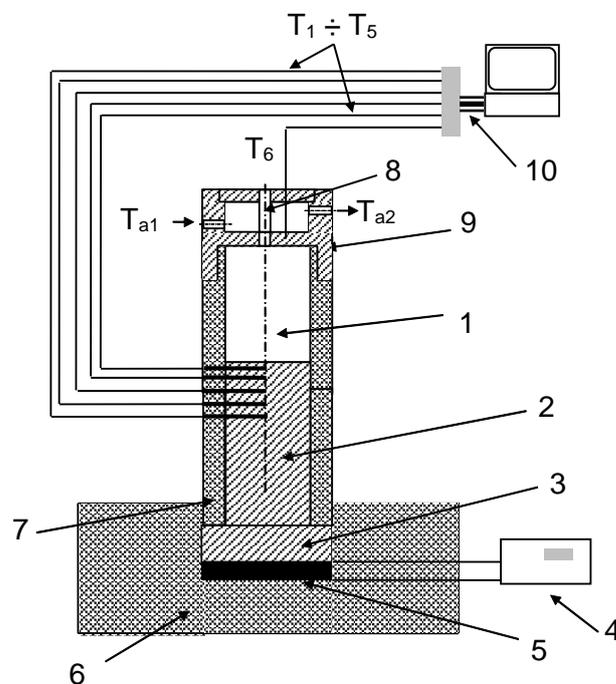


Fig. III.15. Experimental set-up

To determine the heat flux rate and the heated surface temperature, five T-type thermocouples, $T_1 - T_5$, of 0.5 mm diameter, were inserted in the axis of the aluminium bar, at equal distances (5 mm apart, as in Fig. III.12.b) and 5 mm from the heated surface, using a similar procedure as explained before. The cold surface temperature was measured using a thermocouple (6), of the same type, inserted in central position in the lid. The ambient temperature in the proximity of the experimental set-up was also monitored during the steady state measurements, 22.226 ± 0.003 °C.

The experimental set-up used the same data acquisition system (10) type NI DAQCard-6024E, embedded in a PC, and a Labview program for data acquisition and a SCXI 1000 amplifier to obtain highly accurate temperature measurements.

The fluid sample used in this experiment was a water-based MNF, with magnetite nanoparticles dispersed in water, and a double layer stabilization using dodecyl benzene sulfonate (DBS) as surfactant, the main characteristics at 20 °C, are: $\rho = 1066$ kg/cm³ ($\pm 1.5\%$), $\eta = 2.4 \cdot 10^{-4}$ Pa · s, saturation magnetisation $M_s = 55$ G ($\pm 2\%$ accuracy), volume fraction of nanoparticles $\Phi = 0.016$.

III.2.4.2. RESULTS AND DISCUSSION

The measurements were run at first using distilled water, followed by MNF sample testing. Each time the set up was brought to steady state, to collect data for analyses. The methodology presented in Section III.2.2 was used to calculate the heat transfer density, the heat transfer surface temperature and the heat transfer coefficient. The results obtained for water and the water-based MNF sample are presented in Fig.III.16 and Fig. III.17.

The comparison presented in Figure III.16 shows that for the same temperature difference between the hot and cold walls, there is a larger amount of heat transferred by the MNF sample. Also, the experimental data presented in Fig. III.17 show an increase of the heat transfer coefficient of MNF sample compared to that of the carrier liquid (distilled water).

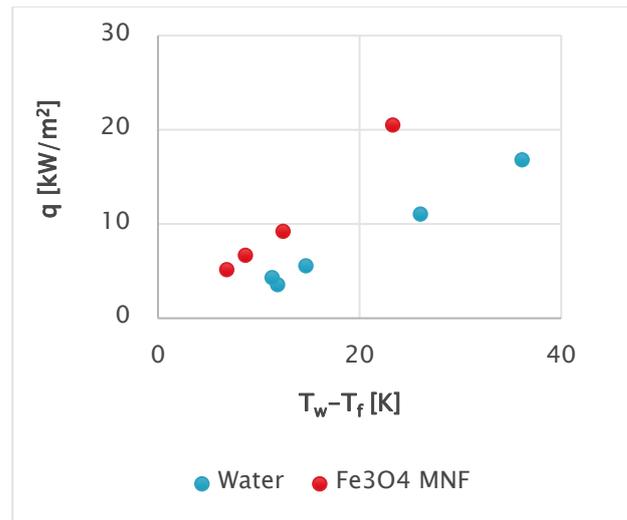


Fig. III.16. Comparison of the experimental data for water and the water-based MNF sample

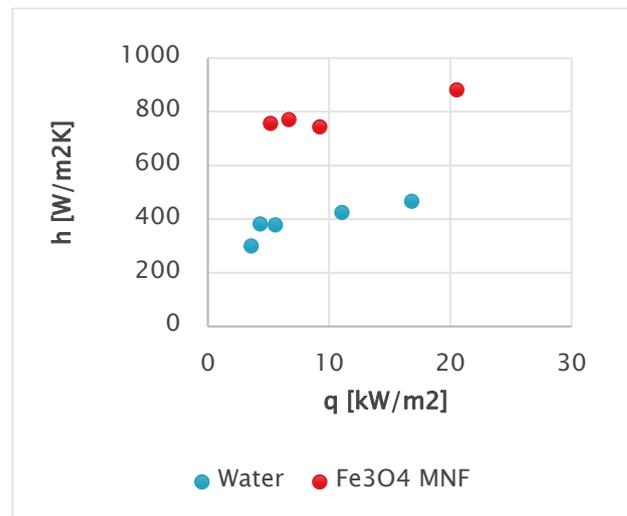


Fig. III.17. Comparison of heat transfer coefficients obtained for water and the water-based Fe₃O₄ magnetic nanofluid

Further, the natural convection heat transfer correlation recommended in (Incropera et al, 2007) for the case of heated lower plate and cooled upper plate, was employed to verify the results:

$$Nu = 0.27Ra^{0.25} \quad (III.19)$$

where the followings were used:

- the mean fluid temperature (taken as $T_m = (T_H + T_C)/2$);
- $\Delta T = T_H - T_C$ is the temperature difference;
- l is the characteristic length (taken as the fluid depth $l = 0.028$ m);

- all fluid properties were taken at the mean fluid temperature, T_m .

The results for both water and the MNF sample are presented in Fig.III.18. We observe that the presence of the stable dispersed nanoparticles led to shift in the flow regime, enhancing the turbulence.

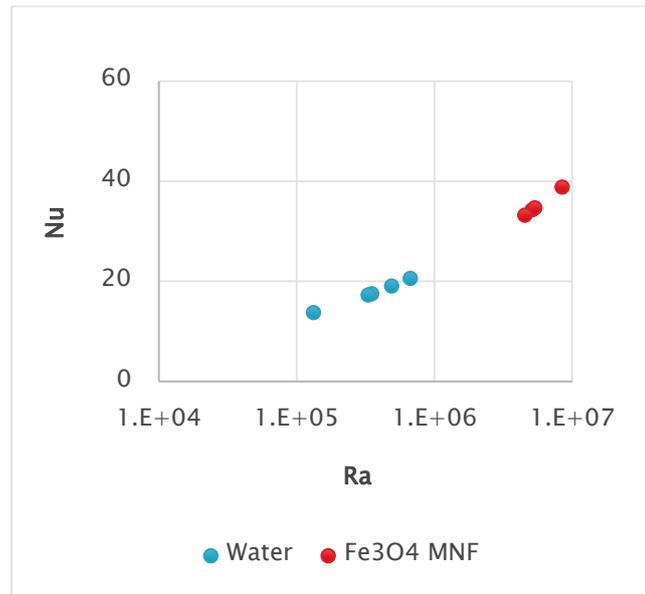


Fig.III.18. Nusselt number correlation Eq.(III.19), for water and MNF sample

Comparing the results for the two experimental set-ups, we observed that, beside to our first experiment (Stoian et al, 2008), when the upper wall of the cell was in contact with the ambient, this time the effect of the driving force largely increased because of a much larger temperature difference $\Delta T = T_H - T_C$. The corresponding data for ΔT in the two experiments were, at the maximum heating regime, of about 4 K and 23 K, respectively.

Let us observe that the results reported in (Putra et al,2003), largely used as a reference for comparison, were obtained with vertical heating and cooling walls, different characteristic geometry ($L/D = 0.5$ and 1.0) and bigger temperature difference, compared to this set-up. Also, in important difference may come from the fact that while the water based nanofluid used in their experiments showed a Newtonian behaviour, in our case it exhibited a non-Newtonian behaviour (Vékás et al, 2005). It is documented up to the present days that the presence of agglomerates and chain-like structures might favour the increase of thermal conductivity of nanofluids and may enhance the heat transfer

in certain conditions (Das et al, 2007; Shima et al, 2009; Ramesh and Prabhu, 2011; Nkurikiyimfura et al, 2013; Nayak et al, 2016).

These experimental results, obtained by changing both operating and geometric characteristics, showed that if conditions are met, natural convection heat transfer enhancement using a nanofluid can be achieved. Additionally, if water-based magnetic nanofluid is used, the non-Newtonian flow behaviour can contribute to a certain extent to the heat transfer enhancement.

III.2.5. Conclusions

The theme presented in Chapter III.2 envisaged the potential of using water-based magnetic nanofluids for heat transfer applications, with a target of using it in a heat exchange device for cooling applications in the automotive industry. The obtained results demonstrated the potential of such application, even without the application of an external magnetic field. Also, it showed for certain working conditions and geometry, a large increase in the heat transfer coefficient can be achieved.

In the framework of the CNCSIS project A665/2005–2007 it was started a pioneer work in our university and our country as well, regarding the use of water-based magnetic nanofluids for convective heat transfer. Its results and experience have been used in the next research theme to be presented.

Reference publication nr.5: Stoian F.D., Holotescu, S., Stoica, V., Bica, D., Vekas, L. (2008), 5th International Conference on New Research Trends in Materials Science, September, 2007, Romania, published in *Journal of Optoelectronics and Advanced Materials*, 2008, Vol.10(4), pp.773–776 (ISI Article/Conference Paper, FI 2015: 0.383)

Reference publication nr .6: Stoian F.D., Holotescu, S. (2012)., *J. Nanoscience Nanotechnology*, Vol.12, Nr.10, pp. 8211–8214 (ISI Article, IF 2015:1.338)

III.3. Thermal properties and heat transfer control by an applied magnetic field, for transformer oil – based magnetic nanofluids

This research topic has been started along with that presented in the previous section. The research regarding the heat transfer control and thermal properties for transformer oil based MNFs envisaged contributions regarding two specific engineering problems: (1) cooling and insulation of power transformers, (2) use of MNFs as core of micro-transformers. The reported results were obtained in the framework of the following projects: PN II 21-043/2007-2010 CFEEL, cooperating as team member with the Laboratory of Magnetic Liquids, RATB, coordinated by National Institute for Research and Development in Electrical Engineering ICPE-CA, Bucharest; FP7 MNT-ERANET, no. 7-018/2009-2011, in an international cooperation, coordinated by the Institute of Experimental Physics, Slovak Academy of Sciences (IEP-SAS); PN II PCCA no.63/2014-2017, in a national cooperation coordinated by ICPE-CA. It represented also the start of a fruitful cooperation, at national level, with research teams from Politehnica University of Bucharest and ICPE-CA, and, at international level, with a research team from IEP-SAS, as well as continued the cooperation with RCESCF – UPT.

III.3.1. Introduction

The idea of using magnetic fluids in power transformers as, either cooling medium or as core, or both, dates back about twenty years ago (Raj, Moskowitz, 1995; Segal, Raj, 1998), and, since then, several research groups studied this topic for which they could find, as always when magnetic nanofluids are used, “a solution” that suited “the problem” and not a generally valid “recipe” (Segal et al, 2000; Morega et al, 2010; Timko et al, 2010; Pislaru-Danescu et al, 2013; Patel et al, 2016). One of the reasons may be the fact that power transformers are electrical equipments that have constructive and working parameters that may vary in large ranges, making it difficult to use one type of magnetic nanofluid for all types of power transformers (Harlow, 2012).

Let us recall that an insulating liquid performs two functions in a power transformer: one is the electrical insulation, which is preventing the electric

contact among the conductive parts, and the other is cooling, by transferring the heat from the windings and core to the tank walls and, further, to the ambient. Usually, the liquids used to accomplish these requirements are mineral oils, synthetic oils or their mixtures. Mineral oils have usually low thermal conductivity and quite high heat capacity, so their cooling efficiency is rather low. On the other hand, synthetic oils can be prepared such that their thermal properties be improved and are the nowadays replacement for mineral oils (Perrier et al, 2006). An additional problem is represented by the thermal aging process to which transformer oils should stand without replacement, for as long as possible (Meshkatoddini, 2008; Alshehawy et al, 2016).

During the last decade, solutions have been proposed and tested, to replace the transformer oils or solid insulators with transformer oil – based MNFs (Segal et al, 2000; Morega et al, 2010, Timko et al, 2010;). Also, very recently, the use of magnetocaloric fluids is also investigated (Patel et al, 2016). Beside the enhanced cooling, improvement of electrical performances and increased life of the equipments can be reached (Kopcanski et al, 2004; Chiesa, Das, 2009; Holotescu et al, 2011; Pislaru–Danescu et al, 2013; Patel et al, 2016).

The possibility of using MNFs as core in power transformers envisaged the miniature size transformers, being at first demonstrated experimentally by (Tsai et al, 2010) and further studied numerically by (Morega et al, 2012), to be applied in a miniature planar transformer. A major difference between the two types of applications is related to the volume fraction of magnetic nanoparticles. Usually, for cooling applications low to very low volume fraction MNFs are used, while for magnetic nanofluid core are needed high volume fraction MNFs. In both cases, the study of their thermo–physical, magnetic and electrical properties should clarify their dependence on the operating conditions in order to find the needed characteristics for the MNF, as well to have the premises of a realistic elaboration and validation of the model for the transformer.

My scientific contribution to the researches carried out in the framework of the above–mentioned projects envisaged the following subjects to date:

- analysis of the cooling potential of the MNF samples and measurements of their thermal properties (heat capacity, thermal conductivity), and
- evaluation of the heat transfer control by an AC applied magnetic field in a transformer oil – based MNF.

III.3.2. Thermal properties of magnetic nanofluids used as cooling and insulating medium in a power transformer

III.3.2.1. INVESTIGATIONS REGARDING THE HEAT CAPACITY OF A MAGNETIC NANOFUID SAMPLE

This work was carried out in the framework of the project PN II 21-043/2007-2010 CFEEL, as a contribution to the characterisation of the magnetic nanofluid prepared at the Laboratory of Magnetic Fluids, RA-TB, that was used as cooling and insulating medium in a low power, single-phase power transformer, for medium voltage installations (TMOF2, 36 kV, 40 kVA), whose prototype was designed by ICPE-CA Bucharest and ICMET Craiova teams, and modelled and numerically analysed by the Politehnica University of Bucharest, as partners in this national project.

To investigate the thermal behaviour of magnetic the nanofluid samples, used an experimental apparatus, realised for this purpose, whose schematic is presented in Fig. III.19. The apparatus was tested at first with distilled water for the interval 40 - 80 °C, to determine the calorimeter constant.

The MNF was prepared using the transformer oil TO-40 as base liquid, in which are stably dispersed Fe_3O_4 nanoparticles using oleic acid as surfactant. The main characteristics of the transformer oil-based magnetic nanofluid sample were: density $\rho_{\text{MNF}} = 963 \text{ kg/m}^3$ and $\rho_{\text{TO-40}} = 866 \text{ kg/m}^3$; saturation magnetisation $M_s = 4010.82 \text{ A/m}$; dynamic viscosity $\eta = 22 \cdot 10^{-3} \text{ Pa} \cdot \text{s}$ (at 20°C), solid volume fraction $\Phi = 0.022$.

The specific heat of the carrier liquid and the magnetic nanofluid sample were determined, for the temperature range 40 - 80 °C, which is the interval of interest for the working conditions of power transformers (Morega et al, 2010; Pislaru-Danescu et al, 2013).

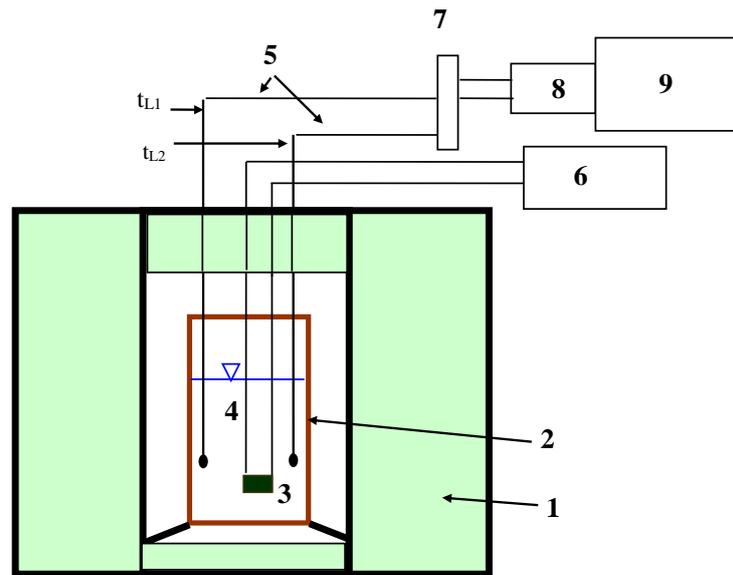


Fig.III.19. Experimental apparatus for calorimetric measurements: 1- insulating housing; 2 - measurement cell; 3 - electric resistance; 4 - liquid sample; T type thermocouples (OMEGA GmbH, 0.5 mm); 6 - power supply (0-32 V, 0-10 A); 7 - conditioning block (NI SC 2345, with thermocouple signal modules SCC TC01); 8 - DAQ system (PCMCIA card NI 6062E); laptop computer

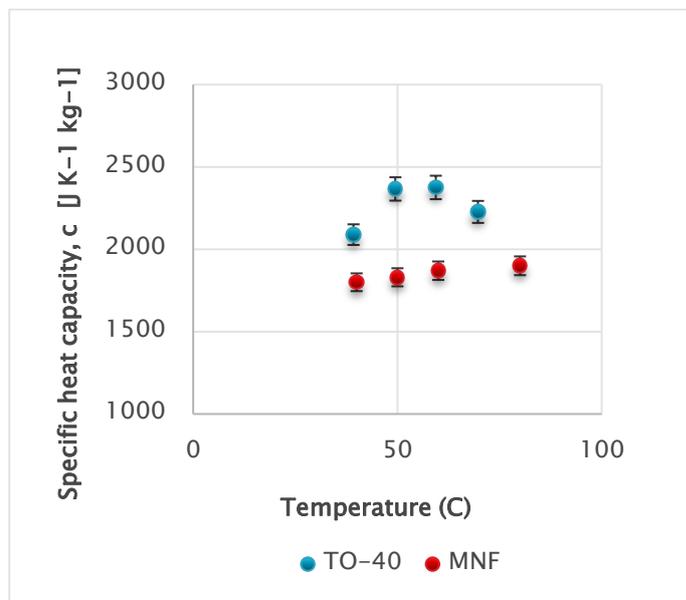


Fig.III.20. Specific heat capacities for the carrier liquid and MNF sample

The comparison of experimental data showed that the MNF sample has a lower specific heat, with respect to that of the carrier liquid (TO-40), and its values are increasing with temperature and agree with the mixture theory formulae (Khanafar, Vafai, 2011).

III.3.2.2. ANALYSIS OF THE COOLING POTENTIAL OF THE MNF SAMPLE

This study was carried out in the framework of the international research project FP7 MNT-ERANET, no. 7-018/2009-2011, that was an interdisciplinary research, in which participated Romanian and Slovak public institutions and companies, coordinated by Dr. Milan Timko, IEP-SAS. The goal of this project was to improve the operating conditions of an existing type of power transformer, produced by the Slovak industrial partner in this project, by replacing the existing insulation with a magnetic nanofluid specially “designed” by the team from the Laboratory of Magnetic Fluids, RA-TB, analysed by the team of RCESCF -UPT and prepared for the Slovak partner by the team of the Romanian industrial partner, ROSEAL S.A. Odorheiu Secuiesc.

In the followings, the analysis that was carried out for the magnetic nanofluid sample chosen for experimentation in the power transformer is presented (Stoian et al, 2013). The MNF sample was prepared, based on transformer oil UTR 40 and Fe_3O_4 nanoparticles, stabilized with a single layer of oleic acid. The main characteristics of the sample are: $\rho_{\text{UTR}} = 867 \text{ kg/cm}^3$ and $\rho_{\text{MNF}} = 937 \text{ kg/cm}^3$; solid volume fraction $\phi = 0.0162$, saturation magnetisation $M_s = 2785.3 \text{ A/m}$, dynamic viscosity $\eta_{\text{UTR}} = 17.5 \cdot 10^{-3} \text{ Pa} \cdot \text{s}$ and $\eta_{\text{MNF}} = 18.5 \cdot 10^{-3} \text{ Pa} \cdot \text{s}$ (both at 20 °C).

The cooling potential of the MNF sample and that of the carrier liquid were determined using the so-called Figure-of-Merit (FOM), as defined in (Bergles & Bar-Cohen, 1993) for natural convection:

$$FOM_{NC} = \left[\beta \rho^2 c_p k^{\frac{1}{n-1}} / \eta \right]^n \quad (\text{III.20})$$

where: $n = 0.25$ for laminar flow and $n = 0.33$ for turbulent flow; β [1/K] – thermal expansion coefficient; ρ [kg/m³] – density; c_p [J/K kg] – specific heat capacity; k [W/mK] – thermal conductivity; η [Pa · s] – dynamic viscosity.

The unknown values of the thermal properties of the MNF sample were determined by calculation, as follows:

- the thermal expansion coefficient was estimated using the mixture formula given in (Ravnik et al, 2010):

$$\beta_{MNF} = \beta_{UTR} \left[\frac{1}{1 + \frac{(1-\varphi)\rho_{UTR}}{\varphi\rho_{NP}}} \frac{\beta_{NP}}{\beta_{UTR}} + \frac{1}{1 + \frac{\varphi}{1-\varphi} \frac{\rho_{NP}}{\rho_{UTR}}} \right] \quad (III.21)$$

where β_{UTR} is the thermal expansion coefficient of transformer oil (Susa et al, 2005), β_{NP} – the thermal expansion coefficient of the magnetite nanoparticles (Nikolaev and Shipilin, 2000).

- the specific heat capacity was determined using the mixture formula for colloidal suspensions, accepted for nanofluids (Khanafar, Vafai, 2011):

$$\rho_{MNF}(t) \cdot c_{p,MNF}(t) = (1-\varphi) \cdot \rho_{TO-40}(t) \cdot c_{p,TO-40}(t) + \varphi \cdot \rho_{NP}(t) \cdot c_{p,NP}(t) \quad (III.22)$$

where the temperature dependence of the transformer oil specific heat capacity was given by:

$$c_{p,TO-40}(t) = 5.025t + 1789.5 \quad (III.23)$$

and the specific heat capacity of the magnetite nanoparticles was given by (Khanafar, Vafai, 2011):

$$c_{p,NP}(T) = 0.6334 + 0.871 \cdot 10^{-3} T \quad (III.24)$$

- the thermal conductivity was determined using the equation from (Holotescu et al, 2011), for the log-normal size distribution of the magnetic core diameter of the nanoparticles:

$$k_{MNF} = k_{UTR} \frac{k_{NP} + 2k_{UTR} + 2\varphi_e(k_{UTR} - k_{NP})}{k_{NP} + 2k_{UTR} - \varphi_e(k_{UTR} - k_{NP})} \quad (III.25)$$

where $\varphi_e = \varphi \left\langle (D_m + \delta)^3 \right\rangle^2 / \left\langle (D_m + \delta)^2 \right\rangle^3$

in which D_m (nm) – magnetic diameter; δ (nm)– non-magnetic layer double thickness, resulted from magnetic measurements (Holotescu et al, 2011).

k_{UTR} – thermal conductivity of the transformer oil UTR 40,

k_{NP} – thermal conductivity of the magnetic nanoparticles, estimated from (Lis, Kellard, 1968)

The data for the transformer oil and MNF sample are presented in Table III.1, all property values being calculated at room temperature.

Table III.1. Calculated thermal properties for the transformer oil and magnetic nanofluid sample

Property	UTR	NP	MNF_UTR	$\Delta X/X_{UTR}$
c_p [J/kg K]	1915.125	1.1092	1743.392	- 8.97 %
k [W/m K]	0.127	1.39	0.13318	+ 4.86 %
β [1/K]	$7.15 \cdot 10^{-4}$	$1.2 \cdot 10^{-4}$	$6.62 \cdot 10^{-4}$	- 7.47 %
FOM (n=0.25)	18.373		18.963	
FOM (n = 0.33)	90.265		92.686	

We observe that, for natural convection, the FOM value for the MNF sample is slightly higher compared to that of the carrier liquid if the flowing regime is turbulent. Thus, for low volume fraction MNFs, with Newtonian behaviour (Stoian et al, 2013), the natural convection heat transfer is only slightly improved, compared to the carrier liquid case.

III. 3.3. Thermal properties of magnetic nanofluids used as liquid core of a miniature planar transformer

This work is part of an ongoing national project, PN II PCCA 63/2014–2017, being an interdisciplinary research, coordinated by ICPE–CA Bucharest. The main contribution from the participating teams from Laboratory of Magnetic Fluids (RA–TB) and RCESCF (UPT) are regarding the magnetic liquid core preparation and characterisation, for use in a miniature planar transformer – a part of the DC/DC converter in an energy harvesting hybrid system, through photovoltaic and piezoelectric conversion.

My scientific contribution to this project is regarding the study of the thermal properties and electrical properties of the MNF liquid core.

Thermal conductivity measurements were carried out for magnetic nanofluid samples, carrier liquids and surfactants used in this project, using two methods and dedicated measuring systems:

- heat flow method, using a FOX 50 Heat Flow meter;
- thermal needle probe method, using a Decagon KD2 Pro apparatus.

The results are part of a patent request at OSIM, the State Office for Patents and Trademarks, Patent request no. A/00713/07.10.2016, approved but not yet published.

III. 3.4. Heat transfer control in a transformer oil – based magnetic nanofluid by an applied magnetic field

III.3.31. THE EXPERIMENTAL SET-UP

The aim of this work, also a part of the international research project FP7 MNT-ERANET, no. 7-018/2009-2011, was to evaluate the cooling potential of a magnetic nanofluid in an applied magnetic field, for use as cooling medium in a power transformer. To accomplish this, the cooling of a coil powered by a 50 Hz AC power supply using Fe₃O₄ transformer oil based magnetic was investigated experimentally (Stoian, Holotescu, 2014). Using this experimental set-up, the MNF sample and transformer oil analysed in Section III.3.2.2 were tested.

The experimental set-up is presented in Fig. III.21. The measurement cell (1) is a cubic vessel with Plexiglas walls, of size 60 x 60 x 60 mm³. A coil (4) of R = 680 Ω, is placed at the center of the cell, on a plastic frame (3), which allows for the circulation of the fluid outside the coil core (of size 15 mm x 15 mm x 18 mm). A constant volume, equal to $V_f = 130 \text{ cm}^3$, of transformer oil and MNF sample were tested. The coil was powered by the AC (50 Hz) power supply (7), voltage set at 40 V.

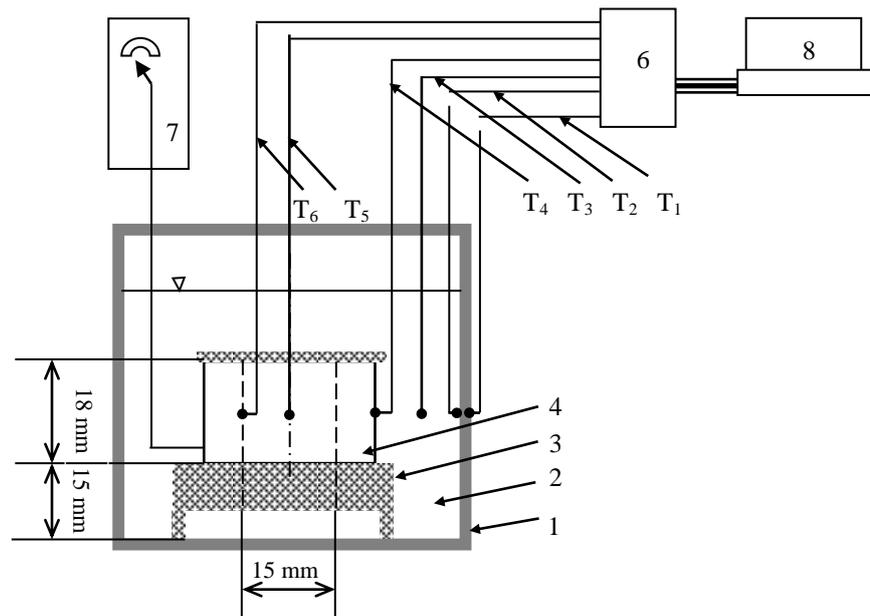


Fig. III.21. The experimental set-up

The temperature inside the cell and at its walls was monitored using six T - type thermocouples (0.5 mm, Teflon insulated, standard error of $\pm 1^{\circ}\text{C}$ or $\pm 0.7\%$), denoted by T_1 to T_6 , all placed in the same in the same horizontal plane:

- (T_1) and (T_2) on the walls of the measurement cell wall,
- (T_4) and (T_6) on the external and internal walls of the coil,
- (T_3) at the middle point between (T_2) and (T_4), and
- (T_5) in the central axis of the coil core.

A DAQ system (6), consisting of a NI DAQ PCMCIA 6062E card, a NI SC 2345 connector block with and NI SCC TC-01 modules to connect the thermocouples, to obtain accurate temperature measurements, and a laptop computer with a LabView program for data recording, were used for running the experiments. The sampling rate was 1 sample/s.

Prior to the experiments, the flux density, B , generated by the coil was measured using a FW Bell 5080 Gaussmeter. The measurement was carried out using the axial or transversal Hall probes, depending on the position of the point of measurement with respect to the generated magnetic field. The measurement points map is shown in Fig.III.22 and the measured values are given in Table III.2.

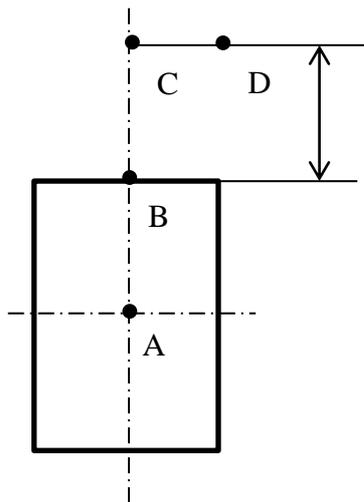


Table III.2. Measured values of the magnetic flux density.

Point	B [mT]
A	8.1
B	4.9
C	1.4
D	1.0

Fig. III.22. Indication of the magnetic flux density measurement points in the coil core and above it

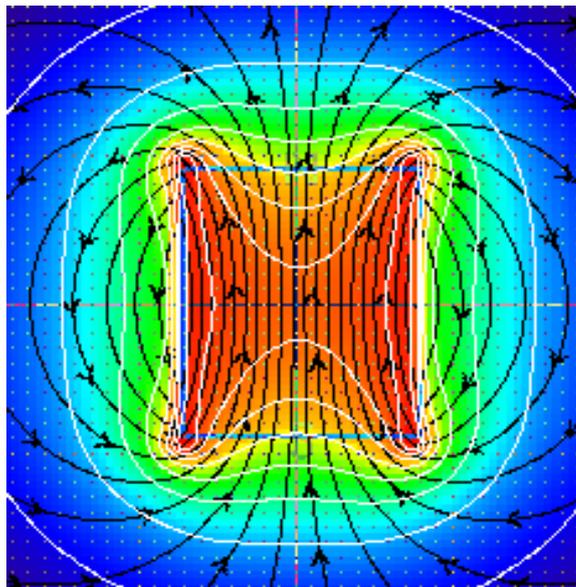


Fig. III.23. Model of the coil magnetic field lines (black lines with arrows) and the flux density contours (white lines)

The model of the magnetic field lines and the magnetic flux density contours, presented in Fig. III.23, were obtained using Vizimag, a software for visualising the magnetic field, to which the coil characteristics were input. We observe that the effect of the magnetic field is limited to the core and its vicinity.

III.3.3.2. RESULTS AND DISCUSSION

Each experiment run had three periods: (I) – a transitory heating regime characterised by a temperature rise rate starting from the room temperature, (II) – a stationary heating regime, of constant average temperature, and (III) – a cooling regime, characterised by a temperature decrease rate until the cell returned to the room temperature. In Fig.III.24 and Fig.III.25 are presented the stationary heating regimes for the transformer oil and the MNF sample, respectively.

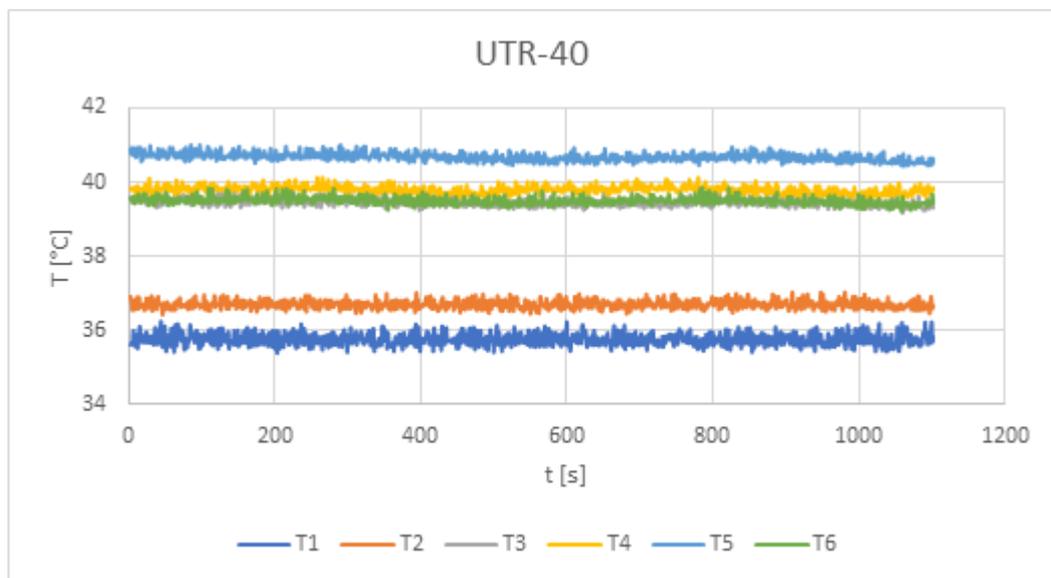


Fig. III.24. Stationary AC heating regime for transformer oil UTR 40

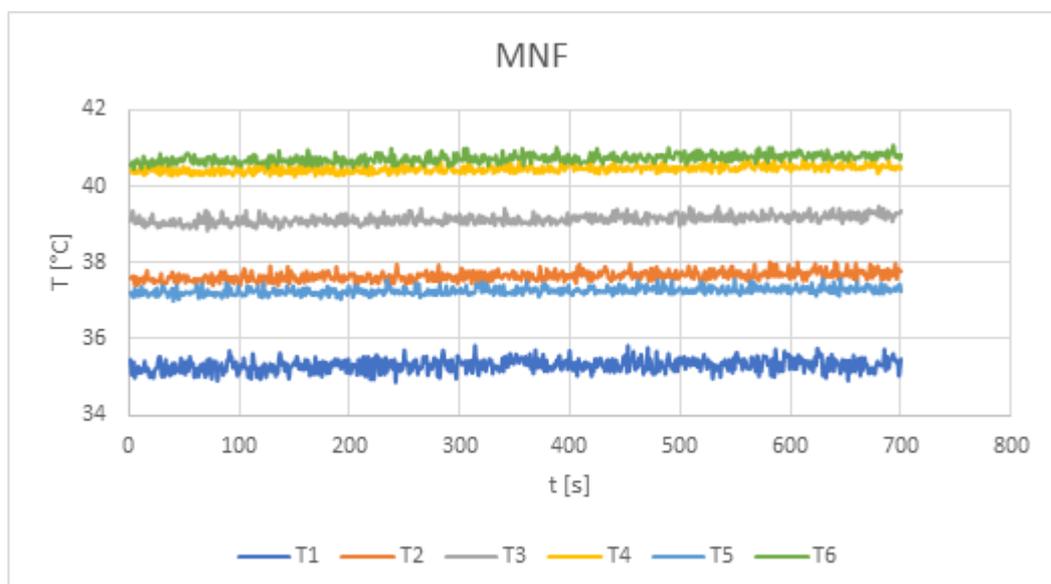


Fig. III.25. Stationary AC heating regime for the MNF sample

The average values for each stationary AC heating regime are presented in Table III.3. It was observed that the coil walls temperatures are slightly higher for the MNF sample compared to the transformer oil, a possible explanation being the adherence of magnetite nanoparticles at the walls. Outside the coil there is no significant difference between the corresponding average temperatures for the two fluids, which may be motivated by the low magnetic volume fraction, $\Phi_M=0.00625$ (Stoian et al, 2013) and the weak magnetic field outside the coil.

Table III.3. Average temperatures for the stationary heating regime

Working fluid	$T_{1,avg}$ [°C]	$T_{2,avg}$ [°C]	$T_{3,avg}$ [°C]	$T_{4,avg}$ [°C]	$T_{5,avg}$ [°C]	$T_{6,avg}$ [°C]
UTR 40	35.7	36.7	39.4	39.8	40.6	39.4
MNF	35.3	37.6	39.1	40.4	37.2	40.7

A significant difference, of 3.4 °C, could be observed between the stationary average values in the coil axis, indicating that the magneto-convection occurred in this region due to higher magnetic field (point A in Table III.2). Therefore, the heat transfer in this region was further analysed using the heating and cooling regimes data. The heating and cooling curves in the coil axis (T_5) are presented in Fig. III.26 and Fig. III.27.

Using the polynomial trend lines obtained from Fig. III.26 and Fig. III.27, the temperature rise rate and temperature decrease rate were calculated, the results being presented in Fig. III.28. We observed that the MNF temperature rise rate (denoted by “heat”) was lower compared to that of transformer oil. Consequently, due to the magneto-convection, the stationary regime was approached at a lower temperature when the MNF was used.

In what it concerns the results for the temperature decrease rate, we observed that this parameter was larger in case of the MNF sample, due to its higher viscosity

The MNF temperature decrease rate for the explored temperature range is larger than that corresponding to the TO-40A. That is, due to higher viscosity of magnetic nanofluid compared to that of transformer oil and the absence of the magnetic field, MNF has a lower heat transfer rate.

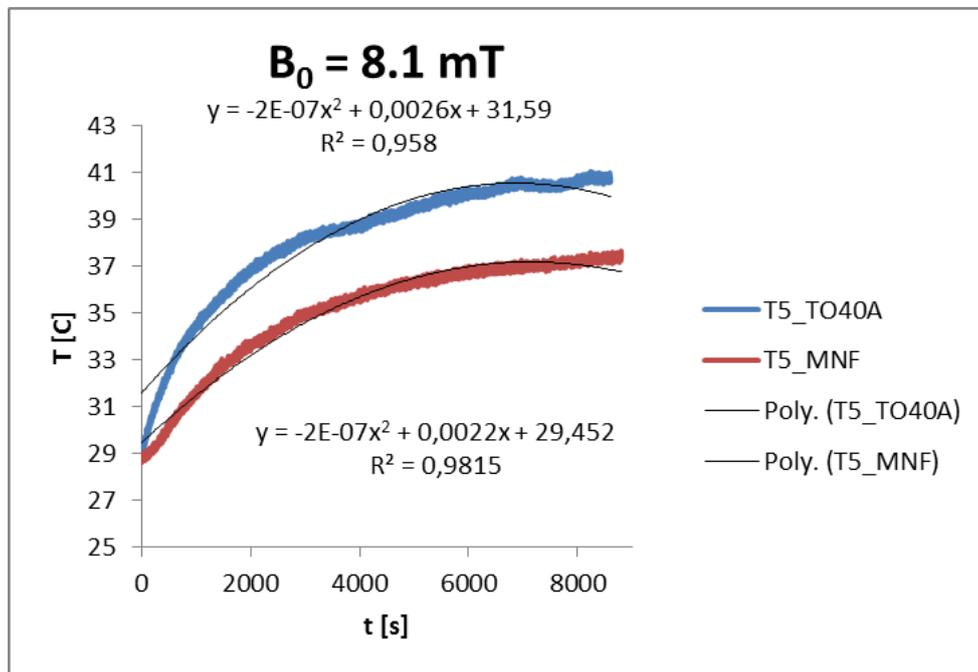


Fig. III.26. Heating curves in the coil axis (T_5), (Stoian, Holotescu, 2014)

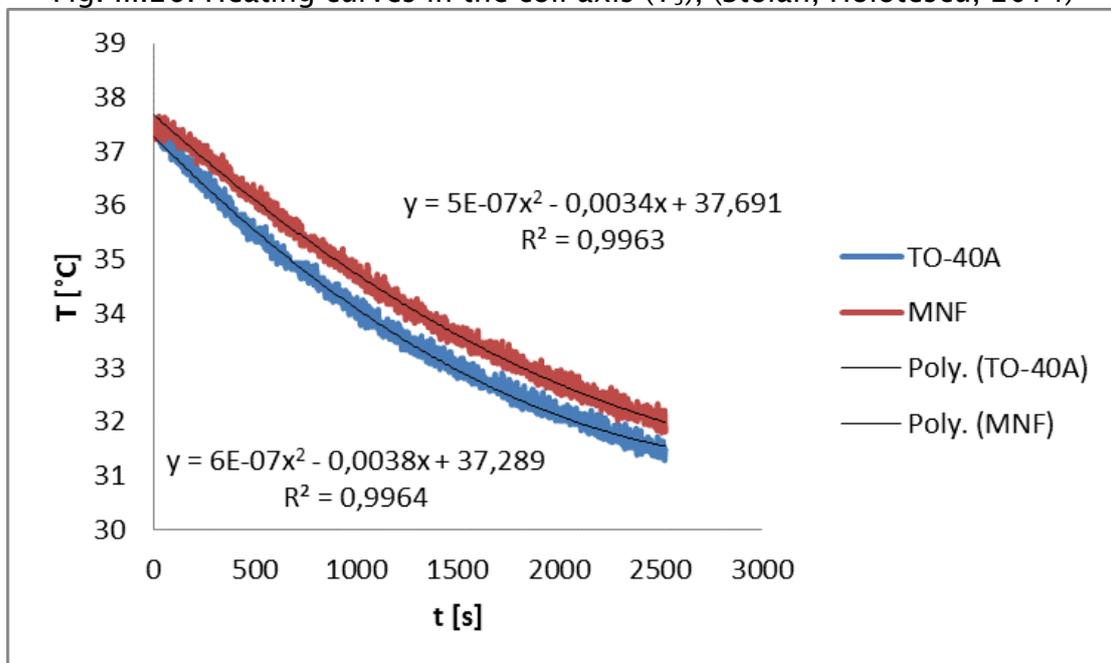


Fig. III.27. Cooling curves in the coil axis (T_5) (Stoian, Holotescu, 2014)

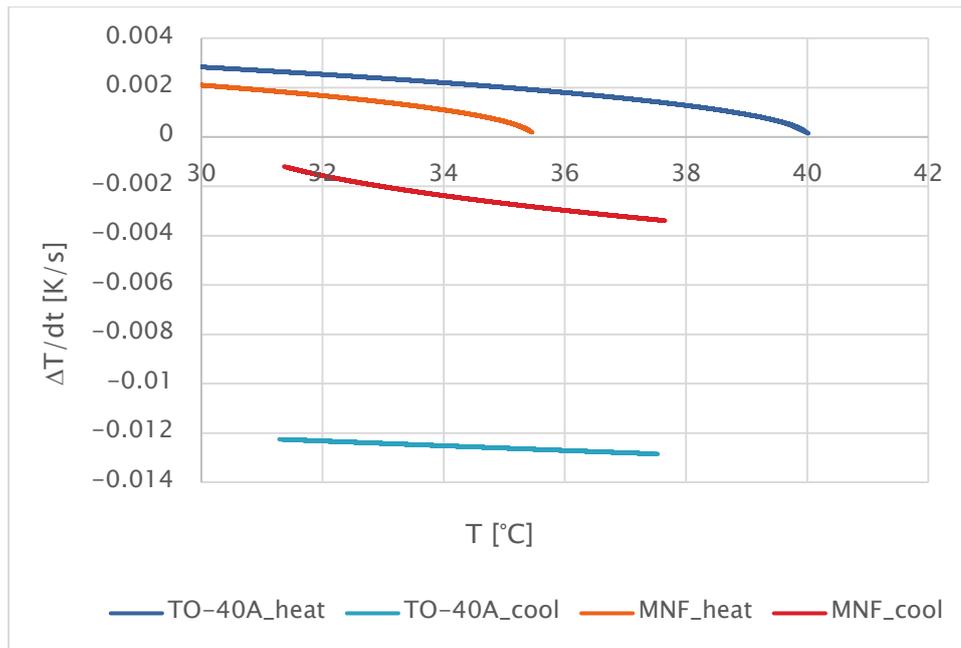


Fig. III.28. Temperature rise rate and temperature decrease rate, for UTR 40 and MNF sample, (Stoian, Holotescu, 2014)

The results led us to the conclusion that the magnetic nanofluid sample, although of low magnetic volume fraction, has potential to be used as cooling medium in a power transformer, if the magnetic field is high enough to generate magneto-convection in the bulk of the fluid.

III.3.5. Conclusions

The research results presented in Section III.3 envisaged the transformer oil - based magnetic nanofluids, with respect to two types of applications: for cooling and insulation in power transformers and as magnetic liquid core in miniature transformers. The themes we are studying are pioneer work and new at national level, and our group is among the few that are working in this area of application of magnetic nanofluids, at international level.

The personal contributions refer to the measurements and analyses of thermal properties and cooling potential of the analysed magnetic nanofluids, including the evaluation of heat transfer control by an applied AC magnetic field.

Reference publication nr.7: Morega, A.M., Morega, M., Pislaru–Danescu, L., Stoica, V., Nouras, F., **Stoian, F.D.** (2010). A Novel, Ferrofluid–Cooled Transformer. Electromagnetic Field and Heat Transfer by Numerical Simulation. 12th International Conference on Optimization of Electrical and Electronic Equipment, OPTIM 2010, 20–22 May 2010, Brasov, Romania, Publisher IEEE, Print ISBN: 978–1–4244–7019–8, , [10.1109/OPTIM.2010.5510425](https://doi.org/10.1109/OPTIM.2010.5510425), (ISI Conference Paper).

Reference publication nr.8: Holotescu, S., **Stoian F.D.**, Marinica, O., Kubicar, L., (2011). Utilization of the magnetogrulometric analysis to estimate the thermal conductivity of magnetic fluids, *Journal of Magnetism and Magnetic Materials*, 2011, Vol.323 (10), pp.1343–1347 (ISI Article/Conference Paper, IF 2015: 2.357)

Reference publication nr 9: **Stoian F.D.**, Holotescu, S., Taculescu, A., Marinica, O., Resiga, D., Timko, M., Kopcanski, P., Rajnak, M. (2013). Characteristic Properties of a Magnetic Nanofluid Used as Cooling and Insulating Medium in a Power Transformer, the 8th International Symposium on Advanced Topics in Electrical Engineering (IEEE Conference), Bucharest, May 23–25, 2013, ISBN: 978–1–4673–5980–1; 978–1–4673–5979–5, (ISI Conference Paper)

Reference publication nr 10: **Stoian F.D.**, Holotescu, S. (2014). Experimental study of cooling enhancement using a Fe₃O₄ magnetic nanofluid, in an applied magnetic field, 32nd Italian Union of Thermo–Fluid–Dynamics (UIT) Heat Transfer Conference Location: Pisa, ITALY Date: JUN 23–25, 2014, Book Series: Journal of Physics Conference Series, Volume: 547, Paper no. 012044 (ISI Conference Paper).

Patents:

1. Patent no. 126613 B1/2014: **Transformator cu agent de racire cu nanofluid magnetic** (Transformer with magnetic nanofluid cooling agent)

Authors: Pislaru–Danescu, L., Macamete, E., Telipan, G., Pinte, J., Nouras, F., Paduraru, N., Vekas, L., **Stoian, F.D.**, Borbath, I., Borbath, T., Morega, A.M., Morega, M.

Owners: ICPE–CA Bucharest, ICMET Craiova and ROSEAL Odorheiu Secuiesc

2. Patent request no. A/00713/07.10.2016: **Transformator planar cu nanofluid magnetic** (Planar transformer with magnetic nanofluid)

Authors: Pislaru–Danescu, L., Popa, M., Ilie, C.–I., Chihai, R.A, Babutanu C.A., Nicolae, S., Bunea, F., **Stoian, F.D.**, Holotescu, S., Marinica, O., Morega, A.M., Morega, M., Dumitru, J.B., Popa, N.C.

Owners: ICPE–CA Bucharest and Politehnica University Timisoara (UPT)

IV. Academic and professional achievements

This section describes the main academic and professional achievements since defending the PhD Thesis, on November 13, 1997, at that date being a fresh Lecturer at the Chair of Chair of Thermodynamics, Thermal Machines and Road Vehicles, Politehnica University Timișoara. The achievements will be presented in their timeline.

IV.1. Academic activities achievements

1. Academic teaching

From October 1st, 1997 till September 30, 2000, I held an academic position of Lecturer, with main teaching activities for the following undergraduate level disciplines: Technical Thermodynamics and Thermal Machines, Bases of Technical Thermodynamics, Thermal Power Plants and Equipments, working with students from the Mechanical Engineering Faculty and the Electrical and Power Engineering Faculty.

Starting October 1st, 2000, till present, I have the academic position of Associate Professor, at the Department of Mechanical Machines, Technology and Transportation, Politehnica University Timișoara.

Over the last fifteen university years I have been involved in teaching activities (lectures, seminaries, laboratory classes), as follows:

- undergraduate level disciplines: Technical Thermodynamics, Thermal Power Plants, Thermal Power Plants and Equipments, Thermal Plants, Energy Cogeneration Systems and Equipments.
- graduate level disciplines: Advanced Management Methods for Thermal Systems, Management and Quality of Thermal Energy, Energy Management and Quality of Energetic Processes, Thermal Energy Conversion Systems, Complements of Thermodynamics, Unconventional Power Plants.

Most of the undergraduate disciplines are taught at the Mechanical Engineering Faculty, except for the lecture Thermal Power Plants, taught at Electrical and Power Engineering Faculty, since 2004. All graduate

disciplines have been taught at Master programs that runned at our department: Master Course Energo-ecology in the field of mechanical engineering and vehicles (2004 - 2013), Master Course Engineering of Renewable Energy Systems (2011-2013), Master Course Engineering of Automotive Propulsion Systems (2016-).

Also, starting 2004, I am teaching at the *ARCE (Romanian Agency for Energy Conservation) Expert training Program*, held at the Faculty of Electrical and Power Engineering, Politehnica University Timisoara. The lectures are held on the following subjects: thermal energy balances, electrical energy balances, energy management in industries.

In 2009, from October 5 to October 11, I was invited at the Tamagawa University, Machida, Tokyo, Japan, to give a class (15 hours of teaching) to graduate students attending the Environmental Soft Energy Master Course at the Graduate School of Engineering, with the subject: *Novel thermal fluids for enhanced heat transfer in energy-efficient equipments*.

In 2014, from September 28 to October 10, I participated as Foreign Expert in the European Structural Funds Project PhysNet - International Virtual Laboratory of Progressive Materials Physics, Project No. 26110230097, 9/2013 - 8/2015, Activity 3.2. International Virtual Laboratory of Physics of Nano and Biomaterials, 28.09.2014 - 10.10.2014, at the Institute of Experimental Physics, Slovak Academy of Sciences, Kosice, Slovakia.

2. New courses for graduate students

Since 2004, I also disseminated some of research results from past or ongoing projects as teaching subjects for the lecture "Advanced Management Methods for Thermal Systems" for the Master Course "Energo-ecology in the field of mechanical engineering and vehicles", at the Mechanical Engineering Faculty, later the title has been changed to "Management and Quality of Thermal Energy" and further to "Energy Management and Quality of Energetic Processes".

3. Textbooks, Lecture notes, teaching materials

I continuously sustained the teaching activity with teaching materials, based on reference textbooks on the fundamentals of the taught subjects, research results obtained in my personal activity as well as from reference

scientific literature, in order to give to my students the essential knowledge and means for understanding each new taught item, through documented examples. A list of published textbooks, lecture notes and applications supporting materials is presented as follows:

1. Floriana Daniela Stoian, Termotehnica (Technical Thermodynamics), Editura Politehnica, Timisoara, ISBN 978-606-35-0091-6, 2016
2. Floriana Daniela Stoian, Procese si instalatii termice pentru centralele termoelectrice (Thermal processes and installations for thermal power plants), Editura Politehnica, Timisoara, ISBN 978-973-625-725-4, 2009
3. Jadaneant Mihai, Nagi Mihai, Stoian Floriana, Ionel Ioana, Laza Ioan, Lelea Dorin, Oprisa-Stanescu Paul Dan, Mihon Liviu, Bazele termoenergeticii – Note de curs pentru managerii energetici (Basics of thermal power engineering – Lecture notes for power engineering managers), Editura Orizonturi Universitare, Timisoara, ISBN 973-638-257-5, 2006
4. Jadaneant Mihai, Oprisa-Stanescu Paul Dan, Laza Ioan, Ionel Ioana, Stoian Floriana, Lelea Dorin, Nagi Mihai, Mihon Liviu, Intocmirea si analiza bilanturilor energetice – Note de curs pentru auditorii energetici (Power balances draw up and analysis – Lecture notes for power auditors), Editura Orizonturi Universitare, Timisoara, ISBN 973-638-259-1, 2006
5. Laza Ioan, Lelea Dorin, Mihon Liviu, Nagi Mihai, Oprisa Stanescu Paul Dan, Stoian Floriana, Echipamente si instalatii termoenergetice – Note de curs pentru managerii energetici (Thermo-energetic equipments and installations – Lecture notes for power engineering managers), Editura Orizonturi Universitare, Timisoara, ISBN 973-638-175-7, 2004
6. Ardelean Z., Neacsu Eleonora, Stoian Floriana, Nagi, M., Jadaneant M., Partea termo a centralelor (Thermal power plants), Editura Mirton, Timisoara, ISBN 973-658-669-1, 1999.
7. Jadaneant Mihai, Ionel Ioana, Stoian Floriana, Pop Gheorghe, Lelea Dorin, Stoica Virgil, Negoitescu Arina, Termotehnica si masini termice in experimente (Technical thermodynamics and heat engines in experiments), Editura Politehnica, Timisoara, ISBN 973-8247-11-X, 2001 (Laboratory textbook)

8. Floriana Daniela Stoian, Termotehnica (Technical Thermodynamics), electronic format of lecture notes and presentation files, available at cv.upt.ro
9. Floriana Daniela Stoian, Partea termo a centralelor (Thermal power plants), electronic format of lecture notes and presentation files, available at cv.upt.ro

Starting the university year 2015/2016, I am teaching Technical Thermodynamics for the students attending the undergraduate TCM-IFR (learning with reduced attendance), run through the Virtual Campus of Politehnica University Timisoara.

4. Coordination of diploma projects and dissertations

Starting 2002, undergraduate and graduating students have been involved in our research projects, developed at the RCESCF (UPT), realising their diploma project or master thesis, based on their participation. Thus, 3 students from our department, as well as 1 student from the Power Systems Department, have been involved directly either one in the following projects: CNCSIS AT.2001–2002, PNCDI I 11657/2001–2004, CNCSIS A665. Also, more than ten other undergraduate and graduate students took use of the infrastructure developed through research projects and that available at the RCESCF (UPT), to accomplish their graduating theses. Moreover, 3 former PhD students were actively enrolled in the research projects that I coordinated at Politehnica University.

5. Contributions to the infrastructure for academic activities

In 2007 and 2008, I participated in the internal project competition in Politehnica University Timisoara, for funding the laboratory infrastructure, for educational purposes. The two projects that I submitted were approved and, thus, the Laboratory of Technical Thermodynamics, Room 111, 1st Floor, of the Faculty of Mechanical Engineering main building, was upgraded and transformed into the Laboratory of Thermodynamic Analysis. Through these projects, at first the lab was equipped with 16 PCs, which are still in use for classes with both undergraduate and graduate students, new electrical installation and internet network connection.

The second project added to the laboratory inventory an academic bench for data acquisition, that included a stand-alone system with PC, 10 student kits

for data acquisition and parts, also in use at present with master students for laboratory classes (e.g. Bases of experimental research for road vehicles – master class) as well as diploma projects.

Also, since 2001, through running research projects, I introduced in the inventory of the UPT Library, 15 technical books, from recognised international publishing houses, in the fields like: Technical Thermodynamics, Heat Transfer, Power Engineering, Thermal Management, and 1 educational software (FEM Heat transfer analysis software), that are used in the teaching activity.

IV.2. Professional achievements

In the followings, I will introduce the most important professional achievements since the defending of my PhD:

1. Research publications:

- **9 ISI Articles** (3 as first/corresponding author),
- **7 ISI Conference Proc. Papers** (5 as first/corresponding author)
- **1 Conference Proc. Papers** in other international databases (SCOPUS)
- **29 Communications** at national and local conferences
- **2 National Patents:** Patent no. 126613 B1/2014 and Patent request no. A/00713/07.10.2016, accepted, to be published in March 2017
- **1 monography** (single author).

2. Coordination of research projects:

- Project coordinator for 1 National Research Grant for Young Scientists, awarded by the National Council of Scientific Research in Higher Education (CNCSIS, RO: Project code CNCSIS AT 147/2001 and code CNCSIS AT 75/2002), entitled “Study of the fluid phase transition near critical state under the influence of external force fields”.
- Project coordinator for 1 National Research Grant, awarded by the National Council of University Scientific Research (CNCSIS, RO: Project code CNCSIS 665/2005–2007), entitled “Researches regarding the use of magnetisable nanofluids as thermal fluids”.
- Project responsible for 1 bilateral cooperation project Romania – Italy, COPBIL Theme 45, (2001–2002).
- Project responsible for the team from Politehnica University Timisoara in five National Research Projects (PNCDI I) realised in cooperation with other Romanian

research teams, in the framework of the 1st National Research, Development and Innovation Plan, between 2001 – 2007: Contract UPT 11657/2001–2004 – PNCDI AEROSPATIAL, Contract UPT no. 15908/2001 – PNCDI MATNANTECH, Contract UPT no. 14266/2003 – PNCDI CERES, Contract UPT no. 4–139/2005–2006 – PNCDI CERES, Contract UPT no. 12811/2007 – PNCDI CEEX .

- Project responsible for the team from Politehnica University Timisoara in 2 National Research Projects (PN II), realised in cooperation with other Romanian research teams, in the framework of the 2nd National Research, Development and Innovation Plan between 2012 – 2017: PN II 157/2012–2016; PN II 63/2014–2017.
- Project coordinator for the Romanian team in an international project FP7 Program MNT.ERA–NET, founded for Romanian partners by the Executive Agency for Higher Education, Research, Development and Innovation Funding (UEFISCDI), Project 7–018/2009–2011.

3. Research stages and specialisations

1. 06.06.1998 – 17.09.1998: Invited researcher, University of Pisa, Italy. Research subject: Effects of electric fields on boiling of dielectric liquids, in the framework of a research project of Italian Space Agency (ASI) and European Space Agency (ESA). Host scientist: Professor Walter Grassi.
2. 17.02.1999 – 16.02.2000: Post – doctoral Fellowship JSPS (Japan Society for the Promotion of Science), Faculty of Engineering, Kawagoe Campus, Toyo University, Tokyo, Japan. Research subject: Electric fields effects on critical fluids – studies of modelling and simulation by molecular dynamics. Host scientist: Professor Toru Maekawa.
3. . 25.08.2004 – 24.10.2004: Senior Fellowship CNR – NATO (Consiglio Nazionale delle Ricerche, Italy – Nord–Atlantic Treaty Organisation), Department of Energetics, Faculty of Engineering, University of Pisa, Pisa, Italy. Research subject: Micro – nanoscale effects of electric fields on boiling – modelling and simulation by molecular dynamics. Host scientist: Professor Walter Grassi.
4. . 13.06.2011 – 18.06.2011: Thermal Measurements and Inverse Techniques Course, Eurotherm Seminar No.94 METTI 5 Advanced Spring School, Roscoff (France), in the framework of PN II ID 76/2010 (Complex Projects of Frontier Research).

4. Presentations at international seminars and workshops abroad:

- 1) Effective thermal conductivity of nanofluids and particulate composites: influence of the particle size distribution, Workshop NANOFLUID IX, 29.09 – 2.10.2014, Stara Lesna, Slovakia.
- 2) Experimental Study of Natural Convection Enhancement Using a Fe₃O₄–Water Based Magnetic Nanofluid, presented at the 6th International Conference Diffusion in Solids and Liquids, 05.07 – 07.07.2010, Paris, France.
- 3) Magnetizable nanofluids: progresses and prospects on their use in thermal applications (invited lecture), presented at the 29th Japan Symposium on Thermophysical Properties, Japan Women University, 08.10–10.10.2008, Tokyo, Japan.
- 4) Effects of an applied magnetic field on the bubble growth and departure in a magnetic liquid, presented at the 3rd International Symposium on Two-Phase Flow Modelling and Experimentation, 22.09 – 24.09.2004, Pisa, Italy.
- 5) Analysis of a thermodynamic system at nanoscale by molecular dynamics simulation, presented at the University of Pisa, 18.10.2004, Pisa, Italy.
- 6) European Space Research & Technology Centre (ESTEC), ESA Topical Team on Boiling Meeting, 24 – 25.03.2003, Noordwijk (Netherlands).
- 7) Effects of external fields on bubble behaviour, presented at the ESA Topical Team on Boiling Meeting, 15.02.2001, University of Pisa, Pisa, Italy.
- 8) Electrohydrodynamic effects on nucleate boiling heat transfer, presented at the University of Pisa, on 22.06.1998, Pisa, Italy.

5. Organisation of seminars, workshops, symposia:

- Coordinator of the organising committee for the Symposium “Man and Environment” (2001, 2003, 2004, 2005, 2007 editions), organised mainly in the framework of the Timisoara Academic Days
- Co-chair of two editions of the international workshop "New Opportunities for Interdisciplinary Research in BIO–NANO–SCIENCE", the 1st edition held on June 21–22 2012, at Politehnica University Timisoara, and the 2nd edition held on June 3rd, 2013, at the institute of Biology, Romanian Academy, Bucharest.

6. Expert membership:

- member of the Topical Team on Boiling, organised at the European Space Agency, between 2001 and 2003;
- member of the Working Group for the preliminary form of the National Research Plan 1998 – 2005, organised at the Ministry of Research and

Technology, Topic: Clean Energy Production Technologies, between August – September 1998.

7. **Awards:**

Award Grant for the published research results, Grant PN-II-RU-PRECISI-2013-7 from 2013, for the article “Experimental Study of Natural Convection Enhancement Using a Fe₃O₄-Water Based Magnetic Nanofluid”, published in Journal of Nanoscience and Nanotechnology, 2012, Vol.12, Nr.10, pp. 8211–8214, (USA).

8. **PhD referee commission:**

Referee for two PhD theses at Politehnica University Timisoara.

9. **Expert evaluator for national research proposals:** expert CNCSIS, expert CEEEX, expert PN II

10. **Referee for ISI publications:** Heat and Mass Transfer, Micro Nano Letters, International Journal of Heat and Mass Transfer, International Journal of Thermal Sciences, Journal of Molecular Liquids, Experimental Thermal and Fluid Science

11. **Professional membership:**

American Nano Society, Society of the Automotive Engineers from Romania (SIAR, SAE affiliate), enrolled in 2011

Researcher ID: B-7397-2011 ISI Web of Knowledge

V. Scientific and academic future development plans

V.1. Scientific development plan

The development plan regarding my scientific activity, presents the potential directions to be followed.

(1) on short term: continuation of the ongoing project PN II 63/2014–2017, regarding the study of thermal conductivity and electrical conductivity of highly concentrated transformer oil based magnetic nanofluids;

(2) on short and medium term: the effect of magnetic fields on the thermal conductivity of magnetic nanofluids, complex magneto–rheological fluids, magnetic composite media;

(3) on medium to long term: study of the potential use of magnetic nanofluid and magnetic composite media, for thermal storage and heat transfer applications;

(4) on medium to long term: exploring the use of molecular dynamics to study the effect of the representative volume size on bulk thermodynamic properties.

1. Thermal conductivity and electrical properties of highly concentrated transformer oil based magnetic nanofluids

As mentioned, this theme is currently under work, only its first results have been obtained. The study of thermal conductivity of nanofluids is a hot topic in science for the last two decades, some progress has been made in order to clarify the heat transfer between the nanoparticles, the surfactant layers and carrier liquids but open problems still exist (Kanafer, Vafai, 2011; Nkurikiyimfura et al, 2013).

Magnetic nanofluids have a much longer history, and analyses of their properties have been made previously (Blums et al, 1997), however very high concentrated samples are less studied. Similarly, the electrical properties of magnetic nanofluids is an open research problem. During previous collaborations and projects, electrical properties of low volume fraction magnetic nanofluids have been measured in cooperation with the team from IEP–SAS (Slovakia) (Timko

et al, 2010; Timko et al, 2012; Stoian et al, 2013). The ongoing research is aiming at developing at RCSCF – UPT an experimental bench for such measurements, in view of future use for water based magnetic fluids, in connection with other potential applications.

2. Effect of magnetic fields on the thermal conductivity of magnetisable materials

This topic is in research for several years worldwide, still few experimental results exist (Nkurikiyimfura et al, 2013; Bahiraei, Hangi, 2015). The aim is, at first, to develop an experimental bench to study the effects of various types of magnetic fields that could be used for both liquid-like and solid materials, based on the existing infrastructure of measuring thermal conductivity and the magnetisable materials, that are prepared by the Laboratory of Magnetic Liquids, RA-TB, with whom we are having a long-term collaboration at RCESCF-UPT.

3. Study of the potential use of magnetic nanofluid and magnetic composite media, for heat transfer applications and thermal storage

In what it concerns the heat transfer applications, this area is envisaged to be developed based on the results obtained so far (Stoian et al, 2008; Stoian, Holotescu, 2012; Stoian, Holotescu, 2014), new applications will be explored such that low volumes of magnetic nanofluid are used, considering the problems related to produce large amount of stable fluid that keeps its stability on long term. At first, our recent results, regarding the use of water based magnetic nanofluids, will be further developed toward an experimental model apparatus in view of an engineering related application.

The problem of thermal storage is of crucial importance for solar concentrated power systems and low grade heat utilisation (Li et al, 2012; Chan et al, 2013). Thus, exploring the use of magnetisable media as thermal fluid in such systems is another direction considered for study.

4. Use of molecular dynamics to study the effect of the representative volume size on bulk thermal properties

The experience gained in the molecular simulation is planned to be used for developing a study regarding the problem of representative volume size for thermal properties. So far, several results regarding the molecular dynamics simulations are reported regarding the representative volume problem for mechanical properties of solid materials, especially composite media (Valavala et al, 2009; Huang, Pizhong, 2011; Zhuang et al, 2017).

V.2. Academic development plan

The development plan regarding the academic activity in general, the following paths, are considered:

- (1) continuing the integration of the research results, when possible, as part of the teaching subjects, especially for graduate students, as well as integration of undergraduate and graduate students in the candidate's research team;
- (2) continuing the development of the course materials and infrastructure, in such a manner to attract students toward the will of learning, academic study and research.

In view of pursuing as PhD adviser, I plan the followings:

- (1) integrating new PhD students in the well-established interdisciplinary research team working at RCESCF-UPT;
- (2) tutoring the new PhD students regarding the way of conduct an ethical research;
- (3) providing the new PhD students with the guidance based on the experience gained as an independent researcher, such that, they will be able to accomplish their research plan and thesis.

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ANNEX I:

List of 10 relevant scientific publications

1. **Stoian, F.D.**, Morimoto, H., Sato, T., and Maekawa, T. (2002). Cluster growth and structures of Lennard–Jones molecules near the critical point. *MICROSCALE THERMOPHYSICAL ENGINEERING* (in 2006 was changed to *Nanoscale and Microscale Thermophysical Engineering*). Vol 6(1), pp. 15–30, ISSN: 1556–7265, DOI:10.1080/108939502753428211 (ISI Article, FI 2015: 2.39)
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3. **Stoian F.D.**, Holotescu, S., Vekas, L., (2010). Bubbles generation mechanism in magnetic fluid and its control by an applied magnetic field, 12th International Conference on Magnetic Fluids (ICMF12), Japan, August 1–5, 2010, Book Series: *Physics Procedia*, 2010, Vol.9, pp. 216–220 (ISI Conference Paper).
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RELEVANT PAPERS

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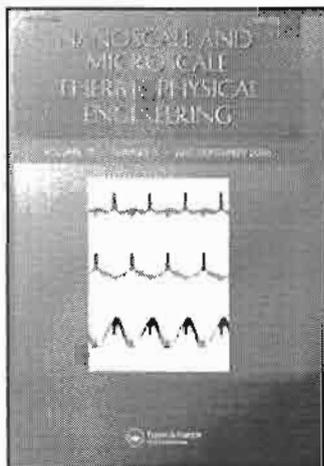
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CLUSTER GROWTH AND STRUCTURES OF LENNARD-JONES MOLECULES NEAR THE CRITICAL POINT

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CLUSTER GROWTH AND STRUCTURES OF LENNARD-JONES MOLECULES NEAR THE CRITICAL POINT

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We considered a two-dimensional system consisting of 529 molecules, interacting with each other via the full Lennard-Jones potential.

Using a constant NVT molecular dynamics simulation method, we studied the behavior of this system and analyzed the cluster growth process near the critical point. We considered several data sets near the critical point. By analyzing the cluster formation, we obtained a power law-like behavior for $N_s(t)$, the number of clusters, and $S(t)$, the average number of molecules in a cluster with respect to time.

We also carried out a steady-state analysis. Starting the molecular dynamics simulation in the canonical ensemble, after about 150 ps we changed the ensemble to micro-canonical for another approximately 300 ps. We calculated the following static properties: isometric heat capacity, isothermal and adiabatic compressibilities.

Beginning with the experimental discovery of the liquid–vapor critical point by Andrews in 1869, considerable efforts have been made to describe theoretically the phenomena at the critical point, of which we shall mention those of van der Waals (just four years after Andrews), Landau, Ising, Onsager, Widom, Kadanoff, and Wilson [1].

It is well known nowadays, both theoretically and experimentally, that the isometric heat capacity, c_v , shows a divergence and a jump at $T = T_c$ [2]. Also, the experiments carried out by Michels, for CO_2 [3], and recently by Pittman for He-3 [4], proved that the thermal conductivity, k , has a sharp increase at the critical point. While k increases when approaching the critical point, the thermal diffusivity, D_T , decreases due to the strong effect of increasing the isobaric heat capacity, c_p .

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A fundamental study regarding the control of nucleate boiling in a complex magnetizable fluid by an applied magnetic field, in microgravity conditions

By: [Stoian, FD](#) (Stoian, FD); [Pop, G](#) (Pop, G); [Bica, D](#) (Bica, D); [Stoica, V](#) (Stoica, V); [Marinica, O](#) (Marinica, O); [Vekas, L](#) (Vekas, L)

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Abstract

A new type of working fluid to be used for boiling or multiphase heat transfer in microgravity conditions is proposed. The advantages of using a complex magnetisable fluid for boiling or multiphase heat transfer in microgravity are presented, based on the theoretical analysis of the body forces exerted by an applied magnetic field in such a fluid, in comparison with the similar phenomena determined by the body forces exerted by an applied electric field in a dielectric fluid. The experimental set-up designed to study the effect-of the applied magnetic field on the bubble dynamics, to be proposed for a microgravity experiment, and currently in preparation for experimentation under terrestrial conditions, are presented.

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12th International Conference on Magnetic Fluids

Bubbles generation mechanism in magnetic fluid and its control by an applied magnetic field

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Abstract

A comparison of the effect of an applied magnetic field on the bubble departure diameter in a magnetic nanofluid, for different orientations of the magnetic field gradient with respect to gravity, is presented. Using the instantaneous relative pressure during the bubble growth and departure and the instantaneous gas flow rate measured for a certain range of magnetic field intensity, the dependence between the average bubbles injection frequency, average bubble volume and the applied magnetic field were studied. An effect of the injection hole size on the bubble frequency was observed.

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Keywords: bubble departure diameter; magnetic nanofluid; nucleate boiling; magnetic field.

1. Introduction

Heat dissipated in various types of devices and equipments that nowadays are getting more and more compact, surpasses the heat transfer performances of conventional cooling fluids. If we consider of combining the most promising passive method, that is the use of nanofluids as cooling fluid, with an active method – application of external fields, which can give the possibility of controlling the enhancement rate, than a solution could be offered by the magnetic nanofluids and an external magnetic field.

Boiling heat transfer is characterized by the highest heat transfer coefficients. Among the characteristic parameters of boiling, the average equivalent bubble diameter (assuming the bubble as a sphere, of an average size) is of major importance for the theoretical study of the nucleate boiling heat transfer but due to the parameters that influence the bubble formation and departure it is difficult to determine it theoretically [1]. Boiling of magnetic nanofluids was studied experimentally and theoretically, both from the point of view of enhancement rate and mechanisms [e.g. 2,3].

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The results of an experimental study regarding the bubble generation frequency and the bubble equivalent average diameter, as a function of the applied magnetic field, are presented in this work.

2. Theoretical background

The condition of bubble departure from the surface can be determined from the force balance on the vapor bubble during the process of growing. The simplest equation for the bubble departure diameter, D_r , includes the buoyancy and surface tension forces:

$$D_r = f(\beta) \left[\sigma / g(\rho_L - \rho_G) \right]^{1/2} \quad (1)$$

where $f(\beta)$ is the liquid to surface contact angle, σ – the surface tension, ρ_L – the liquid density, ρ_G – the vapor density, g – gravitational acceleration. This equation does not include the effects of inertia and drag forces, like more recent and complex ones [1]. In the case of boiling of a magnetic nanofluid in the presence of an external magnetic field, the magnetic body force that acts on the growing bubble must be added in the force balance:

$$\vec{F}_m = -\mu_0 V M(H) \cdot \nabla H \quad (2)$$

where μ_0 – free space permeability, V – bubble volume, M – magnetic fluid magnetization, H – magnetic field intensity. Thus, if the inertia and drag forces are neglected, the force balance results in [4]:

$$\vec{F}_b + \vec{F}_m = \vec{F}_s \quad (3)$$

when buoyancy force and magnetic force act in the same direction. In this case the bubble departure diameter is:

$$D_{r,m} = f(\beta) \sqrt{\sigma / (\rho_L - \rho_G) g \pm \mu_0 M \nabla H} \quad (4)$$

the sign is depending on the relative orientation of buoyancy force and magnetic force.

3. Experimental bench

The experimental bench, presented in Fig.1, was designed to study the phenomena related to the growth and departure of bubbles injected from a flat surface in magnetic fluid and the effects of an applied magnetic field. A non-uniform magnetic field is generated using profiled, V-shaped, electromagnetic poles, which give rise to a constant gradient of the magnetic field, ∇H , parallel with the gravitational field, \vec{g} , with the same orientation as \vec{g} , or opposite orientation to \vec{g} , if the V-shaped poles are switched up-side-down. Prior to actual experiments, the magnetic flux density was measured along the symmetry axis of the air gap, with a 10 mm step. The field measurement was carried out using a Hall probe (6) and Bell Gaussmeter (5). The resulted field gradient at the hole site (50 mm up from the bottom of the poles) was 79.6 kA/m².

The experimental cell (9) is a cylinder with glass walls ($\Phi_{\text{ext}} = 38$ mm and wall thickness 3 mm) and plexiglass bottom cap. The level of magnetic fluid in the cell was 55 mm. The injection hole was drilled in the center of the bottom cap. Three cells, with different hole diameters, d , of 0.3, 0.7 and 1.5 mm, were tested.

The magnetic nanofluid sample was a colloidal suspension of magnetite nanoparticles dispersed in hydrocarbon oil (TR-30), with density 1470 kg/m³. The magnetization of the fluid sample was measured using a VSM magnetometer (DMS/ADE Technology, USA), the saturation value being 44.747 kA/m.

Using the experimental procedure described in [5], we measured the instantaneous relative pressure during the bubble growth and departure, close to the injection hole, and the injected gas flow rate.

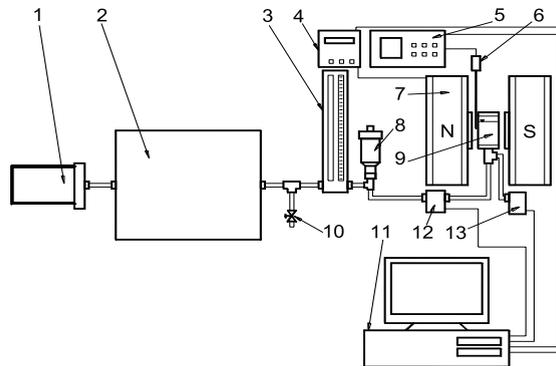


Fig.1: 1 – compressor; 2 –gas reservoir; 3 – manometer; 4 – magnets power supply; 5 -gaussmeter; 6 – Hall probe; 7 – magnetic poles; 8 - micrometer valve; 9 – experimental cell; 10 – flow rate valve; 11 – PC computer with data acquisition system; 12 - flow rate transducer; 13 – pressure transducer

The relative pressure was measured with a differential pressure transducer, accuracy $\pm 1\%$. A flow rate sensor, accuracy $\pm 1\%$, measured the injected gas flow rate. All data were recorded using a computer equipped with a NI DAQ system using the LabView software. By measuring the time period between two peaks we get the bubble emission period, τ_b , and then the bubble frequency, f . By averaging the value of the measured flow rate over the acquisition period, we get the average gas flow rate. The average bubble volume, V_b , and further, the equivalent bubble diameter (of an equivalent spherical bubble of volume equal to V_b), D_{ech} , are obtained.

4. Results and discussion

The experiments showed that the applied magnetic field is influencing the bubble generation frequency, as well as the average equivalent bubble diameter. For the injection hole $d = 1.5$ mm, the average frequency is increasing with the applied field when the field gradient has the same orientation as gravity and if the orientation is reversed, the frequency is decreasing. The values plotted in Fig.2 show the variation of f from approximately -25% to $+55\%$, taking as reference the bubble generation frequency at zero field. The dependences obtained for the average equivalent diameter in the same case are presented in Fig.3. For the investigated field range, D_{ech} ranged from -16% to $+12\%$ of the reference value at zero applied field. Results reported by Bashtovoi et al [6], using a magnetic nanofluid of similar composition and saturation magnetisation, and capillary tubes of 0.8 to 1.8 mm diameter, showed that by applying a much higher field gradient, of different values and about 10 times higher than in this experiment, a corresponding much higher variation of the bubble volume can be obtained.

However, if the injection hole diameter was decreased, the case of $d = 0.7$ mm and $d = 0.3$ mm, the bubble generation frequency followed the ascending trend only to a certain magnetic field, after which the bubble generation frequency started to decrease, as shown in Fig.4. A possible explanation could be that from a certain value of the applied field inertia and drag forces have to be accounted for, as the bubble departure diameter lowers. Also, the characteristic times of the phenomena may play a role. Nucleate boiling experiments of a kerosene based magnetic fluid [7] in uniform field showed a similar trend, by increasing the boiling fluid temperature and the applied magnetic field, the bubble generation frequency increases at first and then decreases.

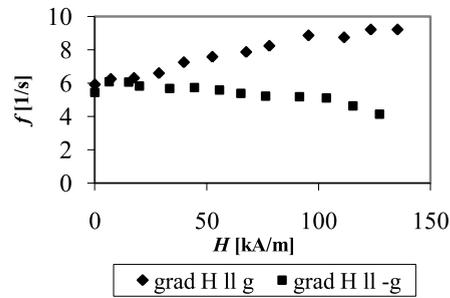


Fig.2. Average bubble generation frequency as a function of the applied magnetic field and its gradient orientation, $d = 1.5$ mm

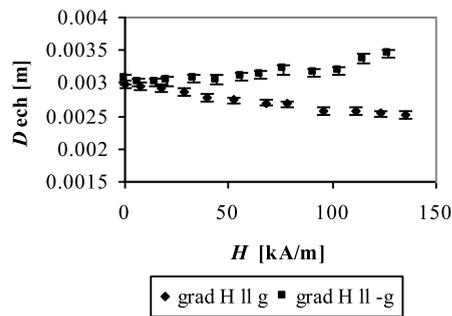


Fig.3. Average equivalent bubble diameter as a function of the applied magnetic field and its gradient orientation, $d = 1.5$ mm

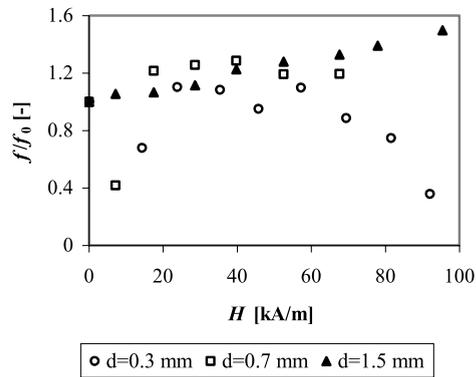


Fig.4. Bubble generation frequency as a function of the diameter of the injection hole, for grad H || g

5. Conclusions

The experimental results showed that the bubble departure diameter and bubble frequency are influenced by the magnetic field, according to the force balance accounting for surface tension, buoyancy and magnetic force. Also, it was observed that the injection hole size might affect the bubble frequency and, consequently the bubble departure diameter, in correlation with the applied field.

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Magnetizable colloids on strongly polar carriers – preparation and manifold characterization

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Abstract The aim of this paper is to present methods applied to obtain and to investigate the properties of magnetic fluids on strongly polar carrier liquids, such as short-chain-length alcohols and water. The high degree of colloidal stability of these colloids is achieved by double-layer sterical stabilization of Fe_2O_3 and CoFe_2O_4 nanoparticles dispersed in the carrier liquid, the adequate choice and efficient adsorption of the surfactant(s) used being one of the key problems to be solved in each particular case, in order to obtain high-quality magnetic fluids. The samples were investigated using a vibrating-sample magnetometer, a rheo/magnetorheometer with various measuring cells, as well as by small-angle neutron scattering, in order to detect magnetic-field-

induced changes at the nanometric level and qualitative differences in the macroscopic behaviour of various samples. The conclusions refer to the dependence of the colloidal stability on the composition and volumic concentration of the magnetic fluid samples, as well as on the applied magnetic field.

Keywords Magnetic nanoparticles · Magnetic fluids · Preparations · Properties

Introduction

Magnetic fluids are a special category of colloids consisting of magnetic nanoparticles, such as Fe_3O_4 , CoFe_2O_4 , Co or Fe, stably dispersed in a carrier liquid. Owing to their high degree of colloidal stability, achieved by specific stabilization procedures, these colloids behave macroscopically as a homogeneous magnetizable liquid, even in strong and nonuniform magnetic fields. Consequently, these kinds of colloids, usually called magnetic fluids or ferrofluids, are smart colloidal systems, which manifest simultaneously fluid and magnetic properties. Macroscopically, the introduction of a controllable magnetic force into the fundamental hydrodynamic

equations for the quasihomogeneous magnetizable liquid medium gives rise to the magnetohydrodynamics of magnetic fluids, known also as ferrohydrodynamics and opens an entire field of new phenomena [1, 2, 3]. From a microscopic point of view, the interactions between magnetic nanoparticles have to be adequately controlled in order to ensure the required high colloidal stability of the system in various operating conditions.

In the case of sterically stabilized nanoparticles in various liquid carriers, the type and quality of surfactants used will determine the efficiency of particle surface covering and, consequently, the balance between attractive and repulsive interactions between particles. The attractive interactions may lead to various types of

Comparative study of convective heat transfer in water and water based magnetizable nanofluid for thermal applications

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An experimental research carried out to investigate the convective heat transfer in a water based magnetizable nanofluid by natural convection is reported. The hot wall was a flat disk, heated from below. The open air played the role of the cold wall, as the upper surface of the fluid was free, at atmospheric pressure and temperature. Comparative tests were conducted using distilled water and water based magnetizable nanofluid. Several heating regimes were tested. The addition of nanoparticles in a usual heat transfer fluid as water led to a decrease in the heat transfer performance by natural convection.

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Keywords: Magnetizable nanofluid, Natural convection heat transfer, Nanoparticles

1. Introduction

The use of magnetic nanofluids (widely known as ferrofluids or magnetic fluids) – a special class of nanomaterials, as heat transfer fluids, had become a topic of intensive research starting about a decade ago [1-9]. The possible applications envisaged the use of phase-change heat transfer (boiling/condensation) or forced convection heat transfer modes. Both terrestrial and microgravity applications were taken into consideration due to the possibility to replace the gravitational field by the magnetic field in micro-g environments. Examples of such possible applications are: cooling systems for electric transformers, magnetocaloric heat pipes, pulsating heat transfer tubes and heat sinks. It should be noted also that, opposite to usual magnetic liquids prepared for their traditional applications (sensors, sealings etc), the magnetizable nanofluids for heat transfer should have a low concentration of magnetic nanoparticles in order to make them competitive with the non-magnetic nanofluids. On the other hand, an advantage of using magnetizable nanofluids, beside the possibility of control by an applied magnetic field, is the vast experience in preparation of such complex fluids gained over more than three decades [e.g. 11-14].

Recent results presented in the open literature state that among the possible mechanisms that may affect the convective heat transfer in nanofluids (similar to the case of magnetizable nanofluid in zero magnetic field) are the transport mechanisms associated to the relative (slip) velocity developed by the nanoparticles with respect to the carrier fluid, such as: inertia, Brownian diffusion, thermophoresis, Magnus effect, fluid drainage and gravity. A model proposed by Buongiorno [15] for convection

transport in nanofluids states that, when the turbulent transport of nanoparticles dominates, the particles are carried by turbulent eddies, while if the opposite case occurs, Brownian diffusion and thermophoresis may become important slip mechanisms. Also, several other mechanisms are related to the larger increase in the effective thermal conductivity for a very low volume fraction ($\Phi \leq 1\%$) of solid nanoparticles of less than 10 nm in size, like ballistic transport of energy carriers within nanoparticles, formation of nanoparticle structures through agglomeration (in fractal-like shape), clustering and networking [16].

Natural convection in usual heat transfer fluids was widely studied both experimentally and numerically, for various types of configurations. However, for various reasons (like low heat transfer coefficient, uniqueness of the problem in closed cavities configuration), in the case of nanofluids this heat transfer mode was less approached [16]. Our work has as final target a new type of heat exchanger for automotive thermal management applications using a magnetizable nanofluid as working fluid and an applied magnetic field to control the heat transfer process. The study reported here envisaged the evaluation of the natural convection heat transfer characteristics for a magnetizable nanofluid of low concentration in comparison with the carrier fluid, for a certain configuration of cavity that may be suitable to accomplish our target.

2. Experiment

The water-based magnetizable nanofluid tested in this work is part of a series of magnetizable nanofluids (water-based and hydrocarbon-based) specially prepared

and characterized within the framework of our research project [14,16]. The sample properties at 20 °C are as follows: density $\rho_{\text{MNF}} = 1082 \text{ kg/m}^3$, viscosity $\eta_{\text{MNF}} = 2.4 \cdot 10^{-4} \text{ Pa}\cdot\text{s}$, specific heat capacity $c_{p,\text{MNF}} = 3863.044 \text{ J/kgK}$, saturation magnetization $M_s = 50 \text{ Gs}$, volume fraction of magnetite (Fe_3O_4) nanoparticles $\Phi = 1.02 \%$. In Fig. 1 is shown the experimental setup used to analyze the natural convection heat transfer in a tube, heated from below.

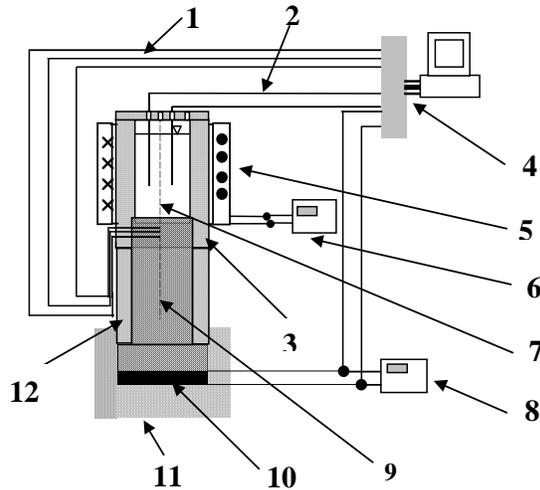


Fig. 1. The setup for the natural convection experiment.

The experimental cell (3) is a cylinder made of acrylic resin ($d_{\text{int}} = 16 \text{ mm}$, $H = 50 \text{ mm}$). The heating surface (9), made of aluminum ($d_{\text{surf}} = 18 \text{ mm}$) is placed at the bottom side of the cell. A nickel heater (10) of resistance $R = 0.14 \Omega$ powered by a power supply (8), is used to heat the surface. The temperature inside the magnetizable nanofluid (7) and in the heating surface was monitored by K-type thermocouples (1,2). The cell and aluminum bar were insulated by acrylic resin (12), while the resistance heater was placed in a ceramic insulating frame (11). The effect of an applied DC magnetic field generated by a coil (5) can be also studied using the same setup. The coil is powered by a power supply (8). The experimental data were collected using a data acquisition system NI 6042E (4), embedded in a PC using a dedicated LabView program. A SCXI 1000 amplifier was used to achieve high accuracy temperature measurements.

The experiments were carried out at constant heat flux. To determine the heat flux and the heated surface temperature, a set of three thermocouples were placed in the axis of the aluminum bar at known distances (3, 5 and 5 mm), close to the heated surface. The heat flux was estimated from the Fourier law, based on the temperature gradient between two thermocouples (T_{p1} mounted at 13 mm and T_{p3} at 3 mm, from surface):

$$\dot{Q} = \frac{\lambda_{Al}}{x} (T_{p1} - T_{p3}) \quad [W] \quad (1)$$

where $\lambda_{Al} = 235 \text{ W/m}\cdot\text{K}$ is the aluminum thermal

conductivity and $x = 10 \text{ mm}$ is the distance between the position of thermocouples in the axis of the bar.

The heated surface temperature was determined using the indication of the thermocouple placed nearest to the surface (at 3 mm) and the heat flux given by Eq. (1):

$$T_p = T_{p3} - \dot{q} \frac{\delta}{\lambda_{Al}} \quad [K] \quad (2)$$

where $\delta = 3 \text{ mm}$.

The average liquid temperature at stationary conditions, far from the surface (T_i), was determined from the readings of the set (2) of thermocouples in Fig. 1 (T_{i1} and T_{i2}):

$$T_i = (T_{i1} + T_{i2})/2 \quad (3)$$

The heat transfer coefficient, h , was determined based on the Newton's law:

$$h = \frac{\dot{q}}{(T_p - T_i)} \quad [W/m^2K] \quad (4)$$

where $\dot{q} = \dot{Q}/A$ and $A = \pi d_{\text{int}}^2/4$.

The experiments were run at first with distilled water, for several regimes of heat flux (heating of the surface) to test the experimental cell and compare the results with known empirical correlations. Then, there were repeated using the sample of water-based magnetizable nanofluid for the same heat flux conditions.

3. Results

The results obtained for the natural convection heat transfer using distilled water were compared with a known correlation from literature. In this type of heat transfer mode the deciding factor is the Rayleigh number:

$$Ra = Gr \cdot Pr = \frac{g\beta\Delta T l^3}{\nu^2} \cdot \frac{\nu}{\alpha} \quad (5)$$

where g is the gravitational constant, β is the volume expansion coefficient, l is the characteristic length ($l = 0.9d_{\text{int}}$), ν is the kinematic viscosity, α is the thermal diffusivity. There are a considerable number of empirical correlations in literature for natural convection heat transfer, for different types of geometries and fluids. Most of these have the form:

$$Nu = CRa^n \quad (6)$$

where Nu is the Nusselt number ($Nu = hl/\lambda$), C and n being constants, depending on the value of Ra .

In Fig. 2 is shown a comparison between the experimental results and a well known correlation for natural convection from a plate heated from below [17]:

$$Nu = 0.54Ra^{1/4} \quad (7)$$

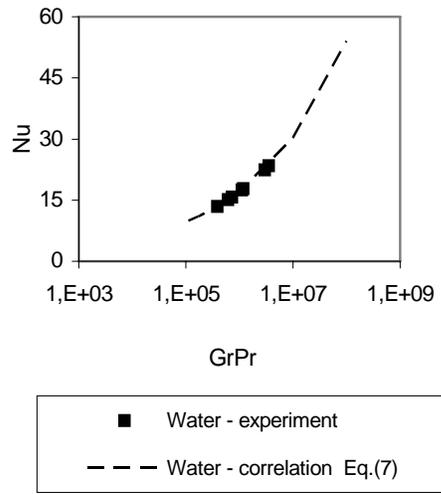


Fig. 2. Comparison of experimental results for distilled water with correlation (7).

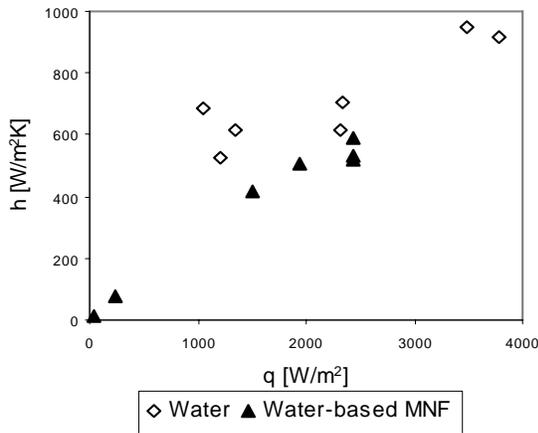


Fig. 3. Comparison of experimental results of water and magnetizable nanofluid at steady state conditions.

One can observe that the present experimental results are well described by Eq. (7).

In Fig. 3 are shown the experimental results for water and the magnetizable nanofluid sample at steady state. It was found that, for the same input of heat flux, the heat transfer coefficient of the magnetizable nanofluid is lower in comparison with that of water. Thus, the effect of addition of solid metallic nanoparticles in water is negative for natural convection heat transfer, similar to other experimental results using non-magnetic water-based nanofluids (Ti-O2 nanofluids, CuO nanofluids) [18,19].

4. Discussion

The analysis of our experimental results, using measured or estimated values of the thermophysical

properties of the magnetizable nanofluid and literature data for water to calculate the Grashof and Prandtl numbers show an interesting behavior, for the same average temperature:

$$T_{med} = (T_l + T_p) / 2 \tag{8}$$

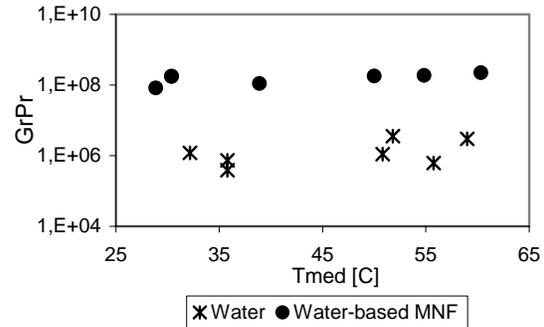


Fig. 4. The influence of the thermophysical properties of the magnetizable nanofluid and water on Gr·Pr.

As shown in Fig. 4, the value of Gr·Pr is in average 10³ times higher for the magnetizable nanofluid but, also due to a higher thermal conductivity of the magnetizable nanofluid, the value of Nusselt number is lower than that for water. It means that in order to obtain a value of the natural convection heat transfer coefficient for the magnetizable nanofluid similar to that of water, in the same working conditions, the turbulence should be further increased. That is, what actually “makes the difference” (influence the convective heat transport) in nanofluids are their thermophysical properties.

The application of a time-varying (e.g. rotating) magnetic field in the case of a nanofluid with particles having rigid dipole moment might give a solution to the problem.

5. Conclusions

The goal of the present research is the development of a new type of heat exchanger for automotive thermal management system using a magnetizable nanofluid as heat transfer fluid of low concentration, specially prepared for this application. This paper presents the results of the evaluation in the case of natural convection heat transfer.

We have tested the heat transfer performance of a magnetizable nanofluid ($\Phi = 1.019\%$) for a certain range of heat flux density and surface average temperature that are of interest for our case and using a configuration suitable for this purpose.

In the explored range of heat flux density, we obtained lower values of the heat transfer coefficient for the magnetizable nanofluid than those for the carrier fluid (water), the relative difference between the corresponding coefficients ranging from 4 to 16%. These results agree with other experimental data for non-magnetic water-based nanofluids, in the case of natural convection heat transfer

mode [18,19].

The influence of the addition of nanoparticles in the carrier fluid on the thermophysical properties reflected in the values of the characteristic numbers Grashof and Prandtl, for the same average temperature of the nanofluid and carrier fluid, is determined.

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Experimental Study of Natural Convection Enhancement Using a Fe3O4-Water Based Magnetic Nanofluid

By: Stoian, FD (Stoian, Floriana D.)^[1]; Holotescu, S (Holotescu, Sorin)^[1]

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Abstract

The effect of nanoparticles dispersed in a carrier fluid on the natural convection heat transfer is still raising controversies. While the reported experimental results show no improvement or even worsening of the heat transfer performance of nanofluids, the numerical simulations show an increase of the heat transfer coefficient, at least for certain ranges of Ra number. We report an experimental investigation regarding the natural convection heat transfer performance of a Fe3O4-water based nanofluid, in a cylindrical enclosure. The fluid was heated linearly from the bottom wall using an electric heater and cooled from the upper wall by a constant flow of water, such that a constant temperature difference between the upper and bottom walls was obtained at steady-state. The experiment was also carried out using water, in order to observe the effect of the addition of Fe3O4 nanoparticles on the heat transfer coefficient. Several regimes were tested, both for water and nanofluid. The experimental results showed that values obtained for the heat transfer coefficient for Fe3O4-water nanofluid were higher than those for water, at the same temperature difference. The present experimental results are also compared with our previous work and the reference literature.

Keywords

Author Keywords: Natural Convection; Magnetic Nanofluid; Heat Transfer Coefficient

KeyWords Plus: HEAT-TRANSFER; FLUID; FIELD; ENCLOSURE; FLOWS; WATER

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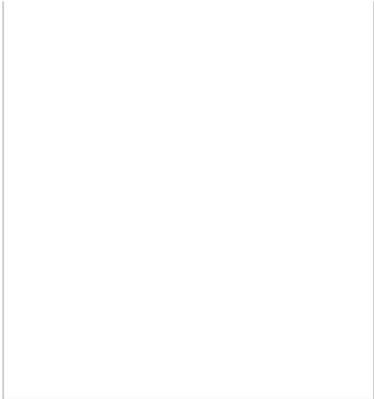
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A Novel, Ferrofluid-Cooled Transformer. Electromagnetic Field and Heat Transfer by Numerical Simulation.

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Abstract—A ferrofluid-cooled low power, single-phased electric transformer was designed and prototyped with the aim of investigating the performance that such an apparatus may exhibit. The nanometric, colloidal, super-paramagnetic fluid used as coolant has specific electric, magnetic, and thermal properties, and presents an overall better stability and capacity to withstanding electromagnetic and thermal stress. This paper addresses also the electromagnetic and heat transfer processes that occur. First, the physical, mathematical, and numerical models are introduced. Numerical simulation results suggest that the magnetization body forces may add to the thermal, buoyancy body forces in providing for better heat transfer. To outline this, several numerical models that may conveniently be treated numerically within the current hardware and software limits, while still providing for satisfactory accuracy were developed. The results may be utilized also in the design phase of the transformer.

I. INTRODUCTION

Ferrofluids are utilized in a number of important industrial applications: “silent” coils (for noise reduction), sensors, (e.g., accelerometers, flowmeters, sensors for inclination, pressure, level, etc.), sealing systems, different types of contacts [1], etc. Recently, it is observed a growing interest in their usage in high power devices for electrical engineering, in particular transformers with colloidal ferrofluids. Magnetizable nanofluids that are usable in heat transfer applications, as an alternative to the regular nonmagnetic fluids, present a lower concentration of magnetic nanoparticles.

The ferrofluids possess heat transfer and dielectric properties that are superior to their classical counterparts (e.g., transformer oils), and may be utilized for improving the heat flow within the aggregate active part contributing thus to increasing their withstanding capacity to faults such as electromagnetic impulses. The ferrofluids may be driven by external magnetic fields, and the magnetization

body forces within the ferrofluids may be controlled by adequately adjusting the incident magnetic field [2–7].

Finer details, such as local spectra of electromagnetic field, flow and heat transfer, and the system’s response (the working ferrofluid) to changes in the incident magnetic field and, or the heat transfer rate are sometimes discernable by numerical simulations only [2], [5], [7].

This paper presents the design of an experimental, low power, single-phased transformer, cooled by natural convection, for medium voltage installations that uses ferrofluid in lieu of the standard oil. The ferrofluid was designed at the Laboratory for Magnetic Fluids at the Timișoara branch of the Romanian Academy (LMF-TB-RA), investigated by LMF-TB-RA and NRCSF-UPT, and produced at ROSEAL. The transformer prototype was designed and fabricated by ICPE-CA and ICMET. The physical, mathematical, and numerical models developed a University POLITEHNICA of Bucharest (UPB) were aimed at investigating the heat transfer. The numerical simulation results evidence the heat transfer processes that occur under the combined action of the thermal, buoyancy forces and the magnetization body forces that occur in the ferrofluid used for cooling the transformer. They provide valuable information for the electromagnetic and thermal design of the transformer.

II. THE FERROFLUID

Ferrofluids have been shown to provide higher capacity to sustain overvoltage, and present better resilience at degradation in time due to the humidity as compared to classic oils. Previous work [3] proves that replacing the transformer oil with a magnetic nanofluid (transformer oil sustained) may be beneficial to the thermal and dielectric performance of the transformer.

The magnetic nanofluid prepared for experiments is a colloidal dispersion of magnetite (Fe_3O_4) nanoparticles in transformer oil TR-40 with oleic acid as surfactant. Laboratory investigations were conducted to obtain a magnetic fluid having surfactant magnetic nanoparticles



Utilization of the magnetogrulometric analysis to estimate the thermal conductivity of magnetic fluids

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ABSTRACT

In this study, the semi-empirical equation for the effective thermal conductivity of the Holotescu–Stoian model was applied to a set of four dilutions of a transformer oil based magnetic fluid with magnetite nanoparticles as magnetic phase, using the results obtained for the size distributions from the magnetogrulometry analysis, followed by a comparison with the measured values of the effective thermal conductivity obtained by the hot ball method. The link between the size distribution by number and by volume used in the magnetogrulometry analysis and the Holotescu–Stoian model adaptation to the lognormal distribution were presented. The comparison between the results given by the model and the corresponding experimental data showed that by using the approximated size distribution to calculate the effective thermal conductivity the analytical results much closer to the experimental ones are obtained, compared to those given by the Maxwell classical model.

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1. Introduction

One of the main parameters that influence heat transfer in various technical applications is the thermal conductivity. Usually, if cooling of a system is needed, the conventional solution is to use liquid like water, ethylene glycol, mineral or synthetic oils and refrigerants. Colloidal suspensions of metallic or non-metallic nanoparticles in the aforementioned liquids named “nanofluids” have higher thermal conductivity compared to that of the carrier liquids, an advantage that makes them a potential replacement for usual cooling fluids.

The use of nanofluids based on transformer oil and stably dispersed metallic nanoparticles as coolant of the power transformers requires the necessity to maintain their insulating properties in allowable limits [1,2]. This requirement limits the possibility to increase the nanofluid effective thermal conductivity by increasing the volume fraction of metallic nanoparticles, so that solutions to enhance the heat transfer for a specified volume fraction need to be found.

Both theoretical and experimental results proved that the size distribution of nanoparticles does influence the effective thermal conductivity of the nanofluid [3–5]. The mechanisms involved are not yet fully elucidated, most probably due to lack of a unitary way to characterize and report the size distributions.

In fact, the size distribution is an approximation that includes simplifying assumptions regarding the shape of the particles that determine the description of the particle using a single descriptor, the equivalent diameter. This leads to major differences between the results obtained using various methods to determine the size distribution, which refer to equivalent diameters specific to each measuring method.

The size distribution analysis of ferrofluids, using various accepted methods (magnetogrulometry, X-ray, electron microscopy, phonon correlation spectroscopy, torsional-pendulum method, etc.), showed also that certain differences exist between the results of various methods [6], even in the case of using the same method [7]. The magnetogrulometric method is based on the analysis of the magnetization curves and the choice of a type of size distribution of nanoparticles. The lognormal [8–11] and gamma [12–15] mono-modal size distributions are the most used ones due to their simplicity and because these depend only on two parameters that can be simply determined using the asymptotic behavior of the magnetization curve.

In the first part of the paper we present the simplest version of the magnetogrulometric method, based on the Langevin behavior of the magnetic fluid (valid for low volume fractions), and outline that depending on the way the lognormal size distribution is interpreted as being by volume [8] or by number [9], different results are to be obtained. This fact requires the necessity of details when results are reported so it can be converted according to a certain case study.

A large number of analytical models for evaluating the effective properties of colloidal suspensions are available, such as mixture

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Characteristic Properties of a Magnetic Nanofluid Used as Cooling and Insulating Medium in a Power Transformer

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Abstract- Magnetic nanofluids (widely known as ferrofluids or magnetic liquids) have a unique property – they are responsive to the application of a magnetic field, which allows for the possibility of controlling the flow and the convective heat transfer. This paper presents the characteristic thermo-physical, magnetic and dielectric properties of a transformer oil based magnetic nanofluid, specially prepared for use as a cooling and insulating medium in a power transformer.

Keywords: power transformer, cooling, insulation, magnetic nanofluid, saturation magnetization, electric permittivity, thermal properties

I. INTRODUCTION

The insulation fluids in power transformers perform two main functions: insulation, to prevent the flow of electric current between conductive components, and cooling, to transfer the heat out of transformer windings and core. The heat transfer rate from the transformer components to its walls and its subsequent dissipation into surroundings are factors that limit the current density in the windings and define its size and weight for a given power rating. Well purified mineral oils (e.g. transformer oil) are characterized by low thermal conductivity, so their efficiency as cooling agent is limited. Therefore, finding an insulating and cooling medium with enhanced electrical and thermal properties, leading to more efficient and environmentally safer power transformers for both energy distribution and industrial applications, has been a goal of the research in the field over the last decade [1-4]. Thus, synthetic oils, mixtures of mineral and synthetic oils were developed for replacing the traditional transformer oil [3-6]. At the same time, during the last decade a new type of coolant for power transformers was proposed and tested: the transformer oil based magnetic nanofluid (MF_UTR) [1, 5]. The unique feature of MF_UTR over any other type of coolant is that is responsive to the magnetic field. This characteristic, that enables the magnetic field lines

to close inside the transformer due to the presence of the dispersed magnetic nanoparticles, can improve the electric performances. In the case when a MF_UTR is used, both thermo-physical properties and heat transfer coefficient can be influenced by an applied magnetic field, such that an active control of the heat transferred in an equipment can be obtained, if the laws of dependence between each property and the applied magnetic field are known. Therefore, the study of their thermo-physical, magnetic and electrical properties have to be carried out both with respect to the temperature dependence and the effect of the applied magnetic field (AC/DC, magnitude, AC field frequency). Moreover, knowledge of the laws of variation for the thermo-physical, magnetic and electrical properties with respect to the volume fraction, magnetic field magnitude and working temperature, offers the premises of a realistic elaboration and validation of a thermal model for a transformer, using this nanofluid as cooling and insulating medium. In this paper are presented the characteristic magnetic, rheological, thermal and electrical properties of a transformer oil based magnetic nanofluid, prepared especially for use as insulating and cooling medium in a power transformer.

II. PREPARATION OF THE NANOFUID SAMPLE

A MNF sample was prepared, based on transformer oil UTR 40 and magnetite (Fe₃O₄) nanoparticles, stabilized with a hydrophobic single layer of OA (oleic acid). The synthesis procedure used the co-precipitation method developed by D. Bica, described in detail in [7, 8].

Density measurements were carried out using an Anton Paar DMA 35N density meter, at room temperature, 25°C. The results were $\rho_{UTR} = 0.867 \text{ g/cm}^3$ and $\rho_{MNF} = 0.937 \text{ g/cm}^3$. The solid volume fraction of the magnetite nanoparticles, φ , dispersed in the carrier liquid was calculated using

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Experimental study of cooling enhancement using a Fe_3O_4 magnetic nanofluid, in an applied magnetic field

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Abstract. This paper presents the results of an experimental study that envisaged the evaluation of the cooling capabilities of a transformer oil based magnetic nanofluid with the solid volume fraction of magnetite nanoparticles equal to 0.0162, in an AC applied magnetic field ($f = 50$ Hz). The heating and cooling regimes of a coil immersed in the magnetic nanofluid were compared to that corresponding to the base fluid (transformer oil). The results of our study indicate that the temperature rise rate of the magnetic nanofluid is lower than that corresponding to the transformer oil and a lower stationary temperature is obtained in the coil core, where the magnetic flux density is the largest.

1. Introduction

Researches regarding the use of nanofluids in thermal applications carried out worldwide are based primarily on the findings that their effective thermal conductivity is enhanced relatively to that of the carrier liquid [1,2]. This offer in turn the possibility to obtain a larger heat transfer coefficient in those devices and equipments where the heat transfer fluid is replaced with an application-compatible nanofluid. The magnetic nanofluids (widely known as ferrofluids or magnetic liquids) have an additional property – they are responsive to the application of a magnetic field, which allows for the possibility of controlling the flow and the convective heat transfer [3-5]. Magnetic nanofluids heat transfer related research is ranging from heat removal in magneto-fluidic devices for maintaining the desired magnetic properties, to their use as heat transfer fluids [3-7]. A field of research that gained promising results up to date is that of cooling and insulation of power transformers and other types of electromagnetic devices using magnetic nanofluids [7-12]. In this case, the magnetic field generated by the windings of a power transformer is used also to generate the flow of the magnetic nanofluid (the magneto-convection), thus enhancing the heat removal from the windings [11,12].

The aim of this work was to evaluate the cooling potential of a magnetic nanofluid for use as cooling medium in a power transformer. To accomplish this, the cooling of a coil powered by a 50 Hz AC power supply using a specially prepared Fe_3O_4 transformer oil based magnetic nanofluid [13], was investigated experimentally.



2. Materials

A magnetic nanofluid (MNF) sample and its carrier fluid – the transformer oil (TO-40A, producer MOL) were used in this experimental study. The MNF consisted of a stable colloidal dispersion of magnetite (Fe_3O_4) nanoparticles in TO-40A, with oleic acid (Vegetable oleic acid, 65-88 %, Merk) as surfactant, and was prepared using an established procedure developed by Bica et al [5,14].

The volume fraction of magnetite nanoparticles was determined based on the measured values of the transformer oil density and magnetic nanofluid density (indicated in table 1) and magnetite density, taken equal to 5180 kg m^{-3} . The resulting volume fraction is $\Phi = 1.62 \%$. The details of the preparation procedure and characterization of the magnetic, electric, rheological and thermal properties of the MNF are given in [13]. A summary of the characteristic properties for the MNF and TO-40A, at $25 \text{ }^\circ\text{C}$, is given in Table 1.

Table 1. Physical properties.

	ρ kg m^{-3}	η mPa s	k W m K^{-1}	c_p $\text{J kg}^{-1} \text{K}^{-1}$	β K^{-1}	ϵ F m^{-1}	M_s A m^{-1}
Magnetic nanofluid (MNF)	937	18.5	0.133	1743.39	$6.62 \cdot 10^{-4}$	$2.25 \epsilon_0$	2785.3
Transformer oil (TO-40A)	867	17.5	0.127	1915.12	$7.15 \cdot 10^{-4}$	$2.2 \epsilon_0$	-

The properties in table 1 are as follows: ρ is the density, η –dynamic viscosity, k – thermal conductivity, c_p - heat capacity, β – thermal expansion coefficient, ϵ – dielectric constant, M_s - saturation magnetization.

3. Experimental Setup

The cooling potential of the fluids was tested in an experimental setup presented in figure 1. A coil (4) of $R = 680 \Omega$ was placed in the center of the measurement cell – a cubic vessel (1) with Plexiglas walls of size $60 \times 60 \times 60 \text{ mm}^3$. The coil has the windings disposed on a rectangular plastic housing (3) and the internal air core dimensions are $15 \text{ mm} \times 15 \text{ mm} \times 18 \text{ mm}$. The coil housing is set on four legs on the bottom of the cell such that the fluid can circulate from the coil core to the outside bulk. The test fluid (2) was consecutively, TO-40A and MNF. The same volume of fluid, $V_f = 130 \text{ cm}^3$ was used. The coil was powered by a 50 Hz AC power supply (7), the working voltage being set at 40 V. Six temperature measurement points were established from the external side of the cell wall to the center of the coil core, as follows: the temperature of the external side (T_1) and of the internal side (T_2) of the measurement cell wall, the temperature on the external wall (T_4) and internal wall (T_5) of the coil, the temperature in the fluid (T_3) at the middle point between T_2 and T_5 , and the temperature in the fluid in the central axis of the coil core (T_6). All measurement points are in the same horizontal plan. The six T – type thermocouples (Omega GmbH, 0.5 mm, Teflon insulated, standard error of $\pm 1^\circ\text{C}$ or $\pm 0.7\%$) were introduced through plastic tubes inserted in a foam lid, placed on top of the measurement cell (not drawn in the schematic, for simplification reasons). Each thermocouple was connected through a NI SCC TC-01 module to a data acquisition system (6) consisting of a NI DAQ PCMCIA 6062E card and the NI SC 2345 connector block. The acquisition system was connected to a laptop computer (8) and the temperature measurements were recorded using the software package LabView 8.6. The sampling rate was 1 sample/s.

The coil magnetic flux density, B , was measured with a FW Bell 5080 Gaussmeter, before the heat transfer experiments. Using the axial and transversal Hall probes, the magnetic flux density was measured in several point of interest in the measurement cell for the applied voltage, as follows: the middle of the coil core axis (A), the top center of the core coil (B), the point situated 15 mm above the

top center of the coil (C) and the point situated 15 mm above the top of the core corner (D), as indicated in figure 2. The obtained values are listed in Table 2.

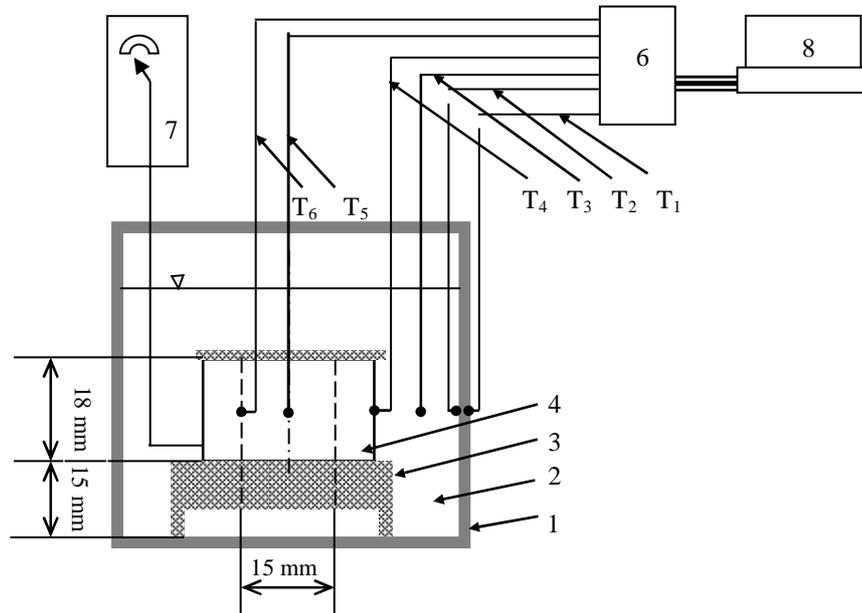


Figure 1. Experimental setup

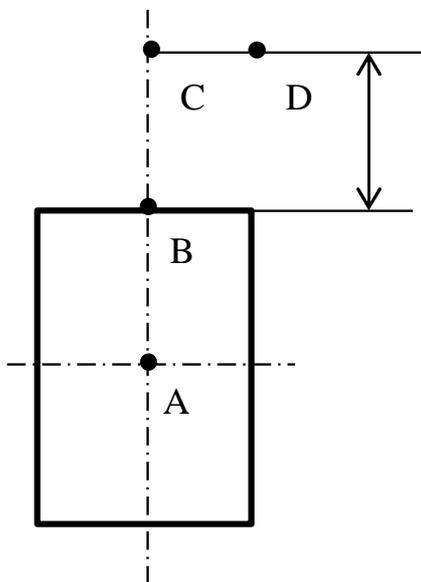


Table 2. Measured magnetic flux density values.

Point	B [mT]
A	8.1
B	4.9
C	1.4
D	1.0

Figure 2. Indication of the magnetic flux density measurement points in the coil core and above it

Using Vizimag software for visualising the magnetic field with the characteristics of the coil and measured magnetic field data for calibration, it was obtained a model for the magnetic field lines and

the magnetic flux density contours of the coil, which are presented in figure 3. It can be observed that the effect of the magnetic field generated by the coil is limited to the coil core and its vicinity.

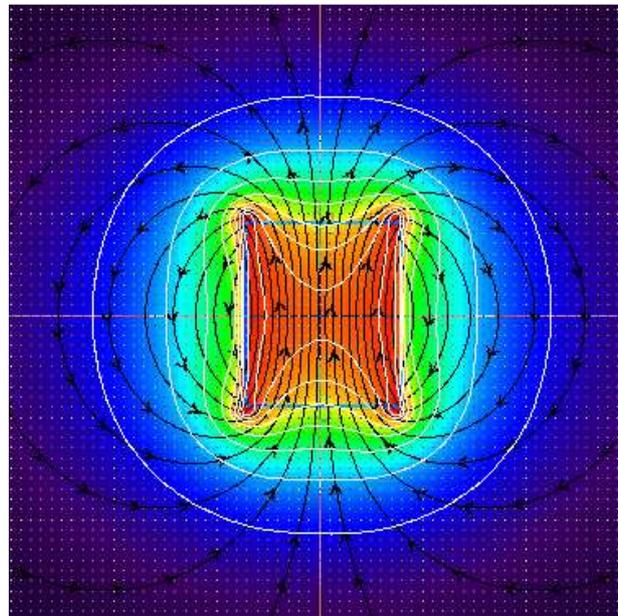


Figure 3. Model of the coil magnetic field lines (black lines with arrows) and the flux density contours (white lines)

4. Results and Discussion

The measurement session for each fluid was divided into three periods: (I) – a non-stationary heating regime characterised by a temperature rise rate, (II) – a stationary heating regime, and (III) cooling regime characterised by a temperature decrease rate.

In each experiment, the temperatures measured by the six thermocouples were continuously recorded by the data acquisition system, starting from the room temperature (28 ± 1.0 °C) and until the end of the cooling regime. During the measurement session, the start of the stationary heating regime was established as the beginning of the period when the temperatures varied less than the standard error (that is less than ± 1.0 °C). The stationary heating regime was attained after about 130 minutes from the start of the experiment. After a stationary period of at least 10 minutes, the power supply was turned off and the experimental system entered the cooling regime until it returned at the initial temperature.

The results of the temperature measurements were compared for the two tested fluids and are presented in figures 4 to 7. Figures 4 and 5 show that, when comparing the stationary values of temperatures $T_1 - T_6$ for the two test fluids, the main differences are obtained for T_5 - the temperature measured in the coil core. The average values of the measured temperatures in the stationary regime are presented in table 3.

Table 3. Average temperatures at the measurement points, in the stationary heating regime.

Working fluid	$T_{1,avg}$ [°C]	$T_{2,avg}$ [°C]	$T_{3,avg}$ [°C]	$T_{4,avg}$ [°C]	$T_{5,avg}$ [°C]	$T_{6,avg}$ [°C]
TO-40A	35.7	36.7	39.4	39.8	40.6	39.4
MNF	35.3	37.6	39.1	40.4	37.2	40.7

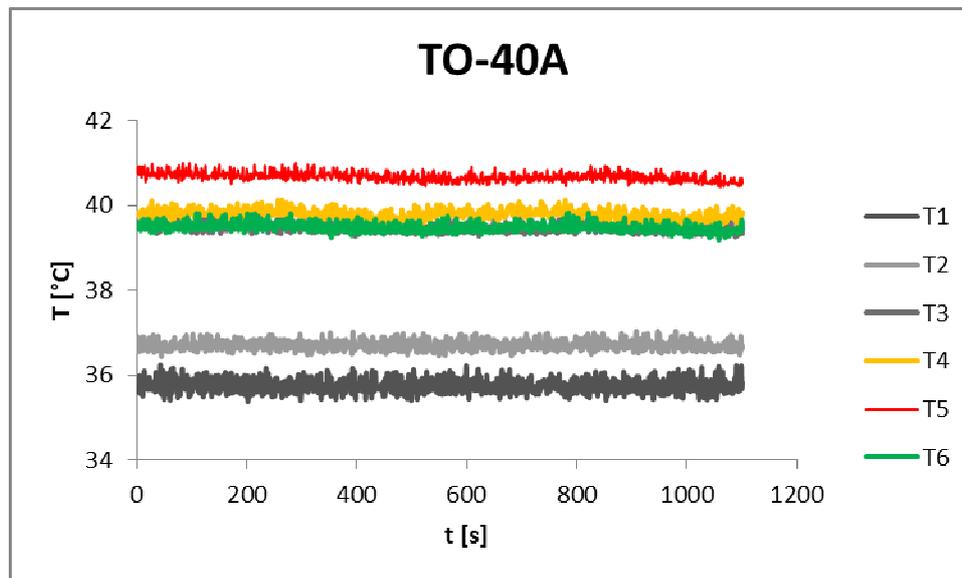


Figure 4. Stationary heating regime for the transformer oil

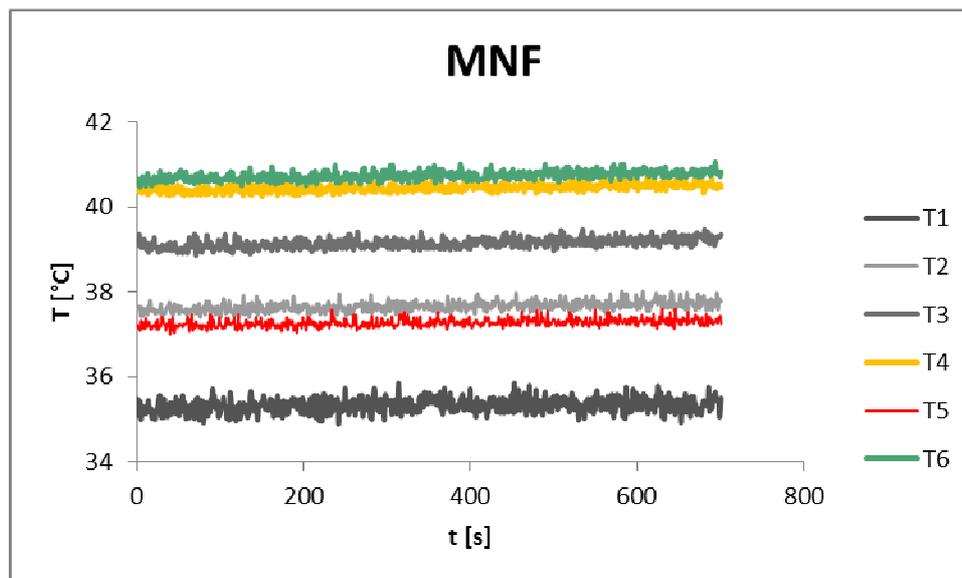


Figure 5. Stationary heating regime for the magnetic nanofluid

The coil external and internal walls temperatures (T_4 and T_6) are slightly higher in the case of magnetic nanofluid test. A possible explanation might be the adherence of some nanoparticles at the coil walls. Due to the low value of the magnetite volume fraction and the low magnetic field outside the coil, there is no significant difference between the corresponding average temperatures in the fluid outside the coil ($T_{3,avg}$). The only clear difference for the temperature measured in the fluid was inside the coil core ($T_{5,avg}$). Here, a difference of 3.4 °C in the corresponding values was obtained. Thus, the effect of the magneto-convection could be observed only in the coil core region, characterised by larger values of the magnetic field density (as indicated by measurements and simulation).

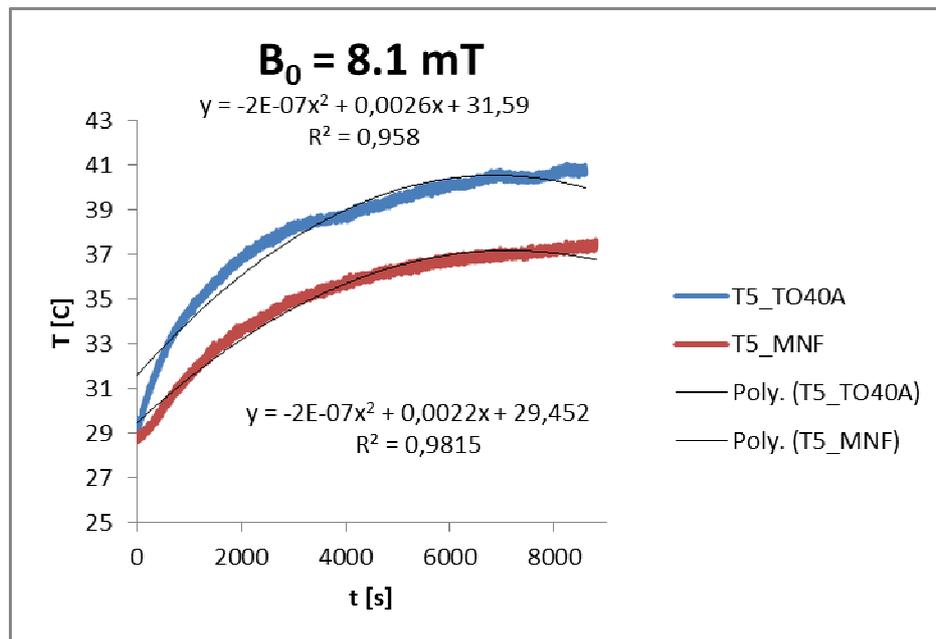


Figure 6. Comparison of heating curves in the central section of the coil core

To study further the heat transfer inside the coil core, the heating curves for TO-40A and MNF at the temperature measurement point T_5 (axis of the coil core) and their polynomial trend lines are compared in figure 6. The magnetic flux density at the measurement point is the one corresponding to the core centre (point A in figure 2). The results of the cooling process using both test fluids at the same point (T_5) are compared in figure 7. The temperature rise rate was calculated from each trend line equation displayed in figure 6 and the temperature decrease rate was calculated from the trend line equations displayed in figure 7. The obtained results are presented as a function of temperature in figure 8. The temperature rise rate of MNF for the explored temperature range is lower than that of the TO-40A. Also, the stationary regime of MNF it is approached at a lower temperature compared to TO-40A case, as observed from figure 5. This result is due to the magneto-convection, a combined effect of the addition of nanoparticles dispersed in the transformer oil, the temperature gradient inside the nanofluid and the applied external field.

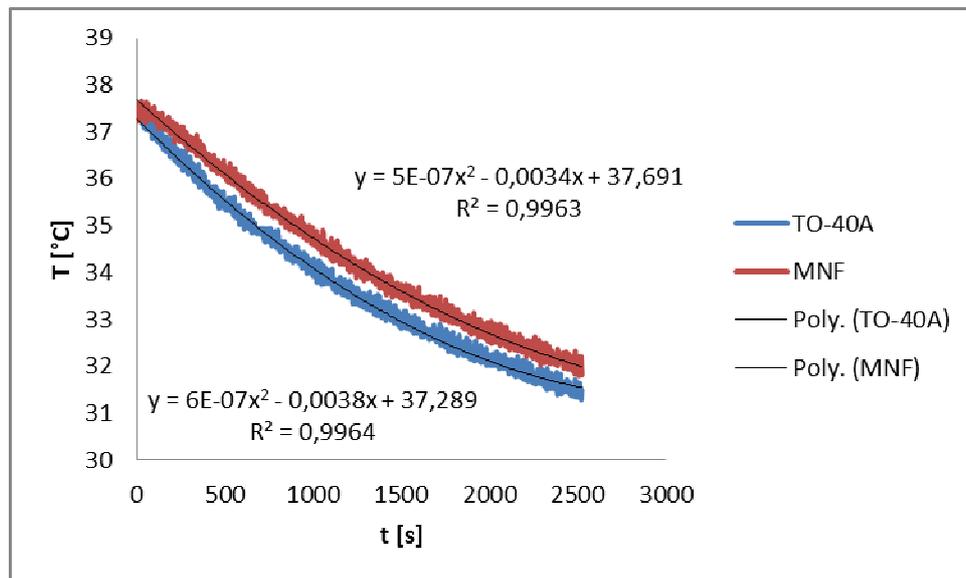


Figure 7. Comparison of the cooling curves in the central section of the coil core (with the coil being powered off)

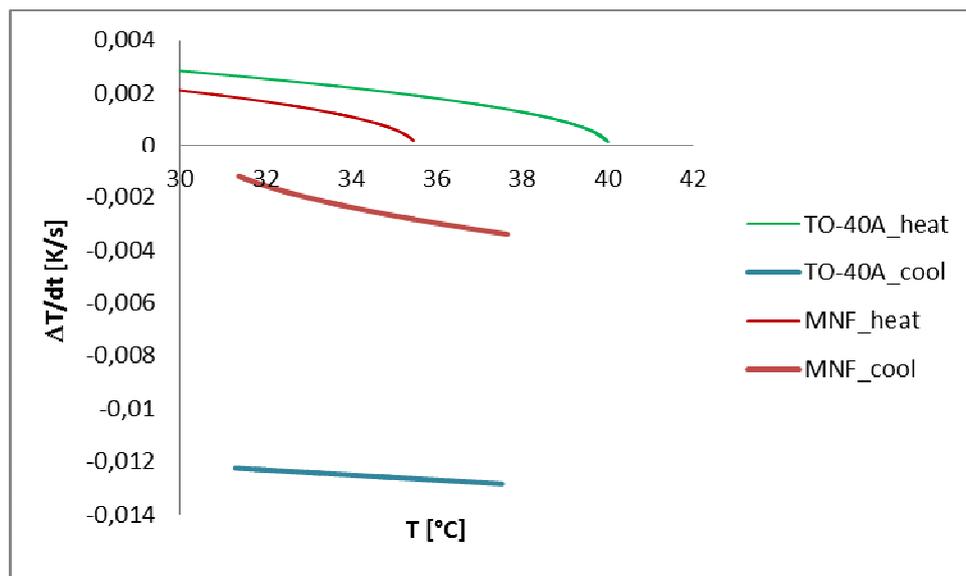


Figure 8. Temperature rise rate (subscript -“heat”) and temperature decrease rate (subscript – “cool”) in transformer oil and magnetic nanofluid

The MNF temperature decrease rate for the explored temperature range is larger than that corresponding to the TO-40A. That is, due to higher viscosity of magnetic nanofluid compared to that of transformer oil and the absence of the magnetic field, MNF has a lower heat transfer rate.

5. Conclusions

The cooling of an air cored coil powered by an AC source ($f = 50$ Hz) with magnetic nanofluid and, for comparison, with transformer oil (its base fluid) have been investigated. The obtained results indicated that the magneto-convection inside the coil core led to a decrease of the temperature rise rate and of the stationary temperature at the related measurement point (T_5). If the magnetic field is low or it is off, the heating and cooling temperature data are similar. The tested magnetic nanofluid has potential to be used as cooling medium in a power transformer, if magneto-convection is generated by the transformer windings magnetic field in the bulk of the fluid. The experimental results will be used as reference for a numerical study dedicated to the exploration of other geometrical configurations for the experimental cell in order to increase the magneto-convection outside the coil.

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