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Combustion synthesis of near-infrared reflective brown pigments based on iron-doped lanthanum aluminate

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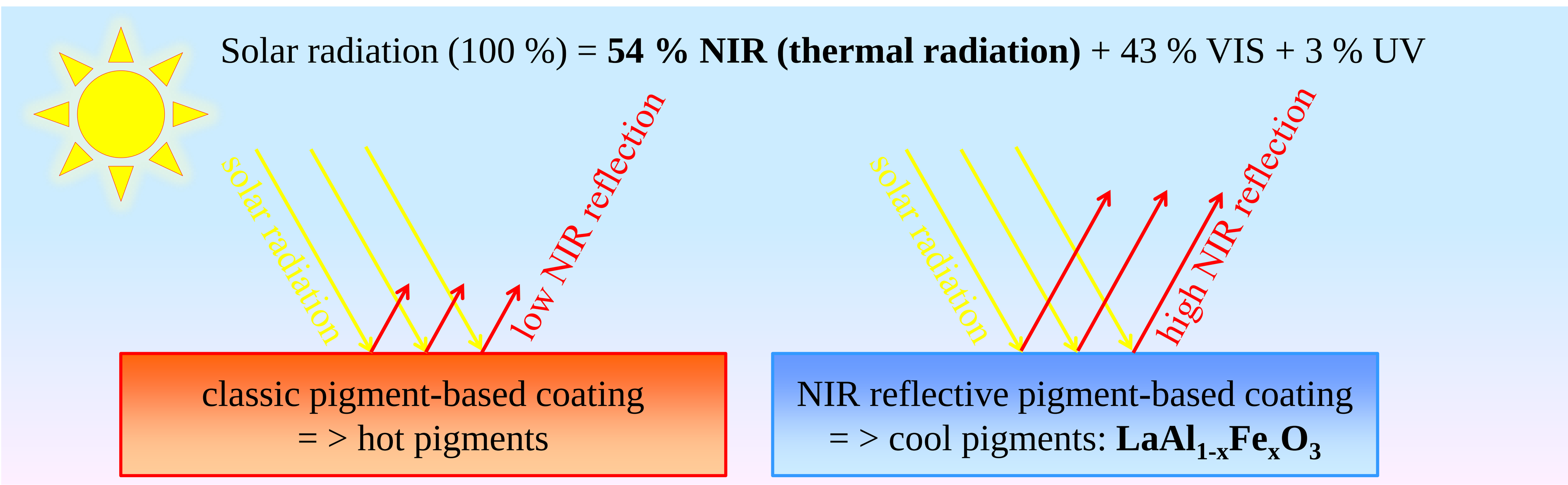
1. Introduction



2. Experimental

OXIDIZERS: $La(NO_3)_3{\cdot}6H_2O$ (Merck) + $Al(NO_3)_3{\cdot}9H_2O$ (Fluka) + $Fe(NO_3)_3{\cdot}9H_2O$ (Sigma-Aldrich)

+

FUELS: CH_4N_2O (Merck) + $C_2H_5NO_2$ (Fluka)

$$6\,La(NO_3)_3 + 10\,C_2H_5NO_2 + 6(1-x)\,Al(NO_3)_3 + 6x\,Fe(NO_3)_3 + 15\,CH_4N_2O = 6\,LaAl_{1-x}Fe_xO_3 + 35\,CO_2 + 55\,H_2O + 38\,N_2$$

Fig. 1. Still images captured during the evolution of a typical combustion reaction (x = 0.75, sample LAF 0.75).

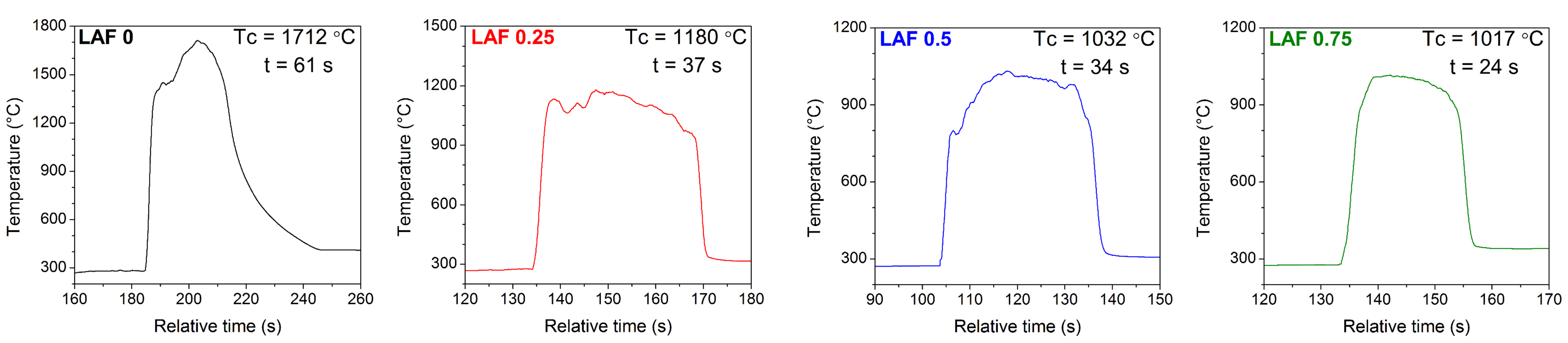


Fig. 2. Temperature variation during combustion reactions.

pigments were used to prepare water-based acrylic paints

paint recipe: 38.7 % acrylic copolymer, 30.0 % pigment, 30.0 % water, 1.3 % additives.
application technique: block applicator, 300 μm gap size.

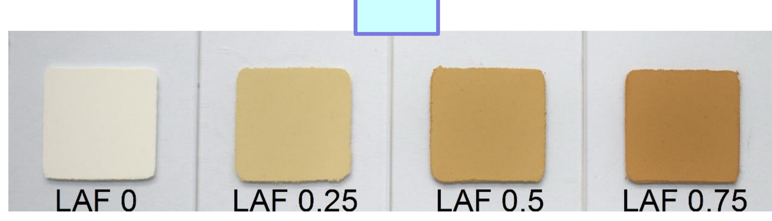


Fig. 3. Images of the resulted pigments.

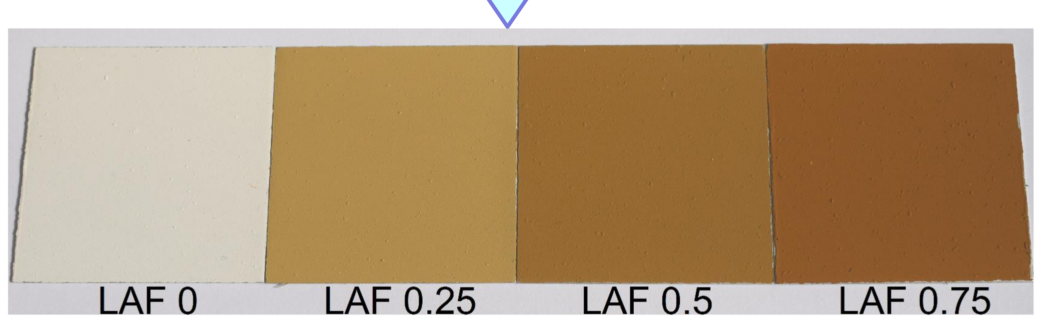
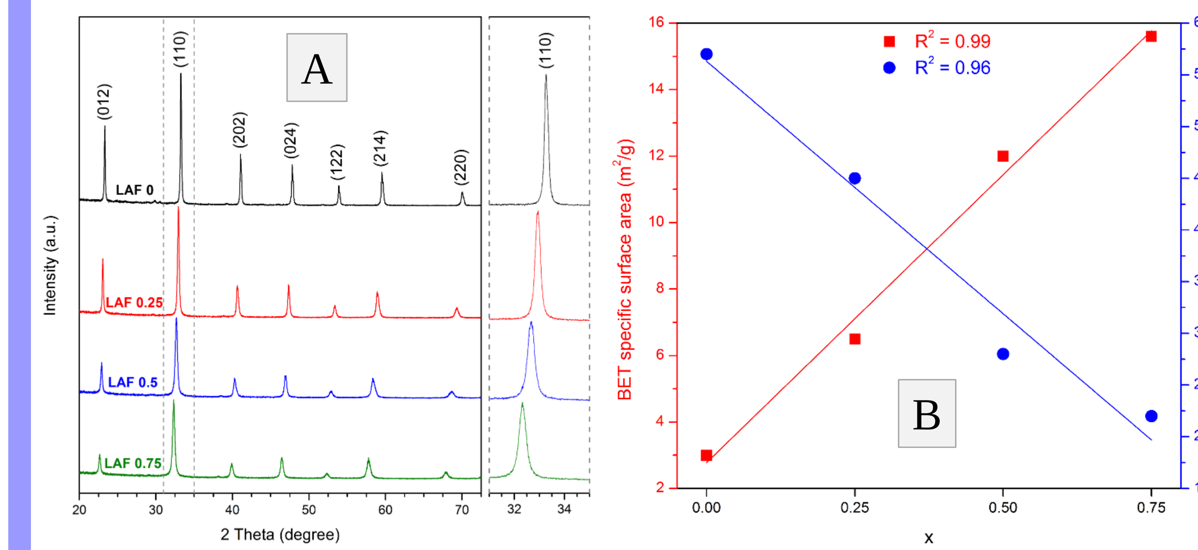


Fig. 4. Images of the coatings applied onto Al sheets (60x60 mm).

Characterization techniques:

- **Thermal imaging**: FLIR T 640, 15 frames/second.
- **XRD**: Rigaku Ultima IV, $Cu_{K\alpha}$.
- **TG-DSC**: Netzsch STA 449, 10 C°/min heating rate, air atmosphere, alumina crucibles.
- **BET**: Micromeritics ASAP 2020, samples degassed at 300 C° and 5 μmHg for 12 hours.
- **TEM**: FEI Titan G2 80–200.
- **DRS**: Perkin Elmer Lambda 950, D65 illuminant, observer's angle 10, CIEL*a*b*, TSR.
- **Coating thickness**: Byko-test 1500 film gauge.

3. Results and Discussion



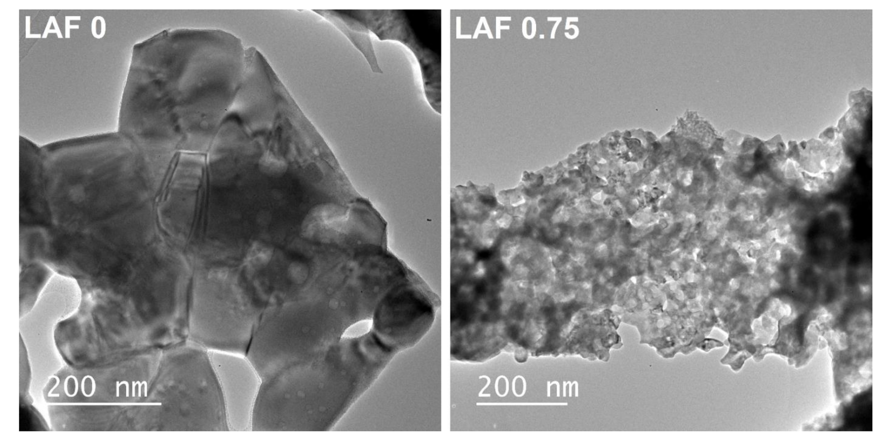


Fig. 6. TEM images of LAF 0 and LAF 0.75.

Fig. 5. XRD patterns (A) of the samples. BET surface area and crystallite size (B) as a function of iron (x) content.

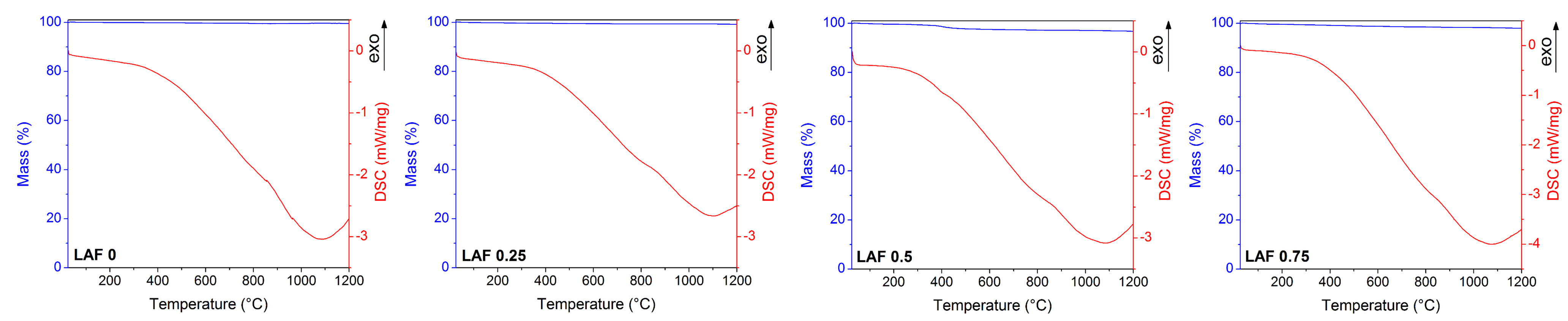


Fig. 7. TG-DSC curves of $LaAl_{1-x}Fe_xO_3$ pigments prepared by solution combustion synthesis.

Table 1. Total solar reflection (TSR) and CIEL*a*b* color evaluation of $LaAl_{1-x}Fe_xO_3$ pigments and coatings.

Sample	Pigment				Acrylic coating (30 wt. % pigment)				Wet / dry coating
	TSR (%)	L*	a*	b*	TSR (%)	L*	a*	b*	thickness (μm)
LAF 0	85.3	98.1	0.3	3.3	87.5	96.5	-0.4	3.1	300 / 94
LAF 0.25	57.3	77.4	1.1	27.1	49.4	69.7	2.7	28.5	300 / 91
LAF 0.5	47.6	65.0	5.0	33.6	38.5	58.1	6.7	32.6	300 / 101
LAF 0.75	42.7	59.0	10.2	39.5	35.8	53.7	12.2	35.1	300 / 99

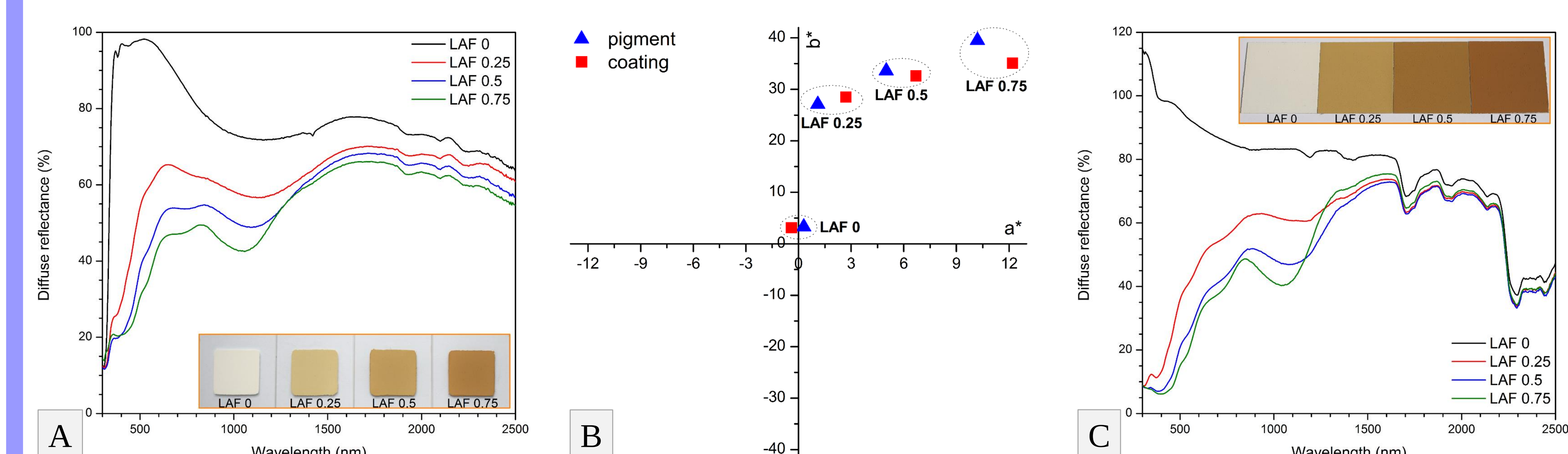


Fig. 8. DRS spectra of the pigments (A) and coatings (C). Colour position in the CIEL*a*b* colour space (B).

4. Conclusions

- NIR-reflective brown pigments based on $LaAl_{1-x}Fe_xO_3$ (x = 0 – 0.75) were prepared by solution combustion synthesis, without any additional thermal treatment.
- As the iron amount increases (0 → 0.75) combustion temperature decreases (1712 → 1017 °C), leading to a decrease of crystallite size (57 → 22 nm) and an increase of BET surface area (3.0 → 15.6 m²/g).
- In terms of colour, the brown shade of the pigments gets more intense for larger iron amounts but the total solar reflectance decreases from 85.3 % to 42.7 %.
- The combustion-synthesized pigments were successfully tested for the preparation of water-based acrylic paints of different brown shades having NIR-reflective capabilities.

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