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**Influence of oxidation of *i*-AlCuFeB particles on the formation of Al-based composites prepared by solid state sintering**

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3 **Influence of oxidation of *i*-AlCuFeB particles on the formation of Al-based composites**  
4 **prepared by solid state sintering**  
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24 We present new results concerning the formation of Al-based composites reinforced  
25 by icosahedral *i*-Al<sub>59</sub>Cu<sub>25.5</sub>Fe<sub>12.5</sub>B<sub>3</sub> quasicrystalline particles. The issue is to limit the diffusion  
26 of Al from the matrix to the quasicrystalline particles during sintering in order to overcome  
27 the transformation of the *i*-phase into the ω-Al<sub>7</sub>Cu<sub>2</sub>Fe tetragonal phase. Here we have  
28 investigated the influence of an oxidation pre-treatment of the quasicrystalline particles onto  
29 the kinetics of this phase transformation by high energy X-ray diffraction. The  
30 quasicrystalline particles were annealed either in vacuum or in air, leading to different  
31 thickness of the oxide layer. We show that the kinetics of Al diffusion is efficiently reduced  
32 by the oxide barrier, enabling the formation of composite materials with no phase  
33 transformation during the sintering process. The dependence of the tribological properties of  
34 these composites is investigated as a function of the volume fraction of *i*-AlCuFeB particles.  
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52 *Keywords:* Quasicrystals; Metal matrix composite; Phase transformation; Tribology  
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## 1. Introduction

The attractive properties of quasicrystals, such as low friction coefficient, low adhesion and high wear resistance [1, 2], could only be used for technological applications in the shape of coatings [3] or reinforcement particles in metal matrix composites (MMC) [4] to circumvent their brittleness. Tsai *et al.* [4] reported an increase of the hardness of aluminium based composites reinforced by *i*-AlCuFe particles and noticed the appearance of the  $\omega$ -Al<sub>7</sub>Cu<sub>2</sub>Fe tetragonal phase during the sintering of the composite at 873 K. In order to preserve the icosahedral phase, Fleury *et al.* [5] made similar MMC by using Ni-coated AlCuFe particles and successfully increased the content of the *i*-phase in the resulting composite materials.

In the present paper, we report an alternative way to preserve the *i*-phase in the aluminium matrix by forming an oxide layer covering the surface of the AlCuFeB particles. We also present the influence of this oxidation pre-treatment onto the kinetics of the phase transformation (*i*-AlCuFeB →  $\omega$ -Al<sub>7</sub>Cu<sub>2</sub>Fe). The icosahedral *i*-Al<sub>59</sub>Cu<sub>25.5</sub>Fe<sub>12.5</sub>B<sub>3</sub> compound was chosen rather than *i*-Al<sub>62</sub>Cu<sub>25.5</sub>Fe<sub>12.5</sub> due to its yield stress, which is three times larger and its lower friction coefficient [6]. The Al-Cu-Fe-B alloy is thus a better candidate for tribological applications than the traditional Al-Cu-Fe system. The results of friction behaviour of sintered composites will also be reported.

## 2. Experimental Procedure

The AlCuFeB particles produced by gas atomization and the commercial Al (99.98 %) powders were sieved with a mesh size of 25  $\mu\text{m}$  and blended in a planetary ball mill under an argon atmosphere at 200  $\text{tr}\cdot\text{min}^{-1}$  for 1 min.

To study the kinetics of the phase transformation, we have prepared two composites by cold compaction (400 Mpa, 300 K). Both contain a volume fraction of *i*-AlCuFeB particles of 15 vol.% that were previously annealed at 873 K for 45 minutes, either in an evacuated tube ( $10^{-5}$  mbar) or in air. This pre-treatment results in almost pure icosahedral phase particles covered by either a thin (a few nm) or a thick (a few tens of nm) oxide layer [7]. In situ measurements by X-ray diffraction were performed on the ID15B beamline at the ESRF (European Synchrotron Radiation Facility, France) in order to quantify the kinetics of the phase transformation (*i*  $\rightarrow$   $\omega$ ) during a thermal cycle similar to the one needed for sintering. Namely, the two composites were heated up to 773 K at 10  $\text{K}\cdot\text{min}^{-1}$  and maintained at this temperature until the phase transformation was completed. The radiation used was a monochromatic beam of 88.67 keV and the  $2\theta$  angles used for analysis were located between 1.00-8.00°.

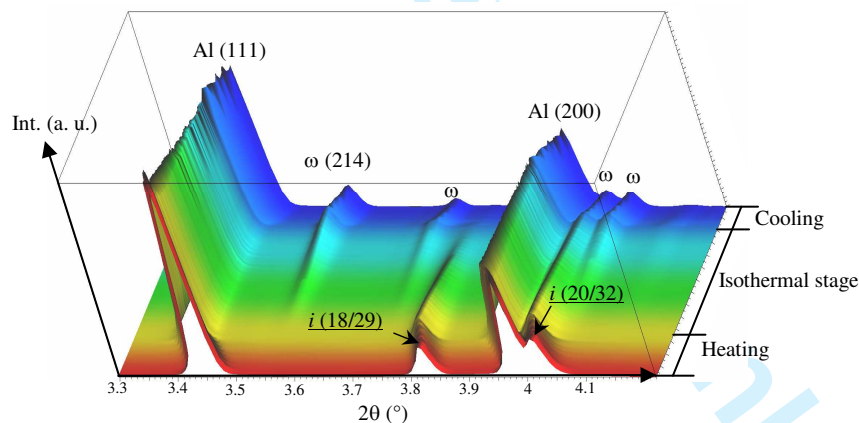
Then, Al/(AlCuFeB) composites were prepared by sintering to study their tribological behaviour. Various volume fractions of AlCuFeB particles (0, 15, 30, 45 and 60 vol.%) have been used, submitted to different oxidizing treatments. The powders were maintained at 773 K in a carbon matrix under low uniaxial pressure of 32MPa and under a helium atmosphere during 3h30. The sintered sample surfaces were mechanically polished (SiC paper in water lubricant, from 500 grit down to 4000 grit). Samples were characterized after sintering by X-ray diffraction (XRD) using a SIEMENS D500 diffractometer (wavelength,  $\lambda_{\text{CoK}\alpha 1}=0.1788965$  nm) and scanning electron microscopy. Tribological properties were investigated with a pin on disk tribometer. All friction tests were carried out at room temperature in ambient atmosphere with a relative humidity of 50 % and under non-lubricated conditions. The

indenter used were 6 mm 100Cr6 hard steel balls, the sliding velocity was  $0.5 \text{ cm.s}^{-1}$  and the normal applied load was 2N. The radius of the trace was 5 mm.

### 3. Results and discussions

#### 3.1. In situ high energy X-ray diffraction

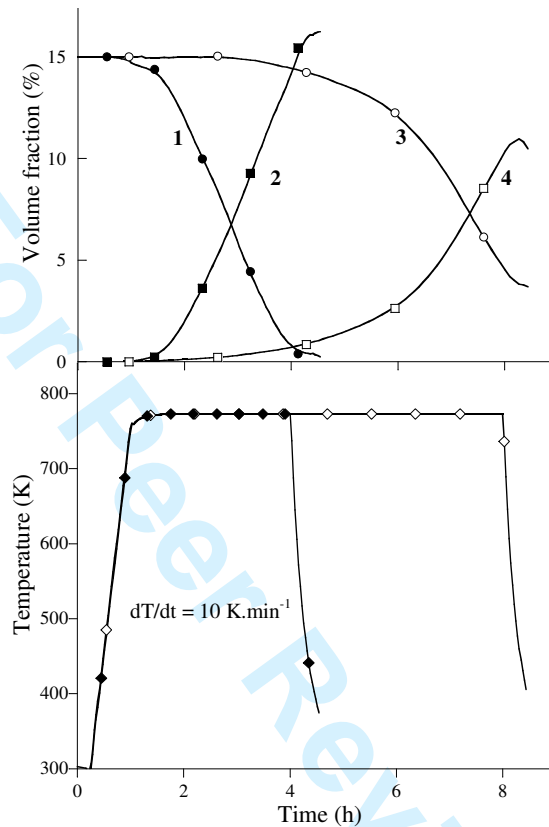
Figure 1 shows a three dimensional representation of the set of diffraction patterns obtained during the heating cycle for the sample containing 15 vol.% of non-oxidized *i*-AlCuFeB particles prepared by cold compaction. Before the heat treatment, we observe that the sample contains only aluminium and *i*-AlCuFeB. The diagram obtained from the composite containing pre-oxidized powders is similar with no additional diffraction peaks, suggesting that the oxide layer is either amorphous or poorly crystallized and very thin. The phase transformation *i*-AlCuFeB  $\rightarrow$   $\omega$ -Al<sub>7</sub>Cu<sub>2</sub>Fe is clearly seen in Fig. 1, with the progressive disappearance of the (18/29) and (20/32) peaks of the *i*-phase during the thermal cycle and the simultaneous appearance of the  $\omega$ -phase.



**Fig. 1.** 3D representation of diffraction patterns obtained during the thermal cycle (heating up to 773 K – holding at 773 K and cooling to room temperature).

Figure 2 presents the evolution of the phase volume fractions during the transformation (*i*  $\rightarrow$   $\omega$ ) for the two composites obtained by cold compaction. The phase fractions are determined using the integrated intensities of the main diffraction peaks for each phase. Absolute values are estimated thanks to calibration using the known initial volume fractions for the *i*-phase/Al composite or by using theoretical intensities calculated from structural data for the  $\omega$ -phase/Al composite resulting from the thermal treatment. The onset

temperature of the phase transformation was determined by the appearance of the first diffraction peak of the  $\omega$  phase.

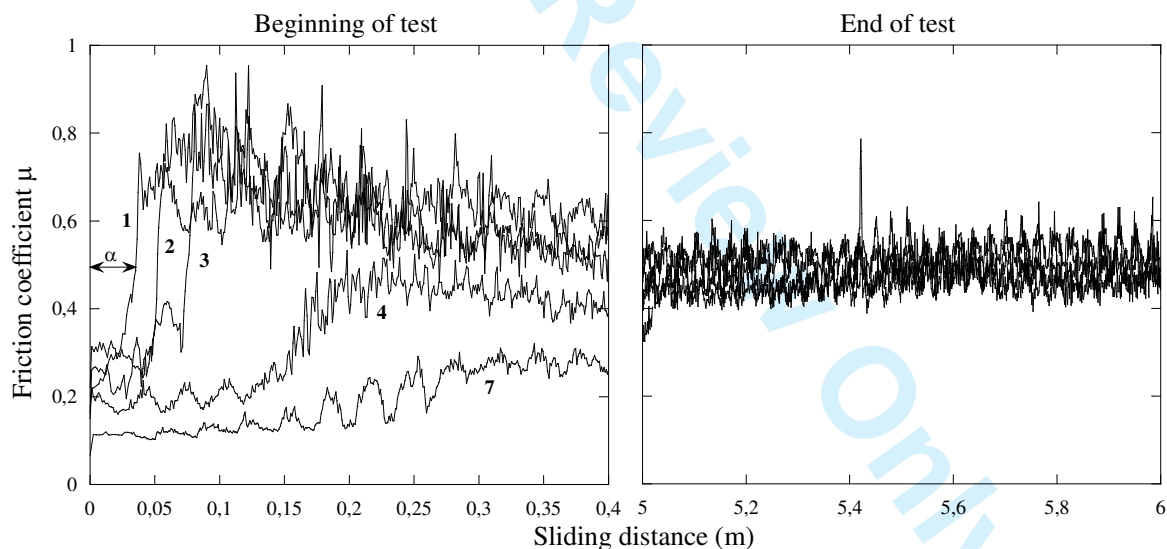


**Fig. 2.** Top: Volume fraction of  $i$  (1 ; 3) and  $\omega$  (2 ; 4) phases for the two composites as a function of time. Solid dots correspond to the composite containing  $i$ -particles oxidized in vacuum for 45 minutes; empty dots correspond to the composite containing  $i$ -particles oxidized in air for 45 minutes. Bottom: Heating cycles used for the two composites.

For non-oxidized AlCuFeB particles (Fig. 2, curves 1 and 2), the formation of the  $\omega$  phase starts at 723 K and the phase fractions evolve very rapidly. The  $i \rightarrow \omega$  transformation is almost completed after three hours at 773 K, which corresponds to the minimum time needed to obtain a sintered composite. On the contrary, the phase transformation is largely inhibited in the case of pre-oxidized AlCuFeB particles (Fig. 2, curves 3 and 4), and the onset temperature is increased to 763 K. This means that the kinetics of Al diffusion is reduced by the oxide barrier and becomes compatible with the sintering process.

### 3.2. Friction behaviour of bulk composites

Using the knowledge gained from the above experiment, several sintered Al/(AlCuFeB) composites were prepared containing different volume fractions and using different oxidation treatments of the AlCuFeB particles, either to prevent or to promote the formation of the  $\omega$ -phase. Table 1 summarizes the main characteristics of the different samples. Figure 3 shows the variation of the friction coefficient for composites containing 15, 30 and 60 vol.% of AlCuFeB particles submitted to different oxidation pre-treatments. For limited sliding distances, composites have a friction coefficient much lower than that of sintered fcc aluminium  $\mu_{Al} > 1$ . However, after five meters of sliding, all coefficients tend towards the same value ( $\mu \cong 0.6$ ) due to the aluminium transfer from the sample to the indenter. As shown in figure 3, we can define  $\alpha$  as the distance before seizing for each composite.

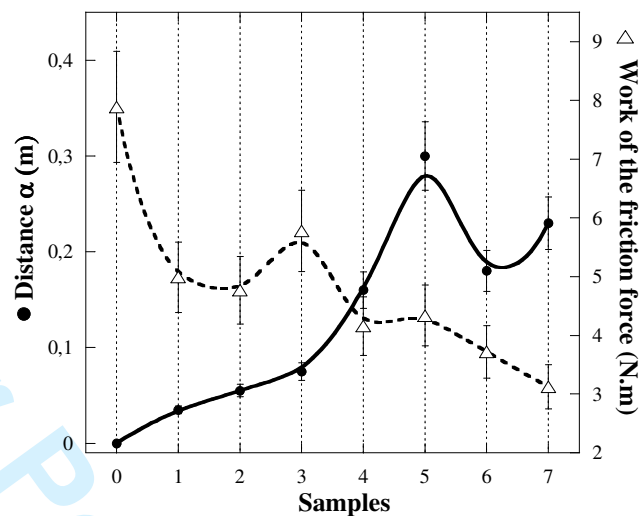


**Fig. 3.** Variation of the friction coefficient for Al/(AlCuFeB) composites with sliding distance. At the end of test, all friction curves merge to the same average value of the coefficient of friction.

The work of the friction force calculated by integrating each friction curve over the first five meters of sliding and the distance  $\alpha$  are plotted for all samples in Fig. 4. The work of the friction force decreases with increasing volume fraction of AlCuFeB particles, while the distance  $\alpha$  increases with the volume fraction and the presence of the  $i$ -phase.



The locally increasing values of  $\alpha$ -distance when the  $i$ -phase is preserved may be explained by the contribution of the low surface adhesion of the  $i$ -phase.



**Fig. 4.** Distance before seizing  $\alpha$  and work of the friction force of the various samples listed in Table 1. Curves serve only to guide the eye.

Table 1  
Summary of the XRD analyses of samples.

Composites N°	Vol.% AlCuFeB	Oxidation Pre-Treatments	XRD before sintering	XRD after sintering
0	0	-	Al	Al
1	15	in vacuum	Al + $i$	Al + $\omega$
2	15	in air for 10h	Al + $i$	Al + $i$ + $\omega$
3	15	in air for 100h	Al + $i$	Al + $i$
4	30	in vacuum	Al + $i$	Al + $\omega$
5	30	in air for 75h	Al + $i$	Al + $i$ + $\omega$
6	45	in vacuum	Al + $i$	Al + $\omega$
7	60	in vacuum	Al + $i$	Al + $\omega$

#### 4. Conclusions

Composites made of icosahedral  $i$ -Al<sub>59</sub>Cu<sub>25.5</sub>Fe<sub>12.5</sub>B<sub>3</sub> quasicrystalline particles dispersed in an aluminium matrix were prepared by solid state sintering with low uniaxial pressure. Oxidation pre-treatments of the quasicrystalline particles prevent the formation of the  $\omega$ -Al<sub>7</sub>Cu<sub>2</sub>Fe tetragonal phase during sintering. Consequently, the oxide layer is an efficient diffusion barrier and the kinetics of Al diffusion becomes compatible with the sintering process. Preliminary friction tests indicate an increase of the sliding distance before seizing when the  $i$ -phase is preserved and a significant decrease of the work of the friction force with increasing volume fraction of AlCuFeB particles, up to 60 vol.%.

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