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Volume-Diffusion of ^{59}Fe in $\text{Al}_{62}\text{Cu}_{25.5}\text{Fe}_{12.5}$ Icosahedral Quasicrystals

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Abstract. The volume diffusion of ^{59}Fe in $\text{Al}_{62}\text{Cu}_{25.5}\text{Fe}_{12.5}$ icosahedral quasicrystals is analyzed using classical diffusion techniques. Over the temperature range from 550°C to 800°C, the volume diffusion coefficients show a classical diffusion behavior and the temperature dependence can be expressed by the following relation:

$$D_v \text{ (m}^2\text{s}^{-1}\text{)} = 4.53 \cdot 10^{-2} \exp - \frac{2.6 \text{ (eV)}}{kT}$$

In the absence of data on a crystalline phase with similar composition, diffusion coefficients are compared to those obtained for Fe in aluminium and in aluminides (FeAl and $\text{Al}_{13}\text{Fe}_4$): no strong peculiarity linked to the quasicrystalline structure appears.

1. INTRODUCTION

Volume-diffusion experiments on quasicrystals are extremely difficult to perform. On the one hand, we have the usual well known difficulties of studying diffusion in aluminides, brittleness, reactivity, absence of a suitable aluminium isotope. On the other hand, there is the difficulty to obtain good quality quasicrystalline samples with a sufficient size.

Recently Kalugin and Katz [1] proposed a possible mechanism for volume self-diffusion in quasicrystals, which could superimpose to the usual vacancy driven mechanism. Their model is based on geometric properties of the quasicrystalline structure (quasiperiodic network is generated by three atomic surfaces; see [2] for AlCuFe). In these materials, collective motions of atoms are expected for chemical species distributed at the peripheries of the atomic surfaces and might lead to long range diffusion. Considering, for instance the atomic structure of AlCuFe icosahedral phase, the acceleration of diffusion is expected especially for copper and above all for aluminium.

This "accelerated" diffusion could be a key point to explain the plasticity at high temperature of quasicrystalline phases. According to mechanical measurements of Bresson and Gratias on AlCuFe [3], a deviation from Arrhenius law might occur at about 660°C (corresponding to brittle-ductile transition). Recently, Nakajima et al.[4] performed self diffusion measurements of manganese in icosahedral AlPdMn . These authors observe a slow diffusion of manganese with diffusion coefficients about two order of magnitude smaller than in the crystalline phase of similar composition. The activation energy deduced for quasicrystal is however relatively small (1.4 eV) which leads (taking into account the values of the diffusion coefficients) to a very small pre-exponential term ($D_0 = 10^{-6} \text{ cm}^2\text{s}^{-1}$). No deviation from Arrhenius law was observed.

This paper reports a study of volume diffusion of an equivalent element (Fe) in an equivalent quasicrystalline phase (icosahedral AlCuFe)

2. EXPERIMENTAL PROCEDURE

$\text{Al}_{62}\text{Cu}_{25.5}\text{Fe}_{12.5}$ flakes were produced by planar flow casting and sintered just below the solidus temperature to produce samples. Compared to samples obtained by classical metallurgy, the sintered

Figure 1 : Diffusion penetration profile (703°C for 3 hours)

ones exhibit less porosity. The samples were 2cm in diameter and 3mm thick. After annealing for 5 days at 1073 K, all the peaks of the X-ray diffraction diagram can be indexed according to the icosahedral scheme with a 6D parameter of 6.31 Å. A few weak peaks indicate the presence of a small amount of cubic Fe-Al type phase. Scanning electron microscopy shows that the cubic phase precipitates are very small and unconnected and should not therefore act as diffusion short circuits. The grain size of the icosahedral phase varies from 5 to 20 μm.

Diffusion coefficients were carried out on the same sample using classical methods: i) electro deposition of ⁵⁹Fe, ii) annealing in high purity argon (6N), iii) mechanical sectioning using a high precision grinding machine which allows the reproducible removal of parallel layers with thicknesses ranging between 0.1 to 10 μm, iv) measurement of the layer activity with a high resolution gamma spectrometer, v) analysis of the penetration profiles with the solution of the Fick equation for an instantaneous source :

$$C(x, t) = C_0 \exp\left(\frac{-x^2}{4Dt}\right)$$

It should be pointed out that: i) before each experiment the sample is preannealed at the diffusion temperature in order to reach thermal equilibrium, ii) autoradiographic analysis is used to check the homogeneity of the deposited film and the absence of contamination.

Table 1: Diffusion of ⁵⁹Fe in AlCuFe icosahedral phase

Temperature (° C)	Diffusion coefficient (cm ² s ⁻¹)	√Dt (μm)
799	5.2 10 ⁻¹⁰	17
752	8.6 10 ⁻¹¹	8
703	3.3 10 ⁻¹¹	6
658	6.0 10 ⁻¹²	4.3
642	3.2 10 ⁻¹²	10
590	6.1 10 ⁻¹³	7.2
568	2.0 10 ⁻¹³	3.2

Figure 2: Temperature dependence of diffusion coefficient (see Table 2 for references)

3. RESULTS AND DISCUSSION

Figure 1 shows a typical penetration profile in standard logarithmic scale; the fitted straight line has a Gaussian behavior. It is to be emphasized that the specific activity range extends over four decades.

The volume diffusion coefficients obtained at each temperature are presented in table 1. They range between $2.0 \cdot 10^{-13} \text{ cm}^2\text{s}^{-1}$ at 568°C to $5.2 \cdot 10^{-10} \text{ cm}^2\text{s}^{-1}$ at 799°C . Figure 2 shows that the temperature dependence of the volume diffusion coefficients follows an Arrhenius behavior. The activation energy and pre-exponential factor deduced from these data are respectively 2.6 eV and $453 \text{ cm}^2 \text{s}^{-1}$.

The "relatively" slow diffusion of iron in icosahedral AlCuFe (with an activation energy of 2.6 eV) is not too surprising. According to Rummel et al.[6], the transition elements (Cr, Mn, Fe, Co) are very slow diffusers in pure aluminium and characterized by pre-exponential factors and activation energies higher than for non transition metallic solutes. This suggests that the interaction between vacancies and transition metals could be strong and repulsive in an aluminium matrix.

Considering self-diffusion in aluminides, it should be mentioned that data are scarce and doubtful, comparisons are thus difficult. However the present diffusion coefficients are comparable to those obtained for AlFe and for $\text{Al}_{13}\text{Fe}_4$ (see figure 2 and table 2).

Therefore, the discrepancy between the self diffusion of iron in AlCuFe ($Q = 2.6 \text{ eV}$; $D_0 = 453 \text{ cm}^2\text{s}^{-1}$) and of manganese in AlPdMn ($Q = 1.4 \text{ eV}$; $D_0 = 10^{-6} \text{ cm}^2\text{s}^{-1}$) remains. It is commonly accepted that the structure of AlPdMn can be modeled using the very same global atomic surface of AlCuFe model [2], by substituting Pd to Cu and Mn to Fe. Nevertheless, the chemical decoration of a few atomic surfaces cells, corresponding to $(12.5 - 8)\%$ of the atoms, must be changed with respect to AlCuFe structure, to fit stoichiometry. These cells, occupied by iron in AlCuFe, are occupied by aluminium in AlPdMn. They are located at the boundaries of the ones of the global atomic surfaces. They are precisely those which are expected to generate the short distance jumps needed for the diffusion process to accelerate. This could lead to a slow self-diffusion of manganese but with a high activation energy.

In the present state of diffusion in quasicrystalline phases, one should also be very careful with experimental processes (quasicrystal quality, diffusion techniques and profiles analysis). It should

Table 2: Results and comparisons

Diffuser	Matrix	Pre-exponential factor ($\text{cm}^2 \text{s}^{-1}$)	Activation energy (eV)	Reference
^{59}Fe	i- $\text{Al}_{62}\text{Cu}_{25.5}\text{Fe}_{12.5}$	453	2.6	our work
^{59}Fe	Al	53	1.9	[5]
^{55}Fe	AlFe (B2, CsCl)	60	2.9	[7]
^{55}Fe	$\text{Al}_{13}\text{Fe}_4$ (monoclinic)	0.001	1.6	[7]
^{54}Mn	i- $\text{Al}_{72}\text{Pd}_{20}\text{Mn}_8$	10^{-6}	1.4	[4]

be stressed that because of different sectioning techniques, the two sets of experiments have been conducted with very different penetration depths. Diffusion profiles are followed over $50 \mu\text{m}$ (and four decades in specific activity) in AlCuFe while they are only followed over less than $1 \mu\text{m}$ (and less than one decade in specific activity) in AlPdMn. Such shallow diffusion profiles may be more sensitive to surface effects often encountered in aluminides.

In conclusion, we have shown that diffusion studies could be performed in quasicrystals with classical methods. These results assess the validity of the experimental procedure which will now be extended to measuring the self-diffusion of aluminium with ^{26}Al isotope.

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References

- [1] P. A. Kalugin and A. Katz, *Europhys. Lett.* **21** (1993), 921–926.
- [2] A. Katz and D. Gratias, to be published in Proceedings of 5th Conference on quasicrystals, Avignon (1995).
- [3] L. Bresson and D. Gratias, *Jour. of Non Cryst. Sol.* **153&154** (1993), 468–472.
- [4] H. Nakajima, J. Asai, K. Nonaka, I. Shinbo, A. P. Tsai, and T. Masumoto, *Phil. Mag. Lett.* **68** (1993), 315–320.
- [5] D. L. Beke, I. Gödény, I. A. Szabó, G. Erdélyi, and F. J. Kedves, *Phil. Mag. A* **55** (1987), 425–443.
- [6] G. Rummel, T. Zumkley, G. Erdélyi, K. Freitag, G.M. Hood, and H. Mehrer, *Def. and Diff. Forum* **95-98** (1993), 715–722.
- [7] L.N. Larikov, V.V. Geinchenko, and V.M. Fal’chenko, Diffusion processes in ordered alloys, (Naukova Dumka Publ., Kiev, 1975).