

Thermal mixing of Al–Fe multilayers

M. Meyer¹, L. Mendoza Zélis², F.H. Sánchez² and A. Traverse^a

*Departamento de Física, Facultad de Ciencias Exactas,
Universidad Nacional de La Plata, Argentina*

*^a Centre de Spectrométrie Nucléaire et de Spectrométrie de Masse,
Université Paris XI, Orsay, France*

Al–Fe multilayers have been mixed by thermal treatment and their evolution followed by conversion electron Mössbauer spectroscopy. The initial and final states have been characterized by Rutherford backscattering spectrometry. The results are compared with those previously obtained in the ion beam mixing of similar systems.

1. Introduction

Ion beam mixing of Al–Fe multilayers produces amorphous, quasicrystalline or crystalline phases depending on the target temperature during irradiation [1]. During this process an effective local temperature, which depends on the target temperature and on the rate of energy deposition, would determine the formed phase. We present here results on the thermal mixing of Al–Fe multilayers made with the aim of determining which phases are formed and how the local order around Fe sites evolves during the process.

2. Experimental

Al–Fe multilayers consisting of four bilayers of 220 Å Al and 35 Å Fe were prepared by electron gun evaporation on a quartz substrate at a base pressure of 1.3×10^{-8} mbar. Additionally 30 Å of Al were deposited on top of the multilayer as a barrier for O penetration. Prior to evaporation iron was enriched in ⁵⁷Fe in order to enhance the Mössbauer absorption. From a total of four samples two were used for the present study (samples I and II).

The multilayers were analyzed by Rutherford scattering of α -particles at 93 and 173 degrees. Also a search for O contamination was made using the ¹⁶O(α , α)O¹⁶ resonance at 3.05 MeV.

¹ Fellow of CONICET.

² Member of CONICET.

The heat treatments were performed in a heating device continuously evacuated by a turbomolecular pump at a pressure lower than 5×10^{-5} mbar.

Conversion electron Mössbauer spectra (CEMS) were obtained in the constant acceleration mode using a $^{57}\text{CoRh}$ source and an electron counter with a constant flow of He-8 at% CH_4 .

3. Results and discussion

3.1. AS-PREPARED MULTILAYERS

The layered structure of the samples is clearly displayed by the RBS spectrum of sample II, mainly in the energy region corresponding to scattering by the Fe atoms (fig. 1). The Mössbauer spectrum of both samples (fig. 2) shows, apart from several magnetic sextets, an important absorption at low velocities, revealing that 13% of the probes are in a paramagnetic environment. Therefore the spectrum was fitted with a superposition of three or four magnetic interactions of varying magnetic field and isomeric shift, plus a quadrupole doublet.

The magnetic interactions could readily be associated with pure Fe and Fe with one or two Al atoms in the first two coordination spheres, as expected for a dilute solid solution of Al in Fe (the mean composition would be about 5 at%). There is, however, an excess population of Fe without non-magnetic neighbors with respect to that expected for a random solid solution.

The quadrupole interaction corresponds to a broad distribution of doublets cen-

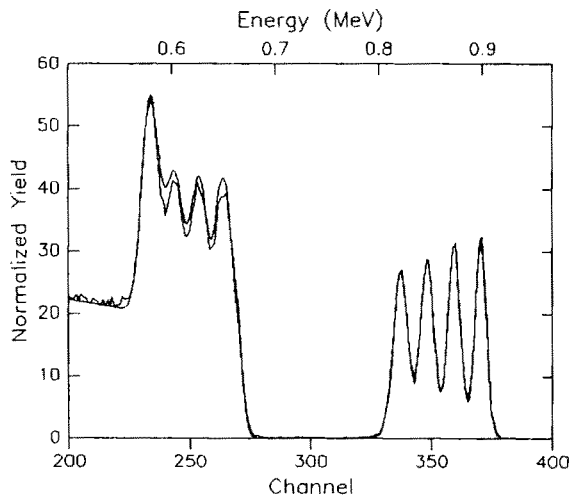


Fig. 1. RBS spectra of the starting multilayer (sample II). The full line represents the simulation described in the text.

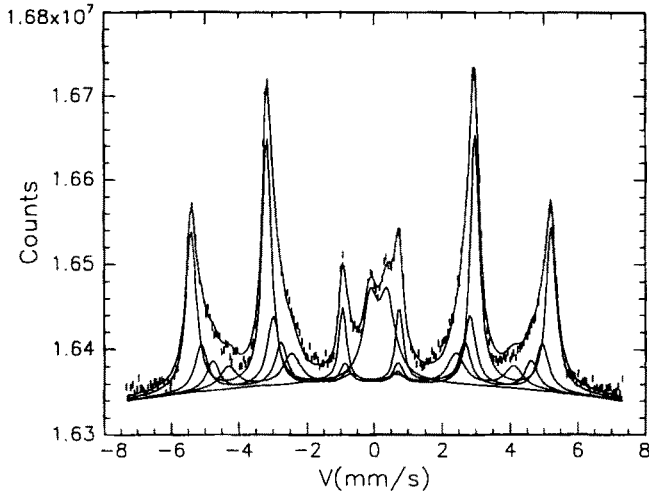


Fig. 2. ME spectra of the starting multilayer (sample II). The resulting fit and the individual components are also shown.

tered at ($QS \approx 0.5$ mm/s, $IS \approx 0.26$ mm/s). It gives no evidence for the formation of Fe aggregates in Al (like dimers or clusters) or some known compounds like Al_6Fe or $Al_{13}Fe_4$ [2]. Its parameters are similar to those obtained in the orthorhombic compound Al_5Fe_2 and in amorphous Al-rich alloys.

This facts were taken into account to simulate the RBS spectra by allowing for the presence of pure Al layers and nearly pure Fe ones separated by regions of an approximate composition: Al_5Fe_2 . The experimental RBS spectrum is very well reproduced in this manner (see fig. 1), resulting in an average Fe concentration of 11.5 at%.

In conclusion, the starting multilayer seems to be partially mixed, with a few percent Al penetrating in Fe on one hand and intermediate disordered regions with 20–30 at% Fe on the other.

3.2. THERMAL MIXING

The samples were thermally treated under the conditions described above, and their evolution observed by CEMS. In the first stages of mixing the spectra could still be interpreted in terms of a dilute solid solution of Al in Fe and a paramagnetic phase. The observed changes in the relative fraction of each magnetic component are consistent with an increase in Al concentration in the dilute alloy region, while the overall fraction of this phase decreases monotonically. Accordingly the paramagnetic phase develops at the interphase. Both the QS and the IS of this phase decrease and the doublet narrows. This phase grows linearly with \sqrt{t} as expected for a diffusion controlled process. From the results of isothermal treatments at

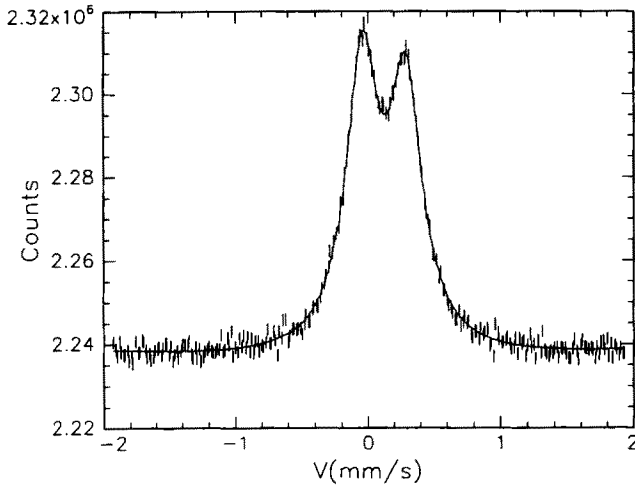


Fig. 3. ME spectra of sample I after 270 min at 550 K.

220°C (sample II) an effective interdiffusion coefficient of about 10^{-15} cm²/s was obtained.

Finally, sample I was sequentially treated at 280°C. All the magnetic sextets collapse in a broad distribution of magnetic fields centered at 305 kOe like that observed in amorphous Fe-metalloid alloys. This phase changes into a paramagnetic one. The IS of this phase remains constant but its QS decreases. After 270 min at this temperature no magnetic interactions could be observed in the spectrum (fig. 3). Accordingly, RBS measurements on this sample show overall compositional uniformity: the Fe concentration decreases slightly from 12.5 at% on top to 10.5 at% on bottom. The results also indicate a slight penetration of Fe in the SiO₂ substrate.

Concerning the paramagnetic phase, as it was already said, the doublet narrows with the thermal treatments (see table 1), and becomes asymmetric. The QS

Table 1

Quadrupole parameters obtained from the ME spectra after several heat treatments.

	Fraction	QS (mm/s)	IS (mm/s)	FWHM (mm/s)
as-made	13 ₂	0.479 ₄₈	0.273 ₂₃	0.698 ₈₆
10 min, 423 K	18 ₂	0.458 ₂₄	0.243 ₁₅	0.499 ₄₁
10 min, 463 K	23 ₃	0.453 ₂₃	0.283 ₁₄	0.485 ₄₁
30 min, 463 K	31 ₄	0.385 ₁₇	0.241 ₁₀	0.446 ₃₂
10 min, 523 K	27 ₃	0.428 ₁₄	0.233 ₈	0.494 ₂₅
30 min, 553 K	41 ₆	0.396 ₆	0.225 ₄	0.411 ₁₂
90 min, 553 K	74 ₅	0.360 ₅	0.236 ₄	0.379 ₈
150 min, 553 K	100	0.332 ₂	0.228 ₁	0.301 ₃
30 min, 623 K	100	0.321 ₂	0.231 ₁	0.291 ₃

decreases monotonically from 0.5 to 0.33 mm/s, and the IS changes from 0.26 to 0.23 mm/s after the first stages of mixing and then remains constant.

The observed quadrupole parameters are alike to those obtained for amorphous and quasicrystalline AlFe phases of similar composition [3], obtained by ion beam mixing of AlFe multilayers at 70 and 520 K respectively. Furthermore, during the thermal mixing our values seem to evolve from "amorphous" to "quasicrystalline" ones. However, it is important to note that practically the same values were observed for metastable Al₆Fe (crystalline) and quasicrystals of the same composition. Nevertheless the observed asymmetry is not consistent with a crystalline compound and would be an indication that a quasicrystalline phase was formed. This needs to be confirmed by X-ray diffraction techniques.

4. Conclusion

Our results are consistent with a diffusion controlled mixing mechanism, through an Al-rich paramagnetic region already present in the starting material.

In the nearly pure Fe regions the Al concentration increases, forming a highly disordered ferromagnetic phase, presumably amorphous, before its transformation to the paramagnetic phase. The latter evolves during the mixing toward metastable AlFe alloys similar to those obtained by ion beam mixing at lower temperatures.

During the mixing at 280°C the formation of quasicrystalline phases would compete with that of crystalline Al₆Fe [4] and probably both phases were formed.

References

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