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Seminar:
Redactarea lucrărilor ştiinţifice:
Partea experimentală
Rezultate şi discuţii

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Facultatea de Chimie Industrială şi Ingineria Mediului
Laborator: Electrochimie, Coroziune şi Inginerie Electrochimică

Cercetarea ştiinţifică

- activitate prin care se dobândesc informaţii şi cunoştinţe despre univers

Funcţiile cercetării ştiinţifice:

- cognitivă
- formativă
- informativă

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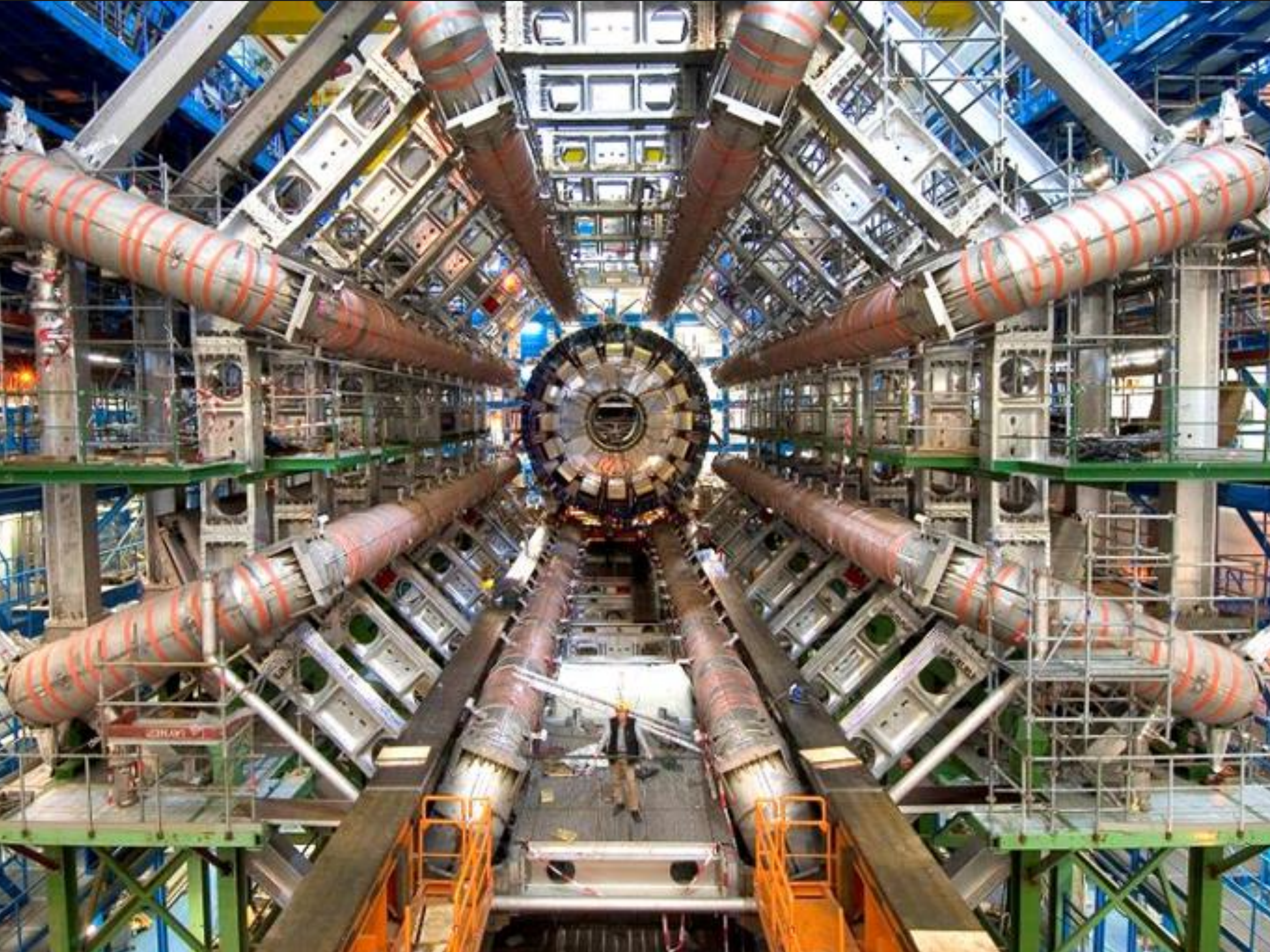
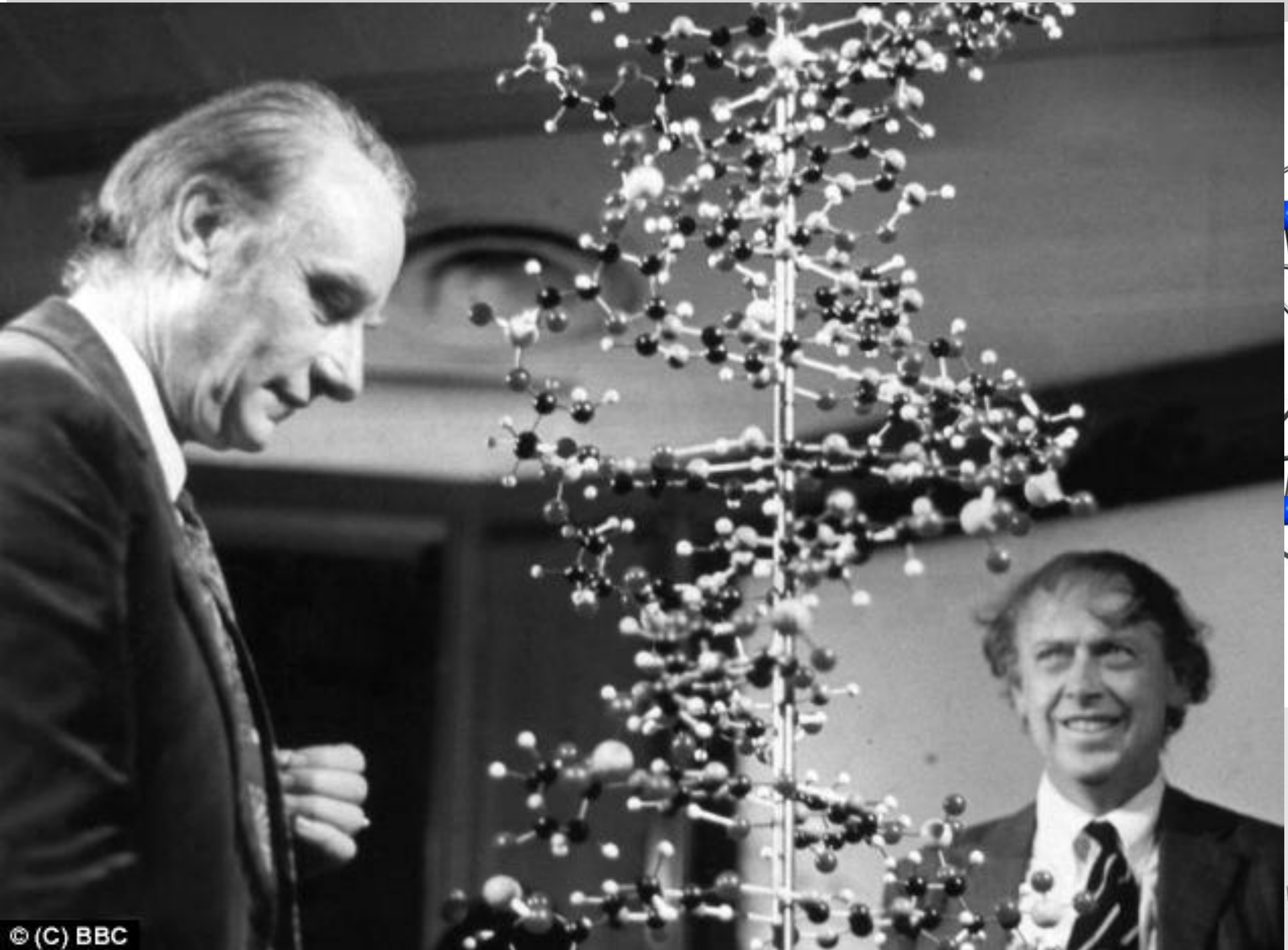
Clasificare:
Cercetare ştiinţifică fundamentală:
Cercetare ştiinţifică aplicată:



Funcţiile cercetării ştiinţifice

- cognitivă



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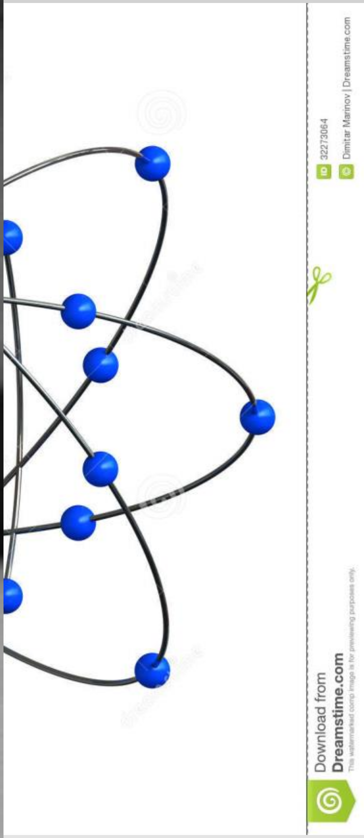

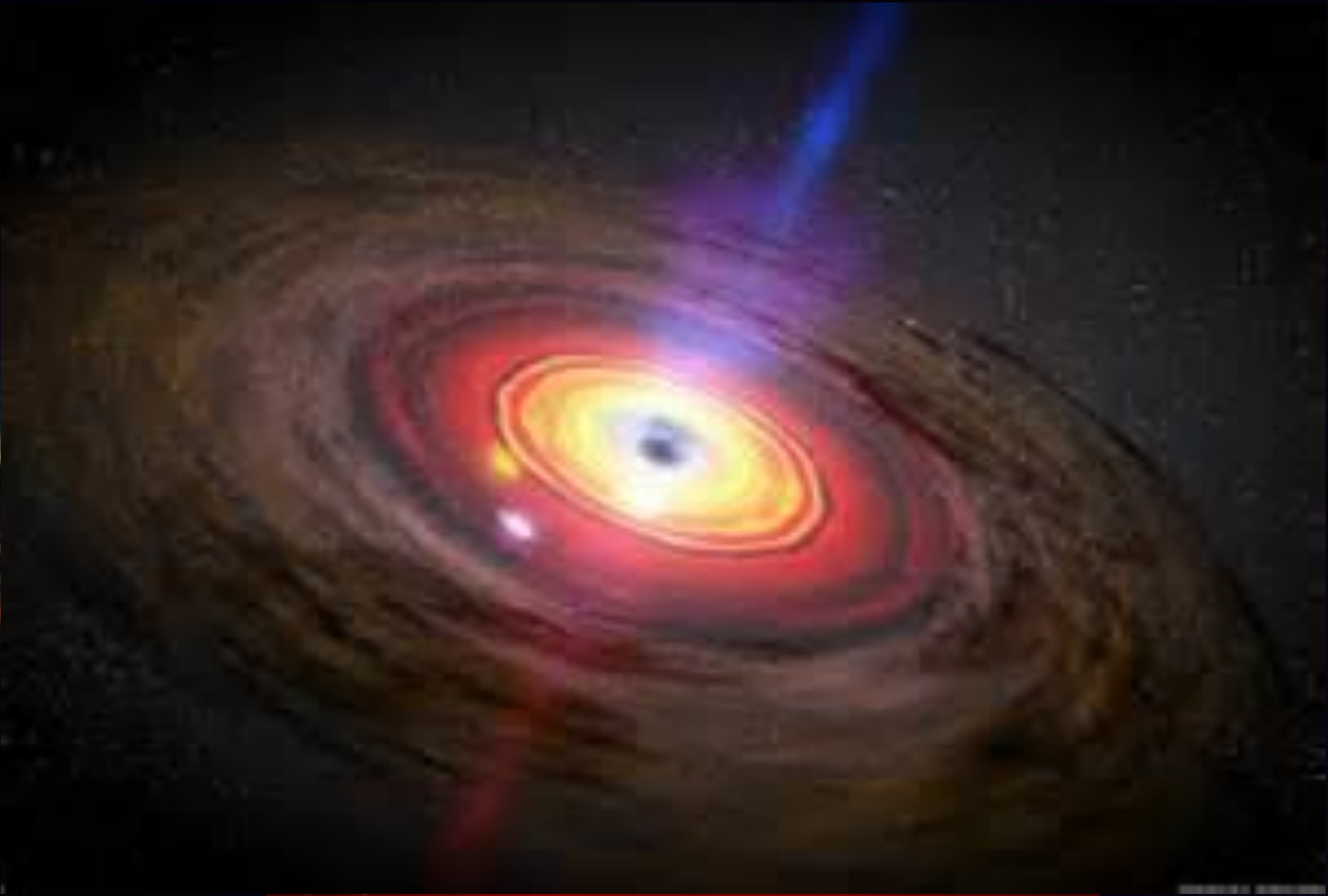


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- informativă

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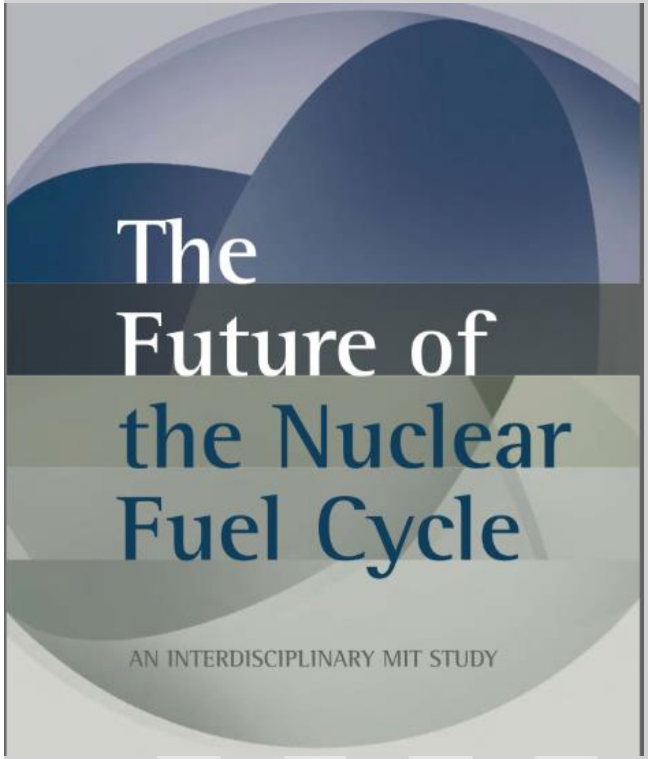

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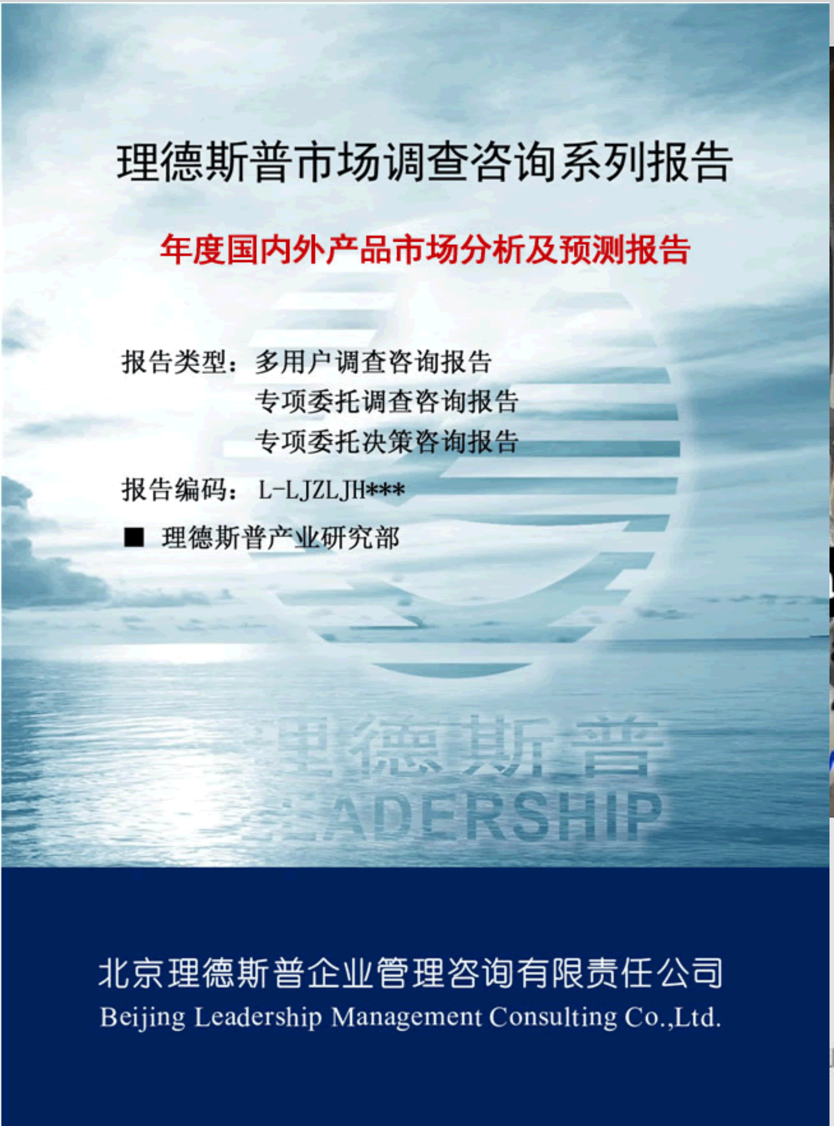

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1. Secţiunea de indexare: titlu, autori, aparteneţă, istoric, rezumat, cuvinte cheie, bibliografie

2. Secţiunea de conţinut: introducere, metode experimentale, rezultate, discuţii, concluzii

3. Secţiunea suplimentară: Menţiuni, multumiri

4. Secţiune complementară: Anexe

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International Journal of ELECTROCHEMICAL SCIENCE
www.electrochemsci.org

Recent Advancements in Electrode Materials for the High-performance Electrochemical Supercapacitors: A Review

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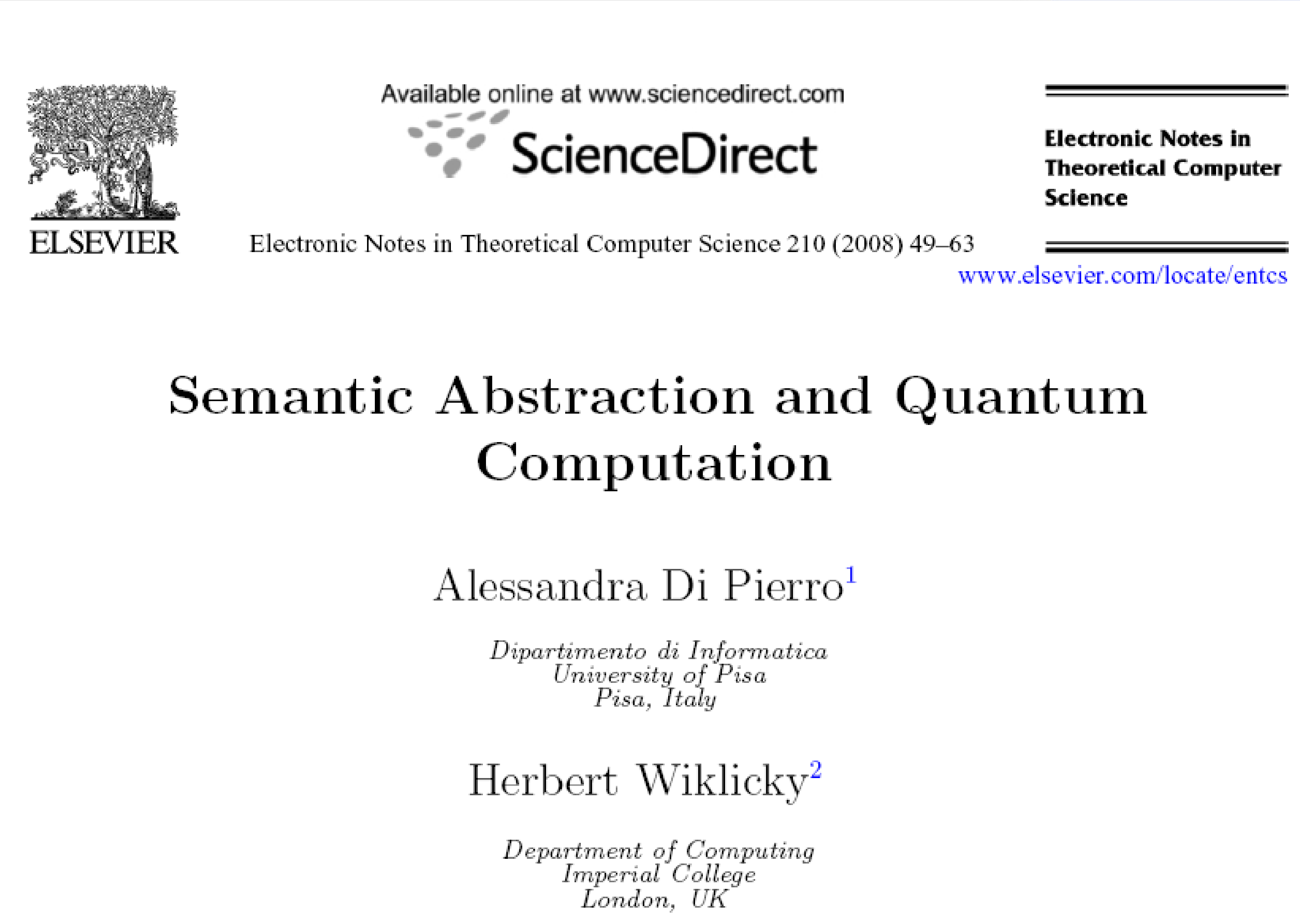
Supercapacitors are energy storage devices emerging as one of the promising energy storage devices in the future energy technology. In this perspective, rapid progress is made in the development of fundamental and applied aspects of supercapacitors. Various techniques have been developed specifically to estimate the specific capacitance. Numerous efforts were made in the literature to increase the specific capacitance value of the electrode materials. The electrode materials which have unique structural and electrochemical capacitance properties, such as high capacity and cyclic stability showed great supercapacitors performances. Recently, there are much new types of electrode materials were developed to play an important role in the capacitance behavior. In this review, we focused on the applications of various nanostructured electrode materials like carbon nanomaterials, metal oxides and conducting polymer towards highly efficient supercapacitors.

Keywords: Carbon based materials, metal oxides, conducting polymers, supercapacitors

Lucrare ştiinţifică teoretică



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Semantic Abstraction and Quantum Computation

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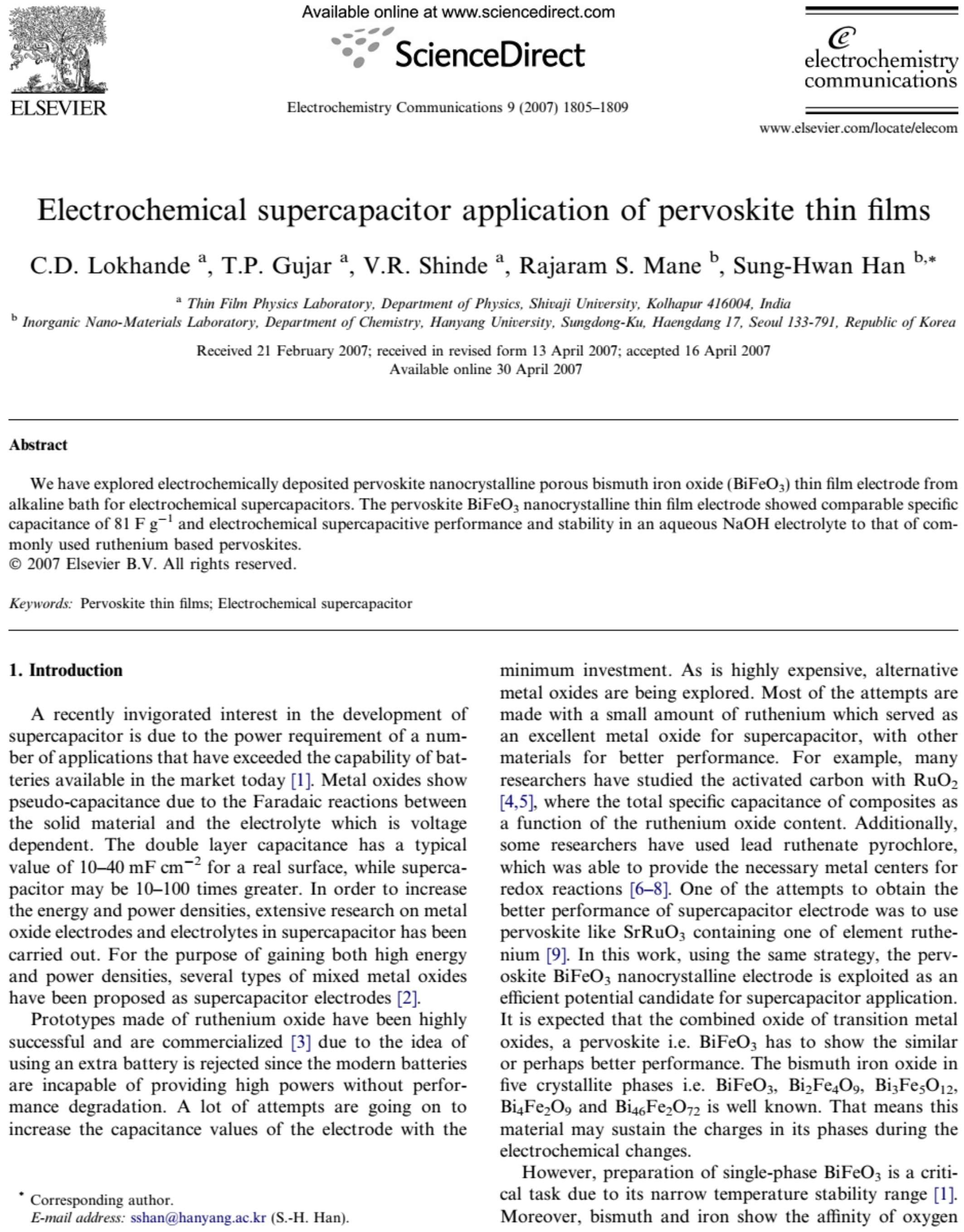
Department of Computing
Imperial College
London, UK

Lucrare ştiinţifică experimentală



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Electrochemical supercapacitor application of pervoskite thin films

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Abstract

We have explored electrochemically deposited pervoskite nanocrystalline porous bismuth iron oxide ($BiFeO_3$) thin film electrode from alkaline bath for electrochemical supercapacitors. The pervoskite $BiFeO_3$ nanocrystalline thin film electrode showed comparable specific capacitance of 81 F g^{-1} and electrochemical supercapacitive performance and stability in an aqueous NaOH electrolyte to that of commonly used ruthenium based pervoskites.

Keywords: Pervoskite thin films; Electrochemical supercapacitor

1. Introduction

A recently invigorated interest in the development of supercapacitor is due to the power requirement of a number of applications that have exceeded the capability of batteries available in the market today [1]. Metal oxides show pseudo-capacitance due to the Faradaic reactions between the solid material and the electrolyte which is voltage dependent. The double layer capacitance has a typical value of 10–40 mF cm^{-2} for a real surface, while supercapacitor may be 10–100 times greater. In order to increase the energy and power densities, extensive research on metal oxide electrodes and electrolytes in supercapacitor has been carried out. For the purpose of gaining both high energy and power densities, several types of mixed metal oxides have been proposed as supercapacitor electrodes [2].

Prototypes made of ruthenium oxide have been highly successful and are commercialized [3] due to the idea of using an extra battery is rejected since the modern batteries are incapable of providing high powers without performance degradation. A lot of attempts are going on to increase the capacitance values of the electrode with the minimum investment. As is highly expensive, alternative metal oxides are being explored. Most of the attempts are made with a small amount of ruthenium which served as an excellent metal oxide for supercapacitor, with other materials for better performance. For example, many researchers have studied the activated carbon with RuO_2 [4,5], where the total specific capacitance of composites as a function of the ruthenium oxide content. Additionally, some researchers have used lead ruthenate pyrochlore, which was able to provide the necessary metal centers for redox reactions [6–8]. One of the attempts to obtain the better performance of supercapacitor electrode was to use pervoskite like $SrRuO_3$ containing one of element ruthenium [9]. In this work, using the same strategy, the pervoskite $BiFeO_3$ nanocrystalline electrode is exploited as an efficient potential candidate for supercapacitor application. It is expected that the combined oxide of transition metal oxides, a pervoskite i.e. $BiFeO_3$ has to show the similar or perhaps better performance. The bismuth iron oxide in five crystallite phases i.e. $BiFeO_3$, $Bi_2Fe_4O_9$, $Bi_3Fe_5O_{12}$, $Bi_4Fe_2O_9$ and $Bi_{46}Fe_2O_{72}$ is well known. That means this material may sustain the charges in its phases during the electrochemical changes.

However, preparation of single-phase $BiFeO_3$ is a critical task due to its narrow temperature stability range [1]. Moreover, bismuth and iron show the affinity of oxygen

---

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Cerinţe fundamentale la redactare

- CLARITATE

- ADECVARE

- NOUTATE

Structura unei lucrări ştiinţifice experimentale:

- Titlu
- Autori
- Apartenenţa autorilor
- Abstract
- Keywords
- Introduction
- Experimental part (details)
- Results and discussion
- Conclusions
- Acknowledgement
- References

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1. confecţionarea de rezultate sau date şi prezentarea lor ca date experimentale, ca date obţinute prin calcule sau simulări numerice pe calculator ori ca date sau rezultate obţinute prin calcule analitice ori raţionamente deductive;
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Experimental part

Redactarea sectiunii "Experimental part"se face pornind la unul dintre principiile de baze ale elaborarii lucrarilor stiintifice:

Orice lucrare stiintifica trebuie sa poata fi verificata de alti cercetatori,
adica experimentele trebuie sa fie reproductibile, fapt care garanteaza validitatea rezultatelor.

- materialele folosite, indicatori de calitate, furnizori, conditionare si depozitare,
- aspecte teoretice ale metodelor aplicate, formule de calcul, standarde, norme specifice,
- descrierea completa si exacta a echipamentului si a tehnicilor folosite,
- prezentarea in mod exhaustiv a conditiilor de lucru si a operatiilor executate,
- parametrii urmariti experimental,
- modul de prelucrare si interpretare a datelor experimentale, tehnici statistice utilizate,
- informatii asupra soft-urilor folosite (licenta!!!)

Cerinte:
- prezentarea sa fie succinta si clara,
- verbele folosite sa fie la timpul trecut!

Results and discussion

Este sectiunea in care sunt anuntate descoperirile autorilor

Sunt prezentate datele obtinute de autori sub forma de:
- tabele,
- grafice,
- diagrame.

Se vor evita redundantele in prezentare.

In mod obligatoriu se includ si rezultatele negative.

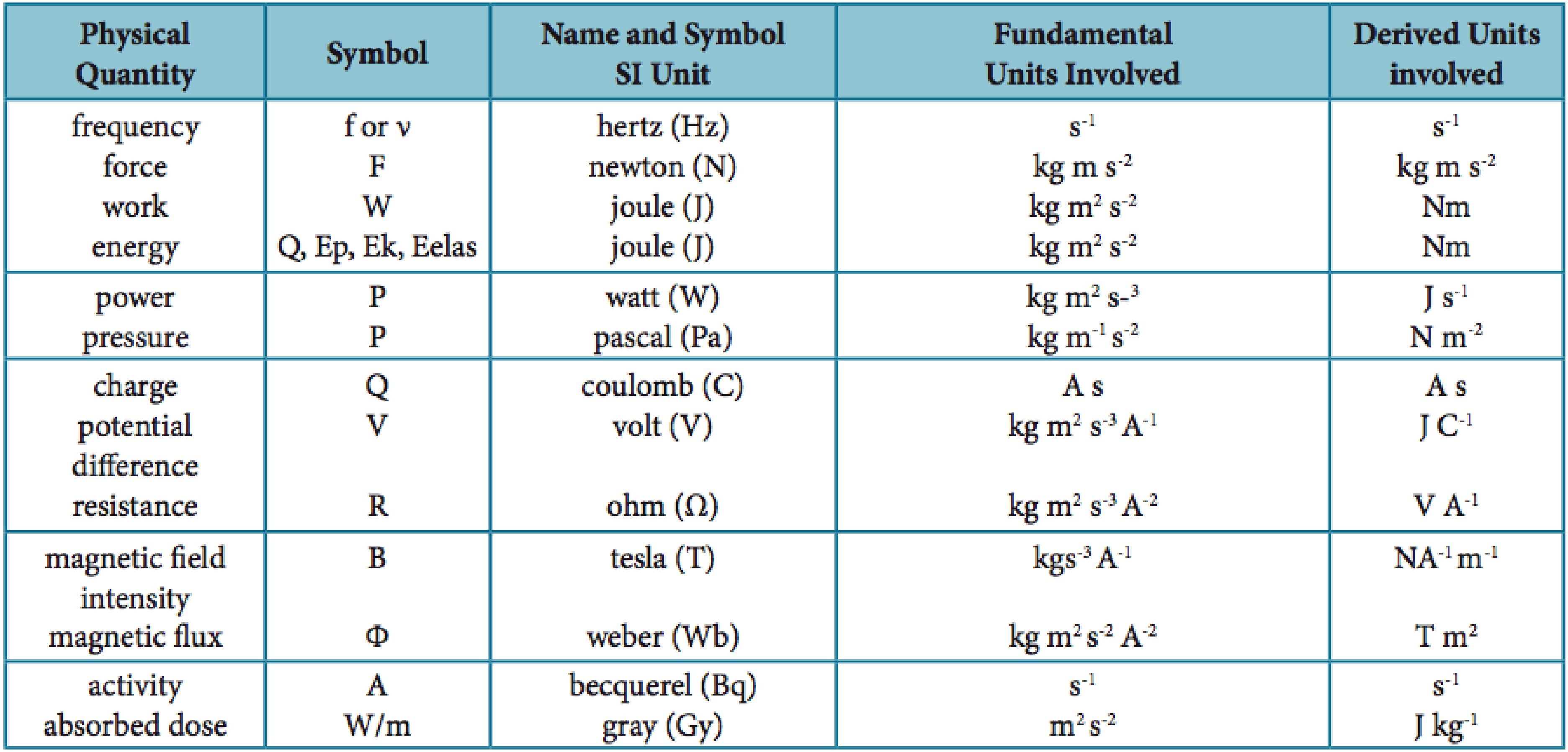
Be sure to include negative results –
writing a results section without them not only invalidate the paper,
but it is extremely bad science.

Unitati de masura - Sistemul International de Unitati

- Unitati fundamentale

Fundamental Quantities	SI Unit	Symbol
Length	meter	m
Mass	kilogram	kg
Time	second	s
Electric current	Ampere	A
Temperature	Kelvin	K
Amount of subtance	mol	mol
Luminous Intensity	candela	cd

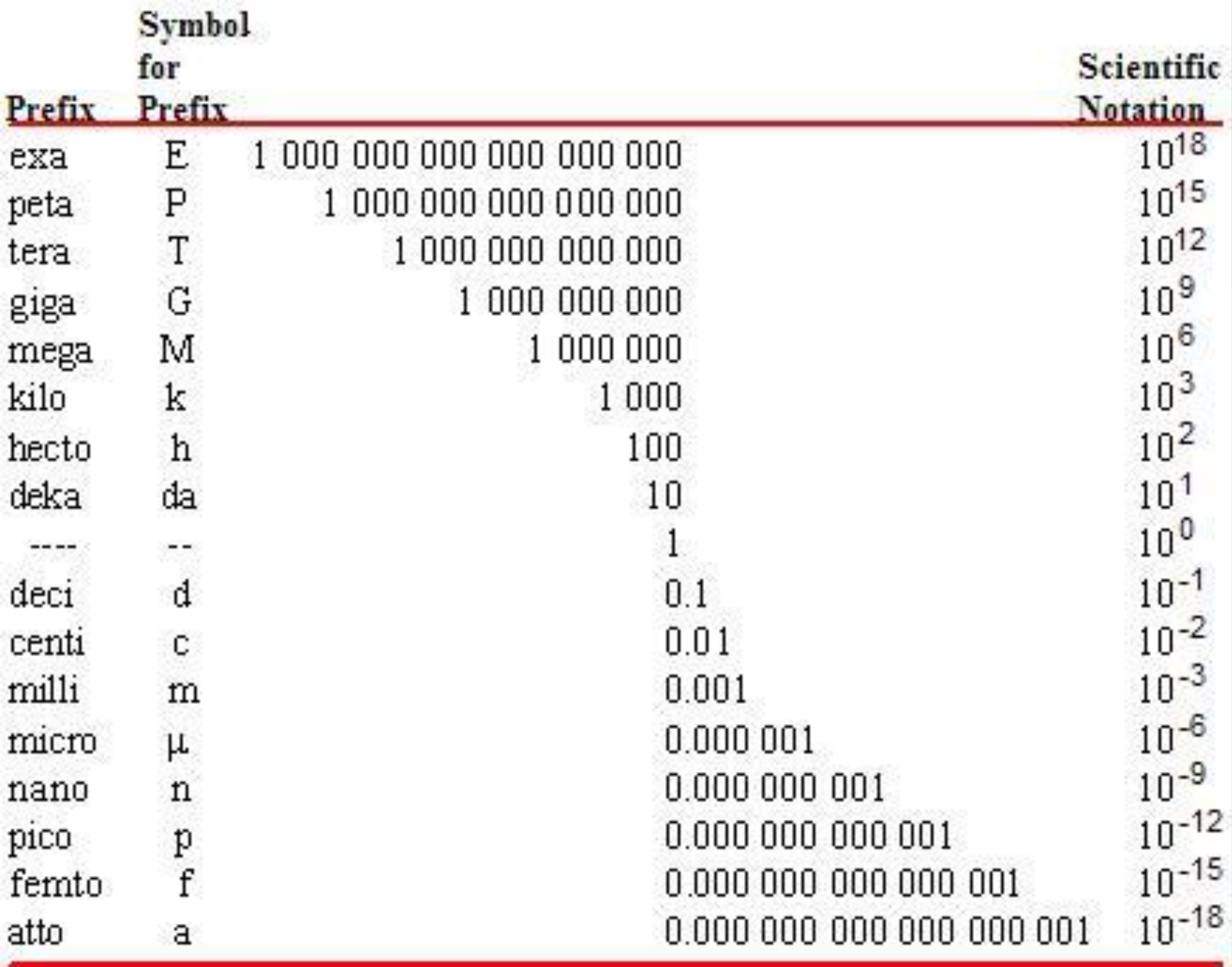
Unitati derivate



Physical Quantity	Symbol	Name and Symbol SI Unit	Fundamental Units Involved	Derived Units involved
frequency	f or ν	hertz (Hz)	s^{-1}	s^{-1}
force	F	newton (N)	kg m s^{-2}	kg m s^{-2}
work	W	joule (J)	kg m^2 s^{-2}	Nm
energy	Q, Ep, Ek, Eelas	joule (J)	kg m^2 s^{-2}	Nm
power	P	watt (W)	kg m^2 s-^{3}	J s^{-1}
pressure	P	pascal (Pa)	kg m^{-1} s^{-2}	N m^{-2}
charge	Q	coulomb (C)	A s	A s
potential difference	V	volt (V)	kg m^2 s^{-3} A^{-1}	J C^{-1}
resistance	R	ohm (Ω)	kg m^2 s^{-3} A^{-2}	V A^{-1}
magnetic field intensity	B	tesla (T)	kgs^{-3} A^{-1}	NA^{-1} m^{-1}
magnetic flux	Φ	weber (Wb)	kg m^2 s^{-2} A^{-2}	T m^2
activity	A	becquerel (Bq)	s^{-1}	s^{-1}
absorbed dose	W/m	gray (Gy)	m^2 s^{-2}	J kg^{-1}

Figure 103 Derived Units

Prefixe pentru unitati



Prefix	Symbol for Prefix		Scientific Notation
exa	E	1 000 000 000 000 000 000	10^{18}
peta	P	1 000 000 000 000 000	10^{15}
tera	T	1 000 000 000 000	10^{12}
giga	G	1 000 000 000	10^{9}
mega	M	1 000 000	10^{6}
kilo	k	1 000	10^{3}
hecto	h	100	10^{2}
deka	da	10	10^{1}
----	--	1	10^{0}
deci	d	0.1	10^{-1}
centi	c	0.01	10^{-2}
milli	m	0.001	10^{-3}
micro	µ	0.000 001	10^{-6}
nano	n	0.000 000 001	10^{-9}
pico	p	0.000 000 000 001	10^{-12}
femto	f	0.000 000 000 000 001	10^{-15}
atto	a	0.000 000 000 000 000 001	10^{-18}

Atentie la prezentarea valorilor numerice: cifre semnificative!!!

0.3452756489 V → 0.3453 V → 0.345 V

pH = 7.67895 → pH = 7.68 → pH = 7.7

0.000000123 m → $0.123 \cdot 10^{-6}$ m → $1.23 \cdot 10^{-7}$ m

Prezentarea unitatilor de masura

$$\frac{\mathrm{kg} \cdot \mathrm{m}}{\mathrm{s}^2}$$

$\mathrm{kg\ m\ s^{-2}}$

Notatii si abrevieri

Temperatura: t T

Energie: E W

Curent: I i

SEM – scanning electronic microscopy;
TEM – transmission electronic microscopy,
DNA – dezoxiribinucleic acid,
NMR – nuclear magnetic resonance

Corrosion Science 69 (2013) 389–395

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Corrosion Science

journal homepage: www.elsevier.com/locate/corsci

Corrosion resistance of carbon steel in weak acid solutions in the of L-histidine as corrosion inhibitor

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L-histidine
Corrosion inhibitor
Inhibition efficiency
Electrochemical impedance spectroscopy

ABSTRACT

This paper presents the inhibitory properties of L-histidine on the corrosi
media containing acetic acid/sodium acetate. The inhibition efficiencies
surements are in good agreement with values given by Tafel method a
spectroscopy. The adsorption of L-histidine obeys the Langmuir isotherm a
energy indicate the nature of interactions between inhibitor molecules
inhibition effect was studied using scanning electron microscopy and en

1. Introduction

The use of chemical compounds in a very wide range of applications is restricted by the protection of the environment and health considerations. Lately, at industrial level, it was required to use chemicals not only due to their efficiency, but also to their safety. These concepts are valid as well in the field of organic compounds which act as corrosion inhibitors for metals and alloys. In this respect, there is a special emphasis on natural compounds, nontoxic and ecologically friendly. In this way, berberine [1], tryptamine [2] and some extracts from natural plants [3–6] were reported as efficient corrosion inhibitors for steel and iron in different test solutions. The amino acids and derivatives have been used as good corrosion inhibitors for mild steel [7–12], carbon steel [13–15], stainless steel [16], iron [17], nickel [18], copper [19,20], aluminum [21], and alloys [22,23] in different aggressive solutions.

Generally, these compounds act on the corrosion process by adsorption on the metal surface, forming a protective film and blocking the active sites. The adsorption mechanism of the inhibitor molecules plays a very important role because the protection degree of the metal or alloy depends on the adsorption process [16].

The aim of this work is to investigate the inhibiting properties of L-histidine on the corrosion process of carbon steel in corrosive solutions containing sodium acetate, the main ingredient used in deicing solutions [24], and acetic acid in order to ensure a constant pH and more aggressive media. Sodium acetate, either liquid or solid, is effective at low temperature in ice and deep snow [24], and
has been used on some winter roadwa
salts, being less corrosive and more su
[25]. Ice Shear is an improved deicing n
acetate and sodium formate which ex
freezing point of water, penetrate ice
bonding between the ice and paveme
deicers with an ecological corrosion i
taking into account that this kind of sol
e.g. highways, airplanes and airports,

Studies on the corrosion process o
solutions are of industrial importanc
boxyl group makes them a basic bui
pounds such as drugs, pharmaceuticals

It is required to mention that L-hi
corrosion inhibitor for mild steel as we
tions [7].

2. Experimental

The weight loss, linear polarization,
spectroscopy and cyclic voltammetry
to observed the inhibitive properties o
process. The surface morphology of th
investigated after removing the produ
tack. The adsorption isotherm of the
studied and discussed.

2.1. Materials

The chemical structure of L-histidin

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http://dx.doi.org/10.1016/j.corsci.2012.12.020

Table 1
Elemental composition of the carbon steel samples.

Element	Fe	C	Si	Mn	P	S	Cr	Ni
wt.%	96.98	0.4184	0.2510	0.7920	0.0132	0.0335	1.162	0.029
Element	Mo	Cu	Al	Ti	V	Co	Nb	W
wt.%	0.2123	0.0234	0.0229	<0.004	0.0124	0.0222	<0.001	<0.010

Experimental measurements were carried out on cylindrical samples of carbon steel with the elemental composition presented in Table 1.

Different concentrations of the inhibitor, L-histidine (Merck, 99%), between 10^{-4} and 10^{-2} M, have been used to determine the inhibition effect of this amino acid on the corrosion rate of carbon steel in 0.5 M CH_3COOH/0.25 M CH_3COONa test solutions. The corrosive media was prepared from glacial acetic acid (Fluka, 99.8%) and sodium acetate (Fluka, 99%).

2.2. Weight loss measurements

Weigh loss (W) experiments were carried out using disc samples of carbon steel having the dimensions Φ 15 mm × 5 mm. Before immersion in the test solutions with or without the inhibitor, the samples were abraded with different SiC paper (220, 500, 800, and 1000), ultrasonically cleaned in distilled water, rinsed with acetone and finally dried. After 240 h exposure time at room temperature in 100 mL corrosive solutions, the specimens were taken out, brushed under running water to remove the corrosion products, washed with distilled water and acetone and dried. Before and after the corrosive attack the samples have been weighed using an analytical balance (precision ±0.1 mg). The weight loss in mg cm^{-2} h^{-1} has been calculated using Eq. (1) [29]:

$$W = \frac{W_1 - W_2}{S \times t} \tag{1}$$

where W_1 and W_2 are the initial and final mass of the samples in mg, S is the total surface area of specimens in cm^2 and t is the exposure time in h.

From the calculated W the inhibition efficiency (η) and surface coverage (θ) of the inhibitor were obtained using Eqs. (2) and (3) [30]:

$$\eta(\%) = \left(1 - \frac{W_{corr}}{W^{o}_{corr}}\right) \times 100 \tag{2}$$

$$\theta = 1 - \frac{W_{corr}}{W^{o}_{corr}} \tag{3}$$

where W_{corr} and W^{o}_{corr} are the weight loss of the samples in the presence and absence of the inhibitor, respectively.

2.3. Electrochemical measurements

Potentiodynamic polarization measurements were carried out in 0.5 M CH_3COOH/0.25 M CH_3COONa test solutions in the absence and presence of different concentrations (10^{-4}–10^{-2} M) of L-histidine, in a conventional three-electrodes cell at room temperature. Carbon steel samples with 1 cm^2 surface area in contact with solution were used as working electrodes, the reference was a silver–silver chloride electrode (Ag/AgCl/saturated KCl) and two graphite rods were used as counter electrodes. The samples preparation before the experiments was similar to weight loss measurements. Before each experiment the test solutions were deaerated by bubbling high purity nitrogen in order to avoid any reaction with dissolved oxygen. Polarization curves were obtained with an Autolab PGSTAT 302N at a scan rate of 2 mV s^{-1} in the potential range of −200 mV to +200 mV relative to the open circuit potential. Electrochemical impedance spectroscopy (EIS) measurements were carried out at E_{corr} at 298 ± 1 K after a stationary state was achieved in the frequency range from 10 kHz to 10 mHz and AC voltage amplitude of 10 mV peak to peak. The EIS spectra were measured in a three electrodes cell connected to a PAR 2263 potentiostat. The working electrode was carbon steel with an active surface of 0.5 cm^2, the counter electrode was a Pt wire and the reference a saturated calomel electrode. Before each experiment the surface of the electrode was prepared similarly as previously described and the test solutions were deaerated by bubbling pure nitrogen for 1 h.

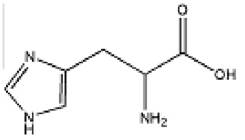

Fig. 1. Chemical structure of L-histidine.

2.4. Surface morphology

Carbon steel surface was analyzed with a Keyence VHX-600 Digital Microscope and a Philips XL 30 ESEM scanning electron microscope (SEM), respectively. The images of the samples, same used in the weight loss measurements, were recorded after 240 h exposure time in test solutions in the absence and the presence of L-histidine.

3. Results and discussion

3.1. Weight loss data

From the weight loss data of carbon steel samples after 240 h immersion time in corrosive solutions, in the absence and presence of different amounts of L-histidine, the inhibition efficiency and surface coverage were calculated and the results are given in Table 2. The results indicate that amino acid reduces the corrosion process and the effect is more pronounced as the concentration of the inhibitor increases.

Table 2
The inhibition efficiency and surface coverage obtained by weight loss measurements.

L-histidine conc. (M)	η (%)	θ (surface coverage)
1×10^{-4}	31.9	0.319
5×10^{-4}	44.5	0.445
1×10^{-3}	58.2	0.582
5×10^{-3}	68.7	0.687
1×10^{-2}	81.6	0.816

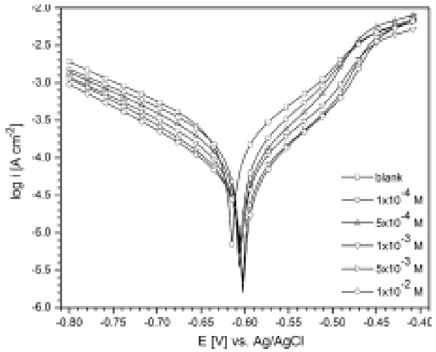

Fig. 2. Linear polarization curves on carbon steel electrode in 0.5 M CH_3COOH/0.25 M CH_3COONa with and without various concentrations of L-histidine.

3.2. *Linear polarization*

One of the most used methods to determine the corrosion rate of metals and alloys in corrosive solutions is linear polarization. Fig. 2 reveals the cathodic and anodic polarization curves obtained for carbon steel in 0.5 M CH_3COOH/0.25 M CH_3COONa at room temperature in the absence and presence of different concentrations of L-histidine.

The values of electrochemical parameters associated with this process, i.e. corrosion potential (E_{corr}), corrosion current density (i_{corr}), cathodic and anodic Tafel slopes (b_c and b_a) and corrosion rate were determined by extrapolating potentiodynamic curves using GPES software. Also, the inhibition efficiency (η) and surface coverage have been calculated with Eqs. (4) and (5) [31] and the values obtained are presented in Table 3.

$$\eta(\%) = \left(\frac{i_{corr} - i^{o}_{corr}}{i_{corr}}\right) \times 100 \tag{4}$$

$$\theta = \frac{i_{corr} - i^{o}_{corr}}{i_{corr}} \tag{5}$$

where i_{corr} and i^{o}_{corr} are the uninhibited and inhibited corrosion current densities, respectively.

The analysis of data given in Table 3 shows that both the cathodic and anodic Tafel slopes decrease indicating that L-histidine is a mixed type inhibitor acting on both the hydrogen evolution reaction and metal dissolution. Moreover, the Tafel slopes do not suffer significant variation with increase of amino acid concentration suggesting that the presence of inhibitor in the test solution does not modify the process mechanism and act as adsorptive inhibitor, retarding both cathodic and anodic reaction by blocking the active sites [17,32]. An increase in inhibitor concentration probably increase the number of inhibitor
interface and decrease the corro
the corrosion potential value slig
The inhibition process can be att
L-histidine molecules on the active
sition of corrosion products on th

From the Tafel polarization res
inhibition mechanism involves bl
the inhibitor molecules via adsor

3.3. *EIS measurements*

Information about the inhibiti
also obtained by electrochemica
advantage of this method is that a
values also double layer capacitar
same measurement.

Fig. 3 gives the EIS spectra of
without and with various concen

The complex plane plots revea
pressed semicircle. The diameter
the polarization resistance and it
of L-histidine.

A simple electrical equivalent
to model the experimental data.
nection between the solution res
tion of the double layer capac
resistance. Due to the deviations f
itance is usually replaced by a
whose impedance is given by:

$$Z_{CPE} = 1/T(j\omega)^{n}$$

where T is a parameter related to t
is an exponent between 0 and 1 de
of the CPE.

The experimental data were fit
linear least-squares (CNLSs) proc
ues of the circuit elements are giv
standard errors.

The inhibition efficiency was
resistance values using Eq. (7) [3

$$\eta(\%) = \left(\frac{R_{p,inh} - R_p}{R_{p,inh}}\right) \times 100$$

where $R_{p,inh}$ and R_p are polarizati
ence and absence of L-histidine.

The obtained values of the in
the concentration of amino acid
the values calculated from linea
weight loss measurements.

Double layer capacitance value
impedance data using Eq. (8) and

$$T = C^{n}_{dl}\left(R_S^{-1} + R_p^{-1}\right)^{1-n}$$

Table 3
Polarization parameters for the corrosion of carbon steel in 0.5 M CH_3COOH/0.25 M CH_3COONa with/without various concentration

Inh. conc. (M)	i_{corr} (μA cm^{-2})	$-E_{corr}$ (mV)	$-b_c$ (mV dec^{-1})	b_a (mV dec^{-1})	R_p (Ω)	Corr. r
Blank	159.0	615	168	118	44.3	1.87
1×10^{-4}	114.1	605	165	102	64.1	1.34
5×10^{-4}	89.3	605	155	102	76.1	1.05
1×10^{-3}	70.1	605	152	102	96.5	0.82
5×10^{-3}	52.2	603	142	95	121.8	0.61
1×10^{-2}	33.1	601	141	87	148.7	0.48

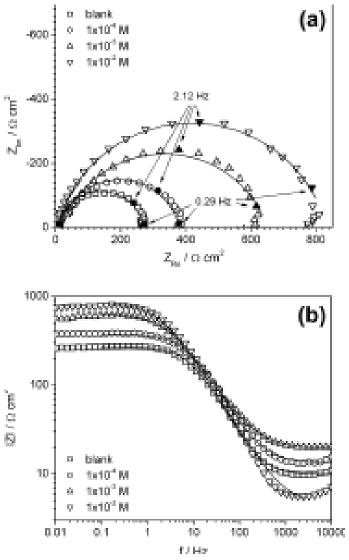

Fig. 3. Experimental Nyquist (a) and Bode (b) plots of carbon steel in 0.5 M CH_3COOH/0.25 M CH_3COONa with and without various concentrations of L-histidine. Symbols are experimental data and continuous lines are obtained by CNLS fitting.

It can be observed that the double layer capacitance in the absence of inhibitor is larger than in its presence. The addition of L-histidine in the test solution leads to a decrease of the double layer capacitances, because the inhibitor molecules adsorb at the inner Helmholtz plane and block the active sites on the metal surface.

3.4. *Cyclic voltammetry*

The manner in which L-histidine can influence the corrosion process is pointed by its electrochemical behavior in test solutions studied by cyclic voltammetry. Fig. 4 shows the cyclic voltammograms recorded in 0.5 M CH_3COOH/0.25 M CH_3COONa on platinum electrode in the absence and the presence of different amounts of inhibitor. On the base curve, in the absence of L-histidine, the anodic branch corresponding to the oxygen release (curve 1) and the cathodic one for hydrogen evolution reaction (curve 2) are distinctive. Further, one can also observe the reduction peak (3) of the oxide layer formed on electrode surface and the peak (4) associated with the oxidation of adsorbed hydrogen. The addition of compound in the test solution leads to an increase of the current (5) that can be attributed to amino acid oxidation. Also, the presence of inhibitor in the test solution reduces the cathodic and anodic processes.

The cyclic voltammograms measured on carbon steel electrode in the same test solutions in the absence and the presence of L-histidine is presented in Fig. 5. The main curve shows, along the oxygen and hydrogen evolution reactions (2 and 6), the characteristic peaks for metal dissolution (1 and 4). On the cathodic branch, one can observe the reduction current (peak 3) corresponding to the reduction of the passive film and the current plateau 5 attributed to the reduction of iron ions to metallic iron. Also, in the anodic sense, peak 7, corresponding to the oxidation of adsorbed hydrogen on metal surface, is observed.

In the presence of the amino acid in the test solution, the passivation of the metal surface occurs more easily, critical current being reduced.

3.5. *Adsorption isotherm*

Useful information about the inhibition mechanism of carbon steel corrosion process can be gained from adsorption isotherm, by determining the standard Gibbs free energy value that characterizes the type of interaction existing between inhibitor molecules and metal surface. The Gibbs energy was calculated with Eq. (9) [35].

$$\Delta G^{o}_{ads} = -RT\ln(55.5K_{ads}) \tag{9}$$

where R is the universal gas constant 8.314 J mol^{-1} K^{-1}, T the thermodynamic temperature in K, K_{ads} is the equilibrium constant for the adsorption process and 55.5 represents the molar concentration of water in the solution.

In the case of amino acids adsorption the most suitable isotherm was found to be Langmuir isotherm, written in the linear form (10) [10,11,23]:

$$\frac{c_{inh}}{\theta} = \frac{1}{K_{ads}} + c_{inh} \tag{10}$$

From the linear dependence $c_{inh}/\theta = f(c_{inh})$ drawn using the experimental parameters computed from weight loss data and Tafel slopes (Fig. 6), the value of the adsorption constant was calculated. The regression coefficient R^2 for all methods was very close to 1, indicating that the adsorption process of L-histidine on metal surface obeys the Langmuir isotherm. The values of standard Gibbs free energy determined by the three methods are −28.7 kJ mol^{-1} by Tafel slopes, −28.5 kJ mol^{-1} by weight loss data and −30.8 kJ mol^{-1} by EIS measurements, revealing that the adsorption of inhibitor molecules on metal surface takes place due to electrostatic interactions between the amino acid molecules and electrode surface. Generally, values of the free energy less negative than −40 kJ mol^{-1} are associated with processes that occur by physical adsorption of the inhibitor on metal surface [36].

It is noted that the mechanism of corrosion inhibition of carbon steel in 0.5 M CH_3COOH/0.25 M CH_3COONa solution in the presence of L-histidine can be deduced on the basis of adsorption isotherms. It is well known that in an acid solution L-histidine exists as a protonated species like some other amino acids [17,20,21]. For acetic acid/sodium acetate the calculated value of pH is 4.46 and the protonation degree of the inhibitor is very close to 1, taking into account the following reaction equilibrium equations:

Table 4
Experimental values of EEC parameters for carbon steel in 0.5 M CH_3COOH/0.25 M CH_3COONa solution with/without various concentration of L-histidine.

Inh. conc. (M)	R_s (Ω)	T (F cm^{-2} s^{n-1})	n	R_p (Ω cm^2)	C_{dl} (μF cm^{-2})	η (%)
Blank	9.3 (1.5%)	1.16×10^{-4} (3.9%)	0.90 (0.7%)	264 (1.1%)	54.6	–
1×10^{-4}	12.5 (1.7%)	1.14×10^{-4} (4.0%)	0.85 (0.8%)	375 (1.3%)	35.7	29.6
1×10^{-3}	18.8 (1.0%)	1.64×10^{-4} (2.2%)	0.80 (0.5%)	642 (1.0%)	38.4	58.9
1×10^{-2}	4.5 (5.6%)	1.39×10^{-4} (2.5%)	0.85 (fixed)	828 (3.8%)	37.8	68.1

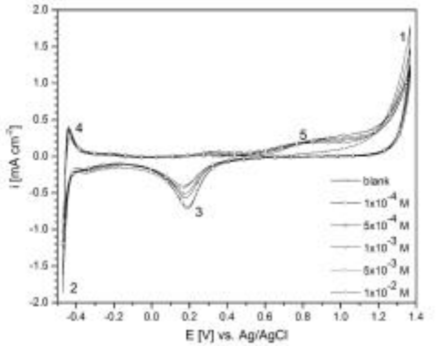

Fig. 4. Cyclic voltammograms on Pt electrode in 0.5 M CH_3COOH/0.25 M in the absence/presence of ʟ-histidine, scan rate 50 mV s^{-1}.

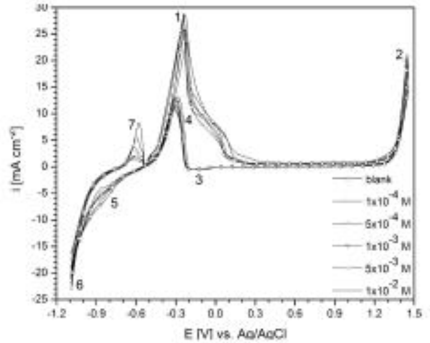

Fig. 5. Cyclic voltammograms on carbon steel electrode in 0.5 M CH_3COOH/0.25 M in the absence/presence of ʟ-histidine, scan rate 50 mV s^{-1}.

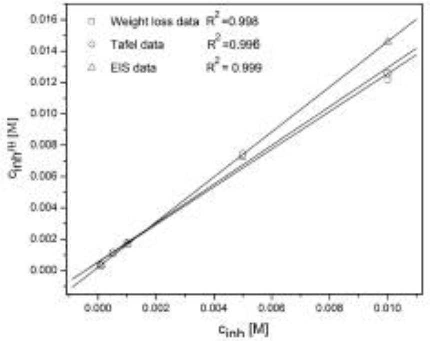

Fig. 6. Langmuir adsorption isotherm for ʟ-histidine obtained by weight loss, EIS and Tafel data.

$$H_3N{-}R(NH^+){-}COOH \overset{pK_{a1}}{\rightleftarrows} H_3N^+{-}R(NH$$
$$\overset{pK_{a2}}{\rightleftarrows} H_3N^+{-}R(N){-}COO^- \overset{pK_{a3}}{\rightleftarrows} H_2N{-}R($$

The values of acidity constants are
$pK_{a3} = 9.08$ [37]. As a consequence, in
almost all amino acid molecul
($H_3N^+{-}R(NH^+){-}COO^-$). Due to the elect
carbon steel and inhibitor molecules,
on the cathodic active sites of metal
evolution reaction. At the same time,
molecules can adsorb on the steel surf
nitrogen atoms of imidazole ring o
unshared electron pairs.

3.6. Surface analysis

The surface morphology of the carb
tigated after corrosive attack by the me
time in the absence and presence of t
graphs have been performed after th
products formed on the sample surfa
of ʟ-histidine the surface of the sampl
(Fig. 7a), while the presence of His fav
compact layer that protects the sampl

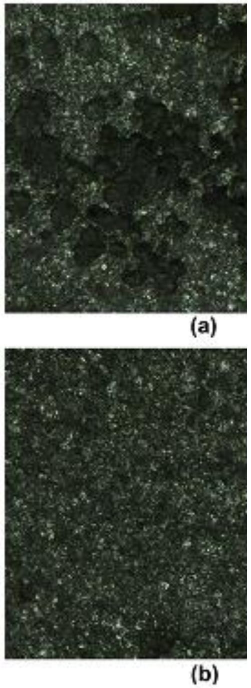

Fig. 7. Microscope images after removing the
(b) with 10^{-2} M ʟ-histidine.

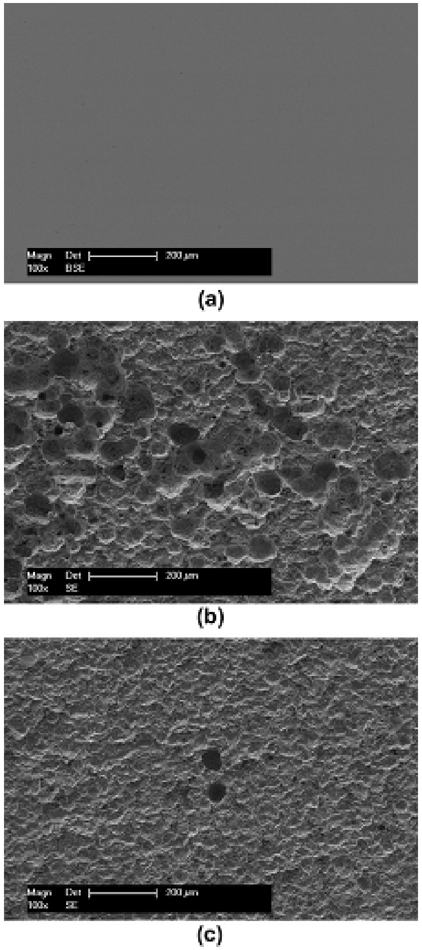

Fig. 8. SEM images obtained for carbon steel samples: abraded without immersion in test solutions (a); after 240 h immersion in the blank solution (b); and after 240 h immersion in the test solution with 10^{-2} M ʟ-histidine (c).

Details about surface morphology were obtained by scanning electron microscopy.

Fig. 8 shows the SEM images recorded for the abraded surface of the carbon steel sample (a) and after removing the oxide layer from the surface. In the absence of inhibitor the image reveals clear pits and cavities with roughness due to metal dissolution under corrosive attack by the media (b). For 10^{-2} M concentration of inhibitor there are fewer corrosion marks observed on sample (c), suggesting that the carbon steel surface was protected by ʟ-histidine molecules.

The atomic percentage of the elements is shown in Table 5.

Table 5
Atomic percentage of the elements.

	Fe	C	Cr	Si	Al	Mo	O
Blank	74.17	22.45	1.5	1.38	–	–	–
10^{-2} M inh.	45.77	38.94	1.6	4.85	1.35	1.56	5.93

4. Conclusions

ʟ-histidine is a mixed safe corrosion inhibitor for carbon steel in weak acid solutions that acts both over the cathodic process of hydrogen evolution and the anodic process of metal dissolution. Inhibition efficiency was determined by three different methods: weight loss, linear polarization and electrochemical impedance, all giving comparable results. Inhibition efficiency increases with the amount of inhibitor and the maximum value was 81.6% for 10^{-2} M ʟ-histidine. The amino acid adsorption on metal surface obeys Langmuir isotherm, and the value of Gibbs free energy reveal the electrostatic interactions between the charged molecules and the charged carbon steel. Generally, values of the free energy less negative than −40 kJ mol^{-1} are associated with processes that occur by physical adsorption of the inhibitor on metal surface. From surface analysis it results that amino acid suppresses the corrosion process by forming a protective film over the metal surface which prevents penetration of the corrosive media. The double layer capacitance obtained from EIS measurements decreases with the increasing of inhibitor concentration, arguing an adsorption process of the ʟ-histidine on the metal surface.

The good inhibition efficiencies of ʟ-histidine in acetic acid/sodium acetate solution makes it a suitable alternative as environmentally friendly corrosion inhibitor in deicing solutions in which sodium acetate is a main component. The advantages of this inhibitor consist in biodegradability and zero impact on the environment.

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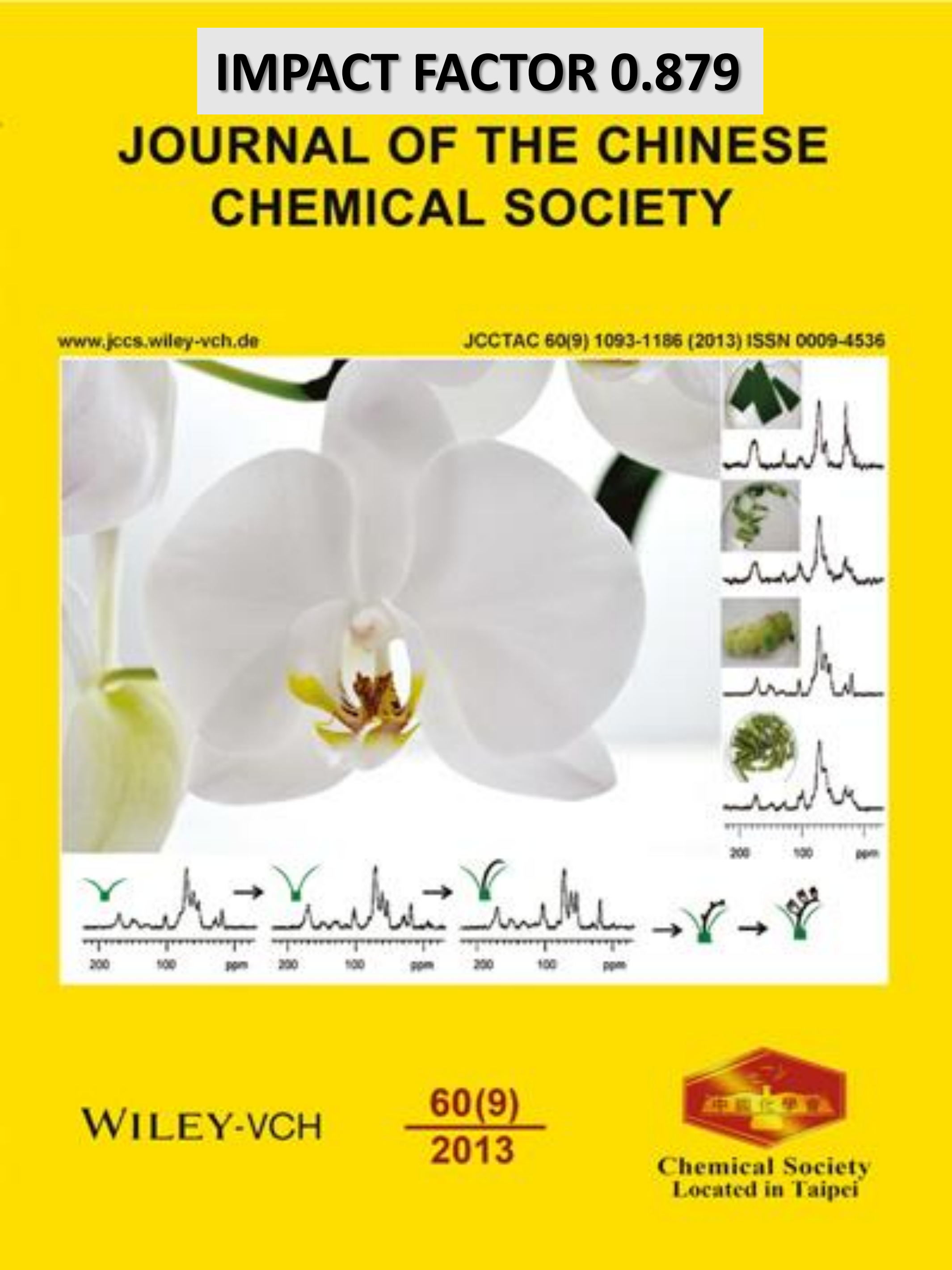

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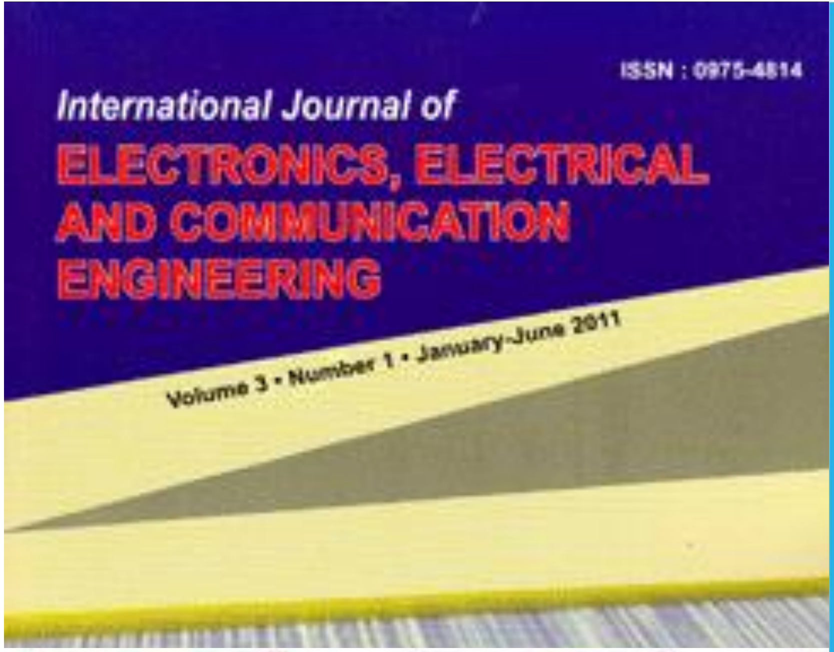

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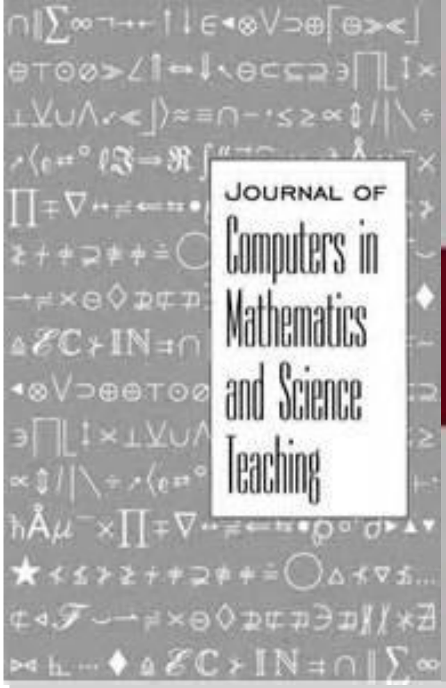
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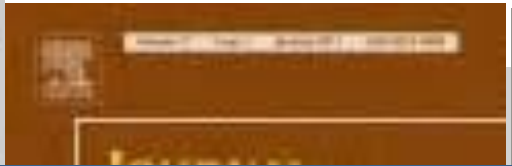


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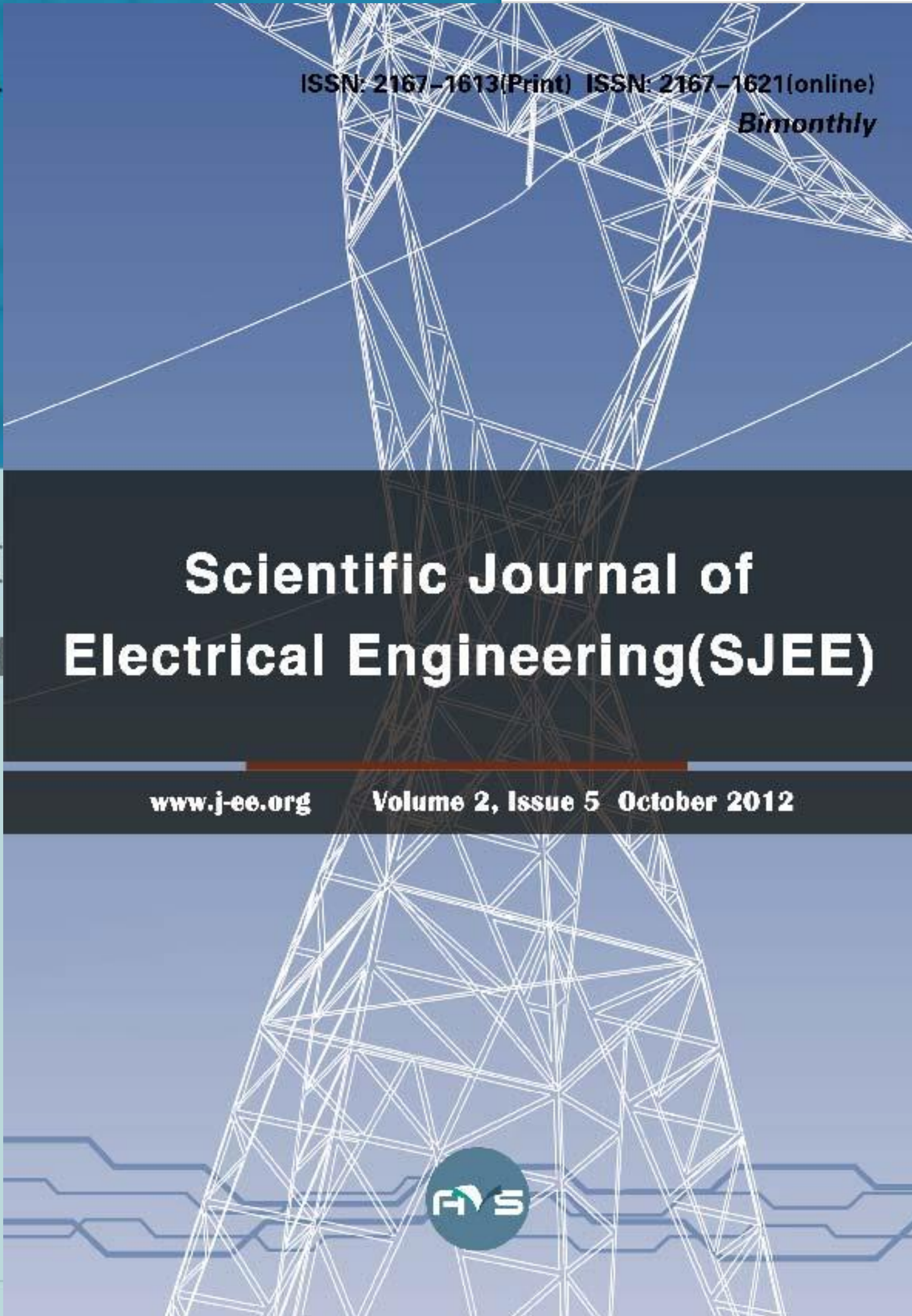
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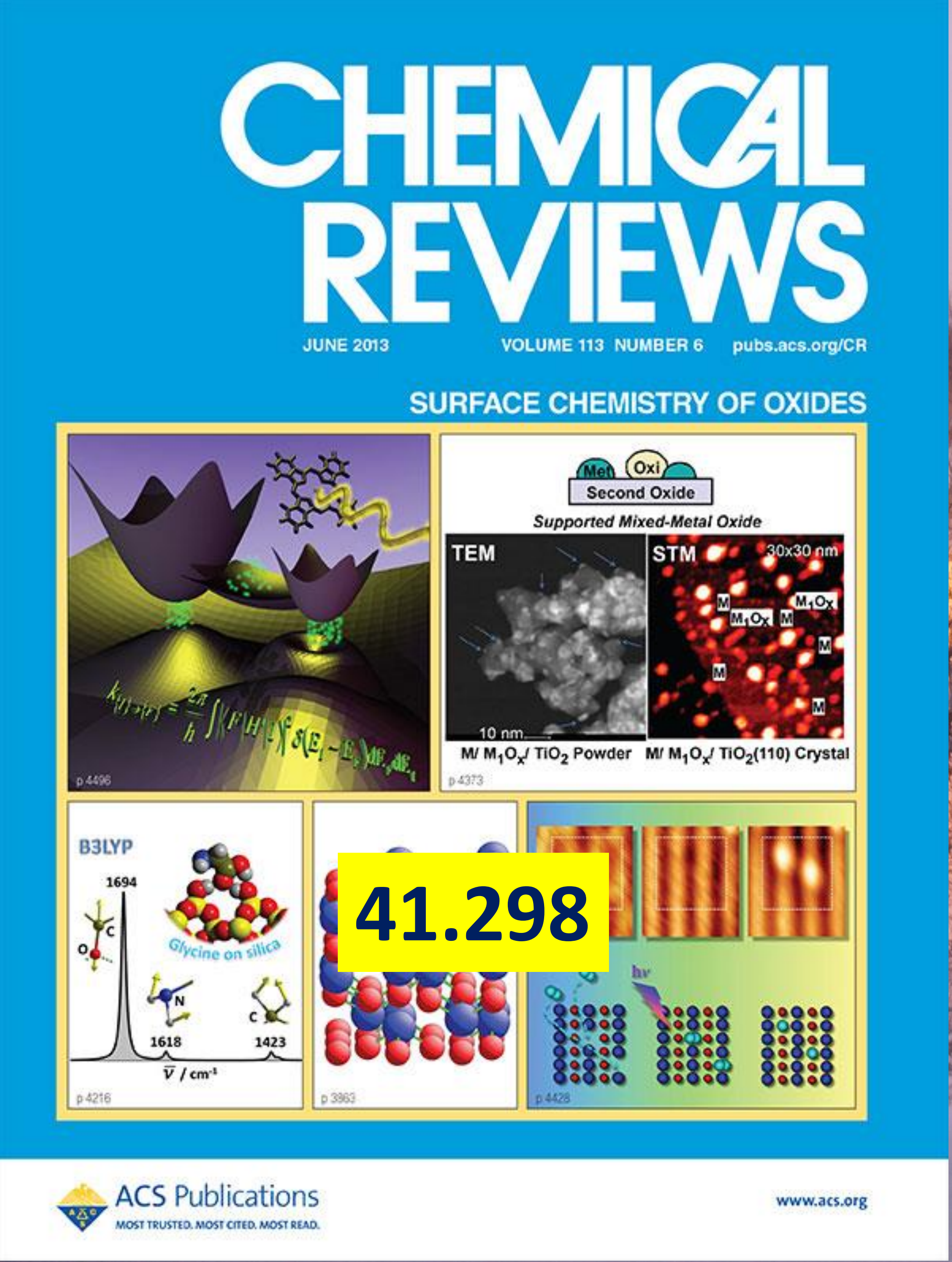

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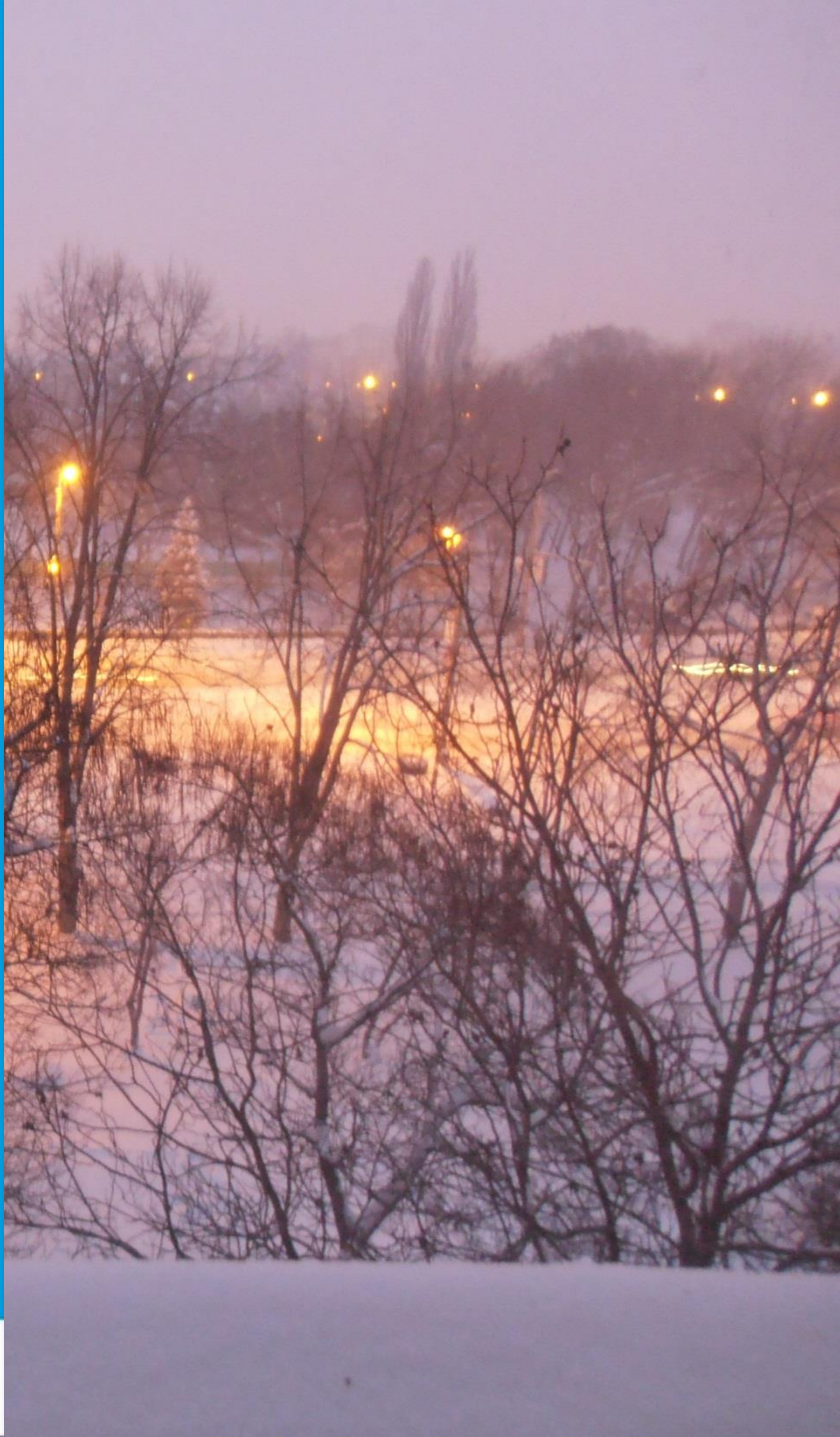

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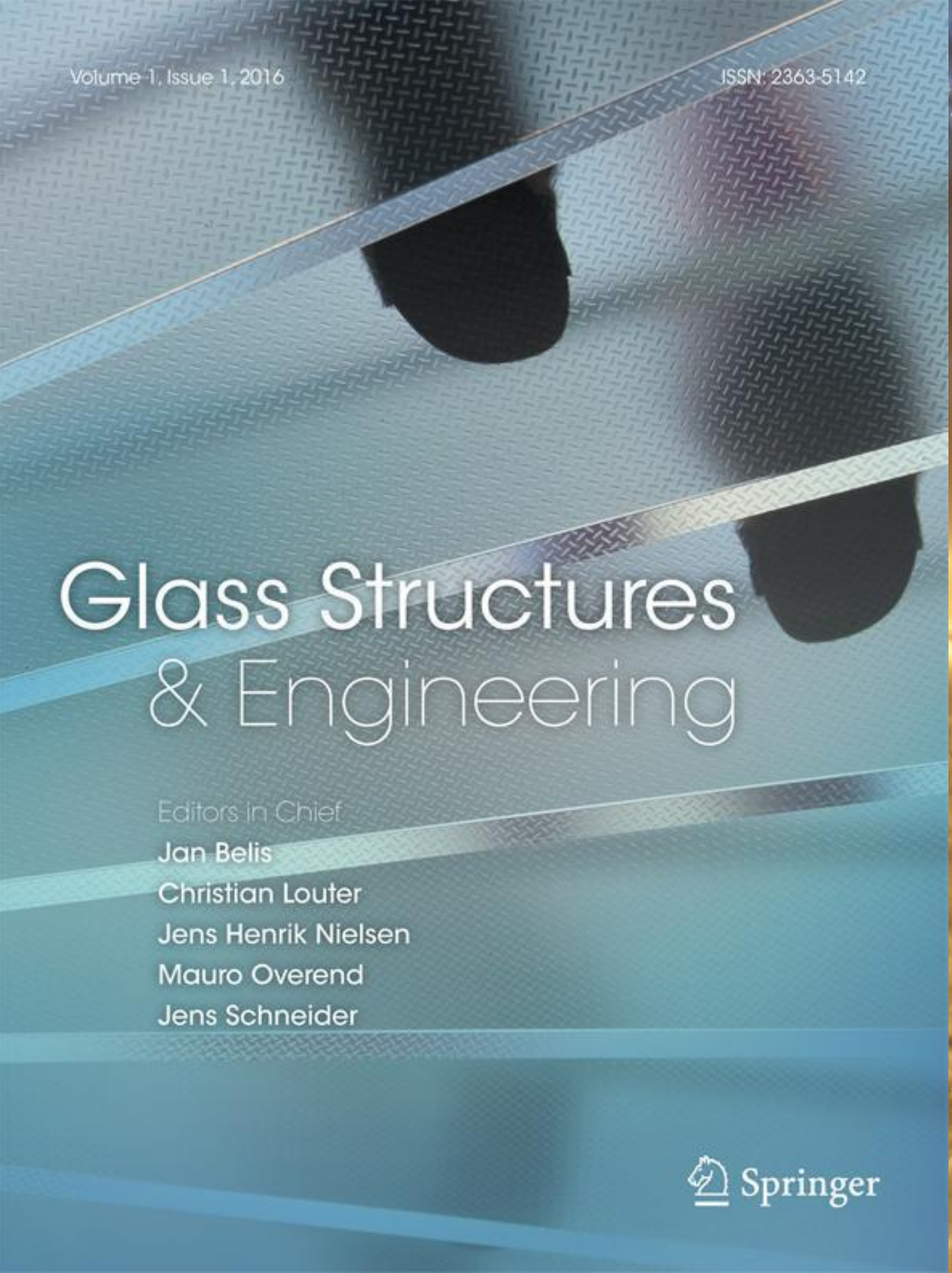

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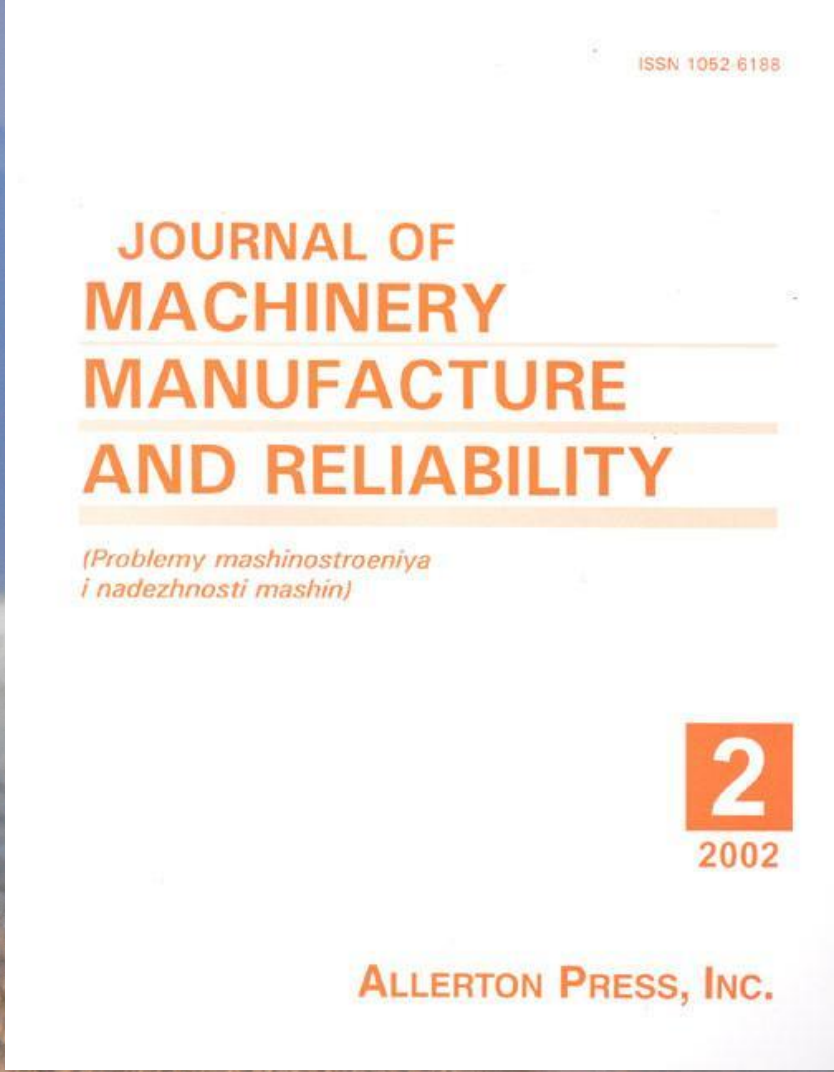

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Green IT in Practice: SQL Server Consolidation in Microsoft IT

Contents

Foreword 1
by Diego Dagum

Environmentally Sustainable Infrastructure Design 2
by Lewis Curtis
A comprehensive understanding of environmental sustainability needs for IT infrastructure system design.

Green Maturity Model for Virtualization 9
by Kevin Francis and Peter Richardson
The authors present a study on the maturity of virtualization practices and offer a sustainability roadmap for organizations planning a green agenda.

Application Patterns for Green IT 16
by Dan Rogers and Ulrich Homann
A complete analysis on power-efficient applications design, considering tools that help the architect achieve scalability without deriving in energy waste.

Architecture Journal Profile: Udi Dahan 22
For this issue's interview, we catch up with Udi Dahan, noted expert on SOA and .NET development, and four-time Microsoft MVP.

Profiling Energy Usage for Efficient Consumption 24
by Rajesh Chheda, Dan Shookowsky, Steve Stefanovich, and Joe Toscano
This article suggests that tracking energy consumption at every level will become the factor of success for green architecture practitioners.

Project Genome: Wireless Sensor Network for Data Center Cooling 28
by Jie Liu, Feng Zhao, Jeff O'Reilly, Amaya Souarez, Michael Manos, Chieh-Jan Mike Liang, and Andreas Terzis
Learn about a research project on an interesting adaptive cooling technique.

Green IT in Practice: SQL Server Consolidation in Microsoft IT 35
by Mark Pohto
This Microsoft IT case study offers useful guidelines for effective resource consolidation for environmental sustainability.

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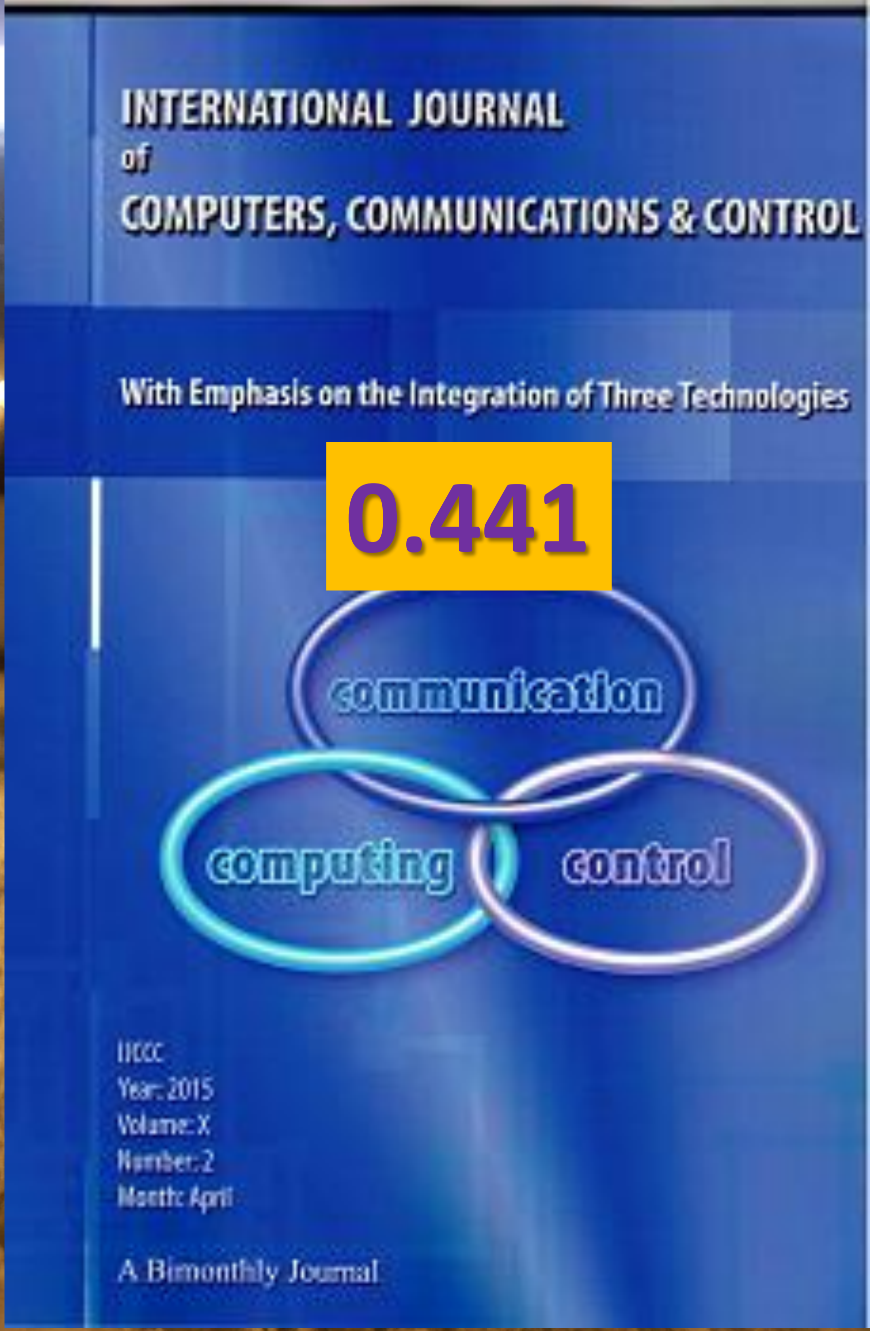

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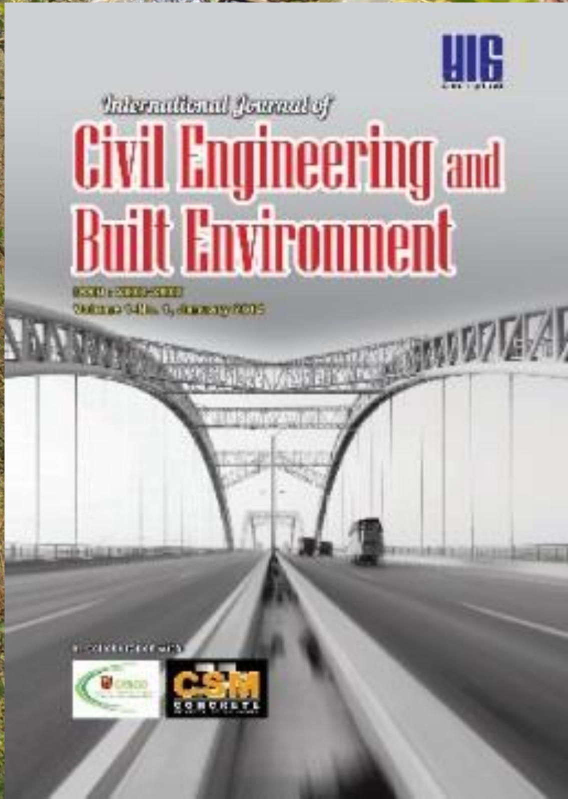

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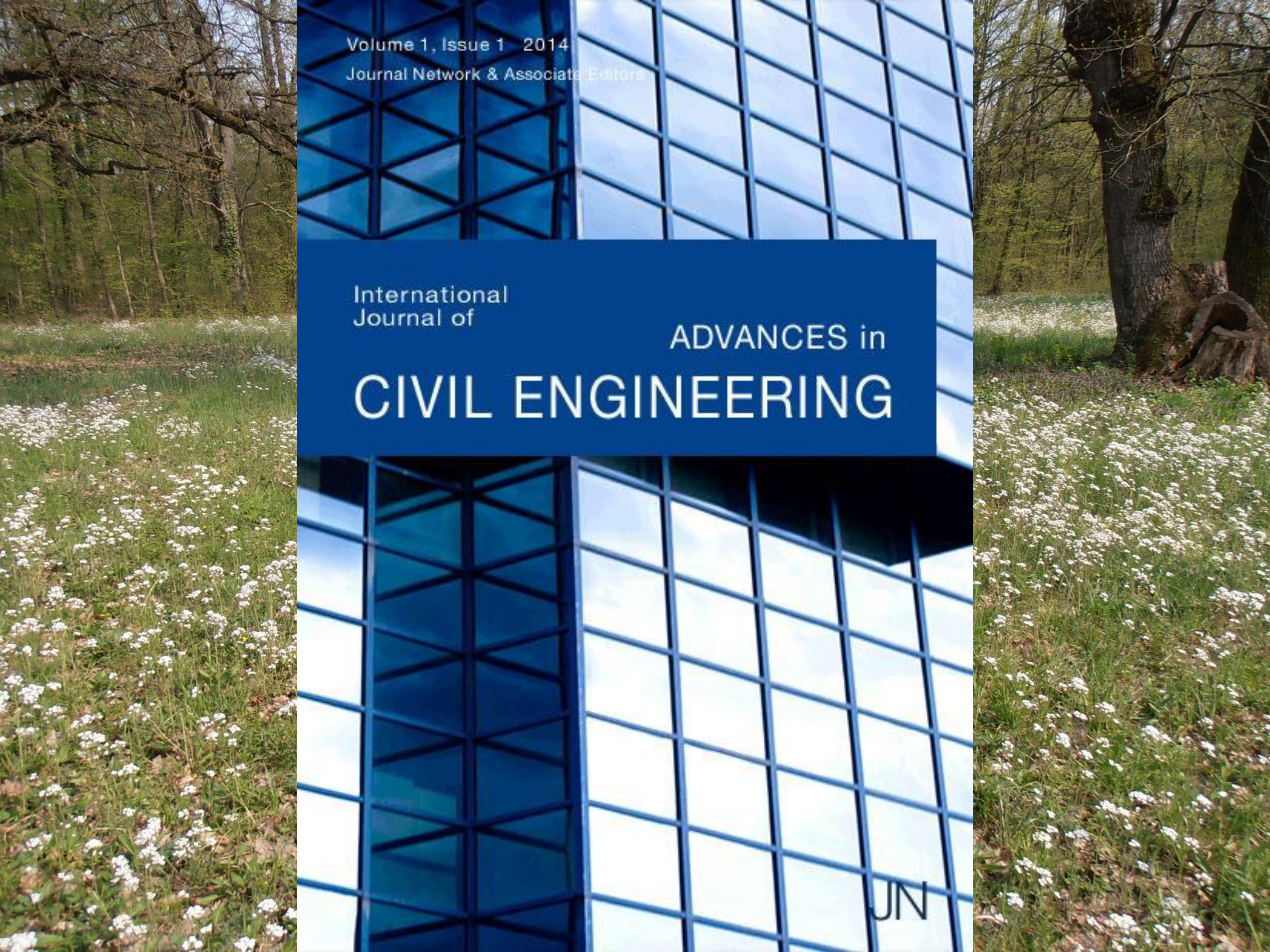

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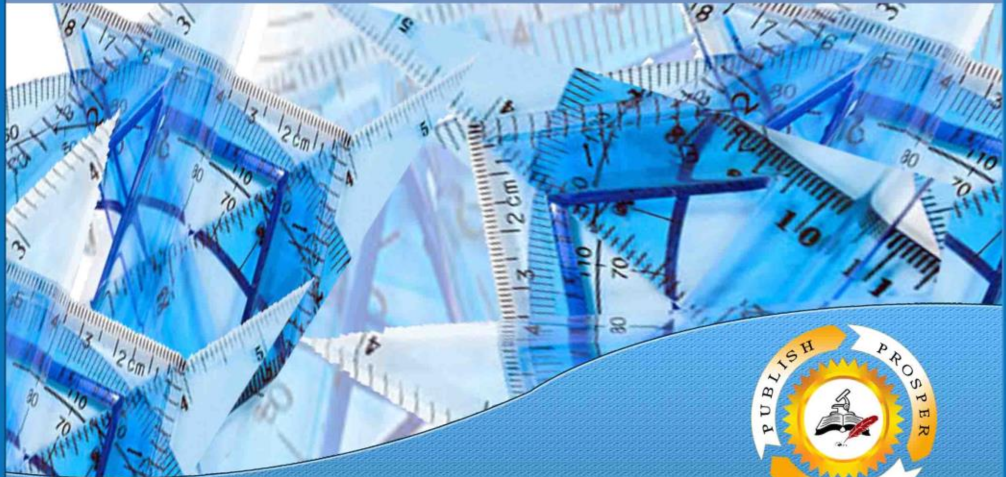

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