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VACANCY ELIMINATION IN FeAl ALLOYS WITH B_2 STRUCTURE

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Résumé. — L'élimination des lacunes a été étudiée après trempe dans des alliages FeAl de structure B_2 contenant de 33 à 51 at % d'aluminium. Cette élimination est inhomogène et sa cinétique dépend fortement de la température de trempe.

Abstract. — Vacancy elimination is studied in quenched FeAl alloys containing 33 to 51 at % Al. The process occurs in localized regions in the specimen and the rate depends strongly on the quenching temperature.

Introduction and experimental methods. — It has been shown that B_2 FeAl alloys may retain a high concentration of vacancies after quenching from high temperatures [1, 2]. Vacancy concentration increases with increasing Al content for a given quenching temperature and does not vary monotonically with increasing quenching temperature. The annealing of a quenched sample at temperatures near 400 °C eliminates the quenched vacancies. The elimination of vacancies during annealing has been previously studied in polycrystalline 40 at % Al alloy by resistivity measurements [3]. In the present study, vacancy elimination is followed in single crystal samples with 42 at % to 51 at % Al (total impurities $\leqslant 5 \times 10^{-3}$ %) by dilatometry, microhardness measurements, transmission electron microscopy, field ion microscopy and positron annihilation.

The foils for electron microscopy were cut with an electrolytic saw and thinned down by a standard method. All the samples were heat treated in a vertical furnace and quenched in a quenching medium equivalent to *oil*.

The characteristics of vacancy elimination. — Vacancy elimination was followed during annealing in a differential dilatometer.

As the lattice parameter a is supposed to be constant [4] the concentration of remaining vacancies at an annealing time t is

$$C_v(t) = 3\left[\frac{\Delta L(\infty) - \Delta L(t)}{L_0}\right].$$

Four parameters influence the vacancy elimination in B_2 FeAl alloys : quenching temperature, annealing temperature, Al content and quenching conditions. In these experiments, all the samples were heat treated and quenched in the same conditions so that the influence of this parameter can be ignored. The influence of the three other parameters is studied below.

QUENCHING TEMPERATURE. — The influence of quenching temperature was examined for 42 at % Al samples quenched from various temperatures and annealed at 400 °C. As the initial vacancy concentration is different for the different quenching temperatures, comparison between the curves is facilitated by considering the time $t_{1/2}$ to eliminate half the initial vacancy concentration. It is clear that $t_{1/2}$ increases markedly with decreasing temperature below 1 050 °C and appears relatively insensitive to temperature above 1 050 °C.

A model proposed by Damask, Danielson and Dienes [5] leads to the expression for the vacancy concentration as a fonction of annealing time

$$C_v(t) = C_v(0) (\cosh \alpha t)^{-2/p}.$$

The theoretical curves giving $\log C_v(t)/C_v(0)$ versus time are linear after a certain time.

This model seems only to be applicable to low quenching temperatures [3] as these curves are no longer linear for quenching temperatures above 1 000 °C (figure 1).

ANNEALING TEMPERATURE. — For the same samples (42 at % Al) quenched from 950 °C vacancy elimination was studied for different annealing temperatures from 300 °C to 400 °C. These relatively low annealing temperatures were chosen in order to obtain as low a concentration of residual vacancies as possible after annealing. The vacancy elimination curves (figure 2) show that $t_{1/2}$ increases very strongly with decreasing annealing temperature. No vacancy

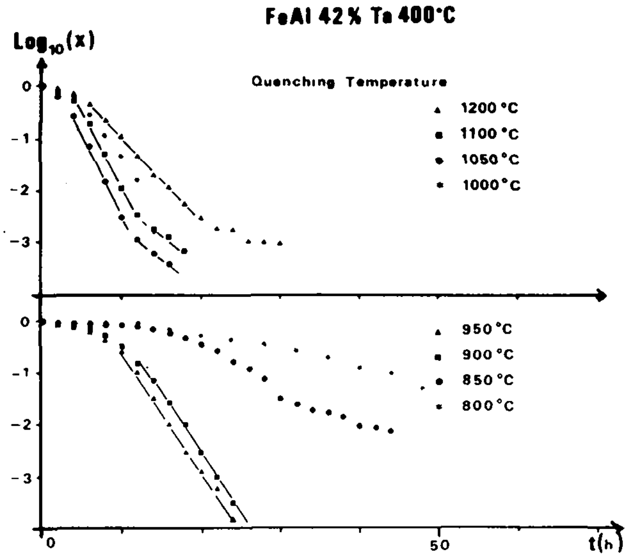


FIG. 1. — Vacancy elimination curves in 42 at % Al alloy during annealing at 400 °C.

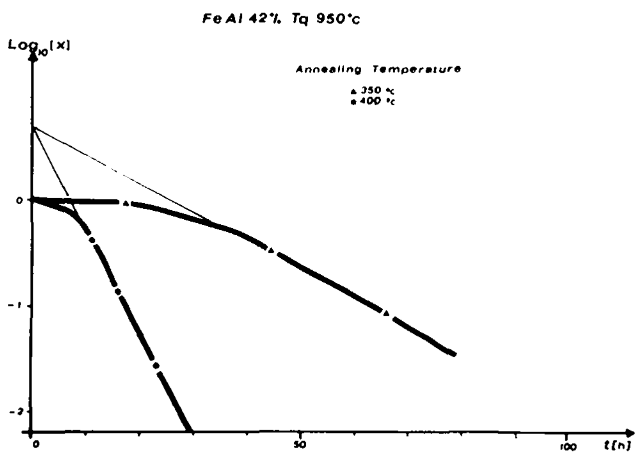


FIG. 2. — Vacancy elimination curves in 42 at % Al alloy after quenching from 950 °C.

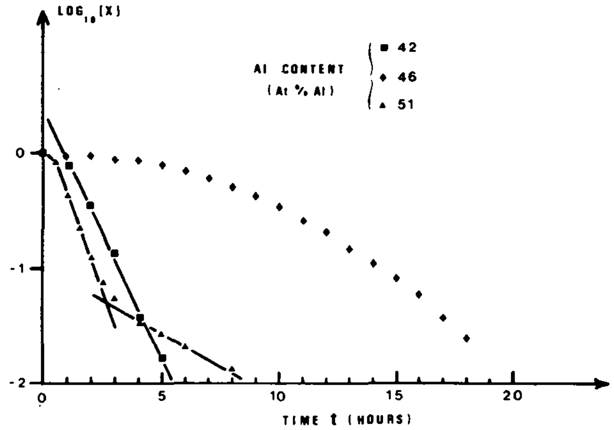


FIG. 3. — Vacancy elimination curves in FeAl alloys for different Al contents (T_q = 950 °C, T_A = 425 °C).

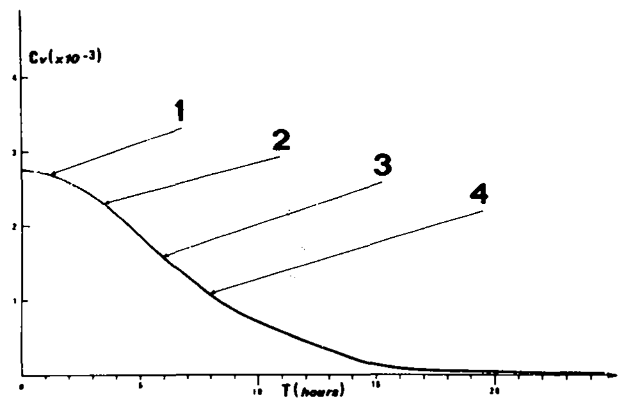


FIG. 4. — Vacancy elimination curve in 42 at % Al alloy (T_q = 1 000 °C, T_A = 400 °C) : Stage 1 : after 75 min. ; Stage 2 : after 3 h 30 ; Stage 3 : after 6 h ; Stage 4 : after 8 h.

elimination was observed at 300 °C. It must be noticed that the parameter p is the same for annealing at 350 °C and at 400 °C (p = 0.89).

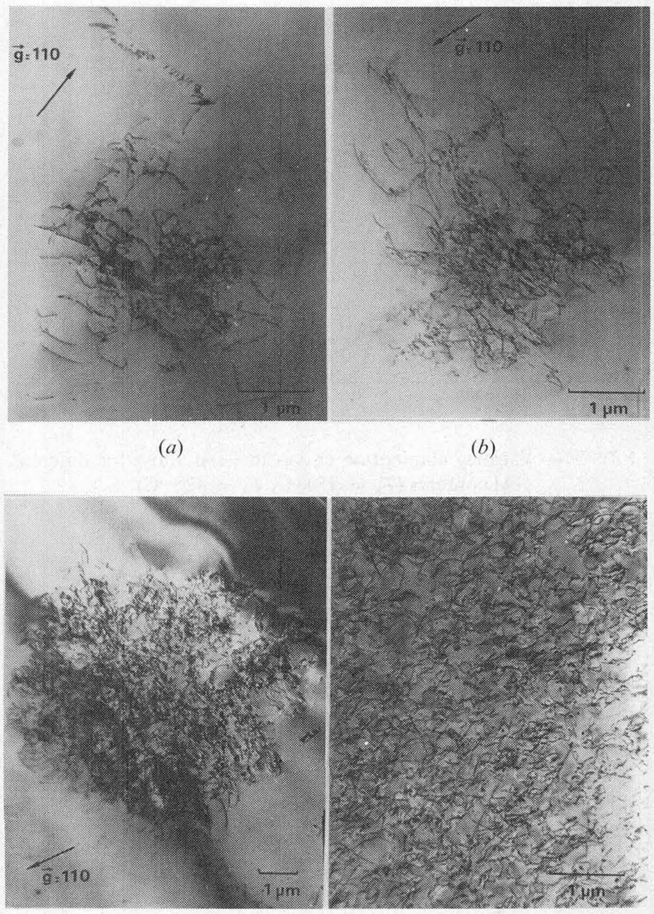
ALUMINIUM CONTENT. — The influence of aluminium content has been examined on samples quenched from 1 000 °C and annealed at 425 °C. The curves (figure 3) show that for high Al content the model is inapplicable. It therefore seems apply to low vacancy concentrations and low quenching temperatures.

Electron microscopy observations. — The structure of a 42 at % Al alloy has been studied by transmission electron microscopy after quenching from 1 000 °C and at different stages during ageing at 400 °C. These stages are denoted 1, 2, 3, 4 in figure 4. The foil plane was (001).

Very few straight dislocations have been observed in the as quenched alloy. After ageing 75 min. (stage 1) the structure of the alloy is inhomogeneous. In some areas, localized regions containing high densities of dislocations can be seen. The rest of the specimen is free of dislocations (figure 5*a*, *b*, *c*). The volume of these areas increases progressively from stage 1 to stage 4, where the foil is full of tangled dislocations (figure 5*d*). In the initial stages, tangled dislocations are observed in the center of the areas (figure 5*b*) and numerous dislocation loops along the edges (figure 6). These dislocation loops are not entirely contained in the foil. The dislocation loops show a fringe contrast when imaged using a superlattice reflection under two beam conditions, and a residual fringe contrast when imaged using a fundamental reflection under two beam conditions (figure 7*a*, *b*). Such a contrast is typical of antiphase boundaries inclined to the plane of the foil.

Inside some loops, *dendritic* dislocations can be seen. Similar dislocations have been observed in β brass [6]. *Dendritic* dislocations give rise to the disappearance of the fringe contrast.

It can be shown that these loops lie on { 111 } planes. This is verified by observation of a (111) foil. Figure 8 shows a well developed loop with a dendritic dislocation inside. This peculiar configuration could be



(*a*) (*b*)

(*c*) (*d*)

FIG. 5. — Micrographs corresponding to the 4 stages of figure 4 : 1 in (*a*), 2 in (*b*), 3 in (*c*), 4 in (*d*). The foil plane was (001).

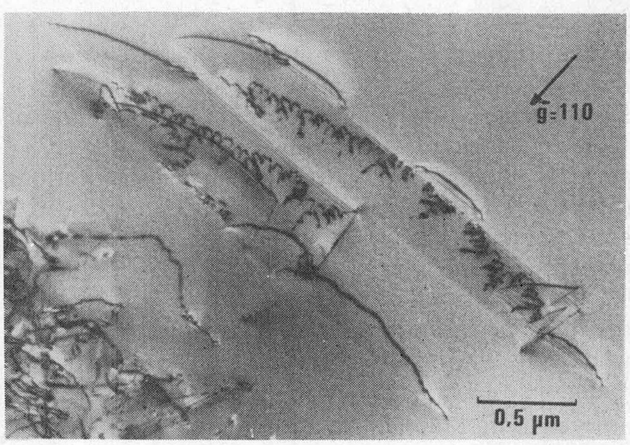
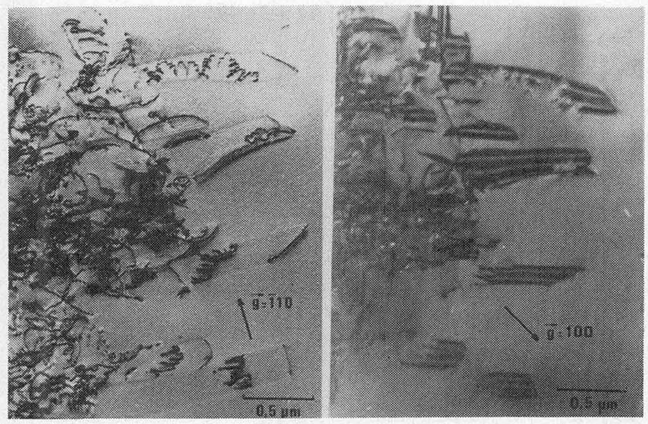


FIG. 6. — Micrograph of dislocation loops observed in stage 2 (figure 4). The loops were imaged using a fundamental reflection under two beam conditions. The foil plane was (001).

explained as follows : a dislocation is first formed by clustering of vacancies on a (111) plane, but the condensation of a single layer of vacancies generates an antiphase boundary ; the clustering of vacancies on the neighbouring plane leads to the creation of another



(*a*) (*b*)

FIG. 7. — Micrographs of dislocation loops observed in stage 2 (figure 4). The loops were imaged under two beam conditions using a fundamental reflection in (*a*), and a superlattice reflection in (*b*). The foil plane was (001).

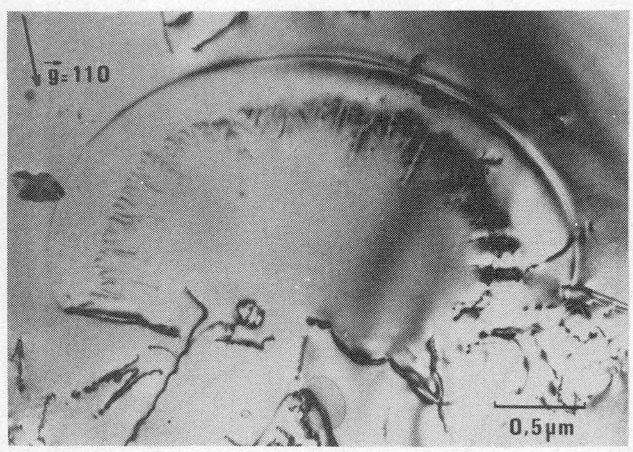


FIG. 8. — Micrograph of a well developed dislocation loop observed in stage 3 (figure 4). The loop was imaged in the same conditions as in figure 6. The foil plane was (111).

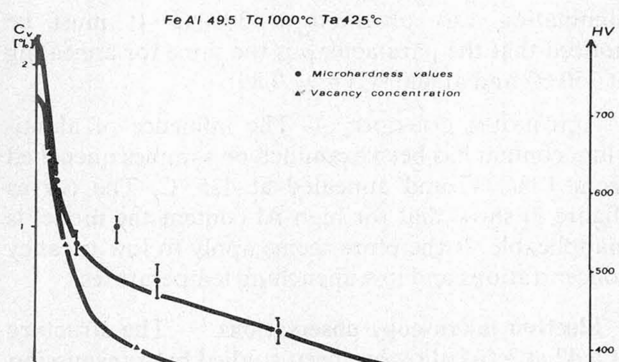
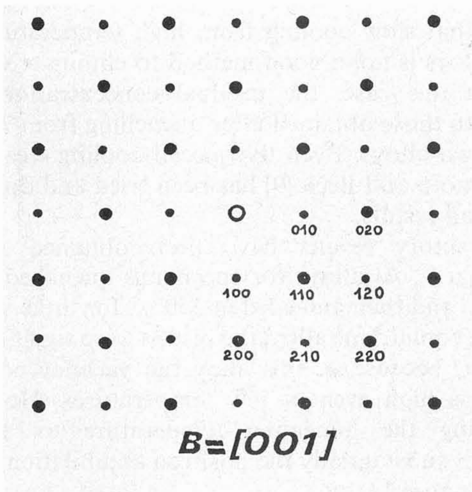
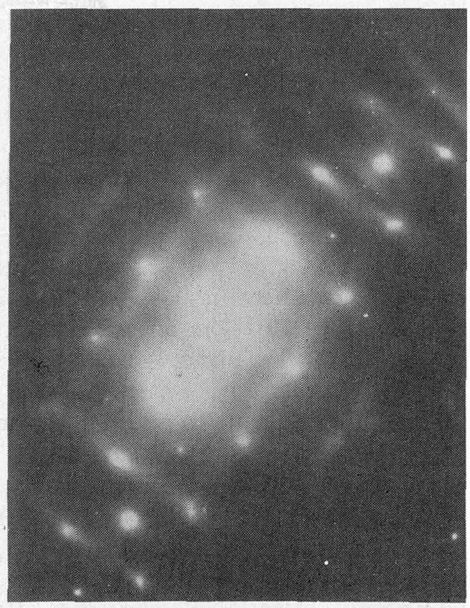


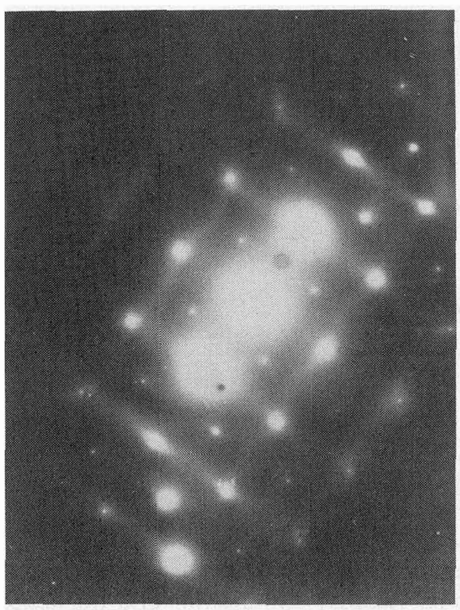
FIG. 9. — Vacancy elimination and microhardness decreasing in 49.5 at % Al alloy (T_q = 1 000 °C, T_A = 425 °C).

dislocation (the dendritic dislocation) which sweeps out the antiphase boundary.

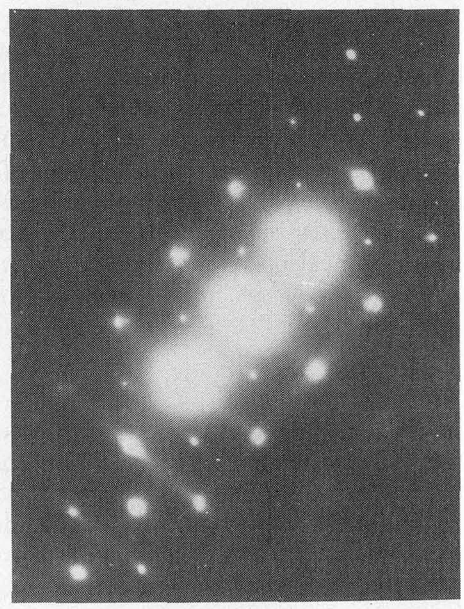




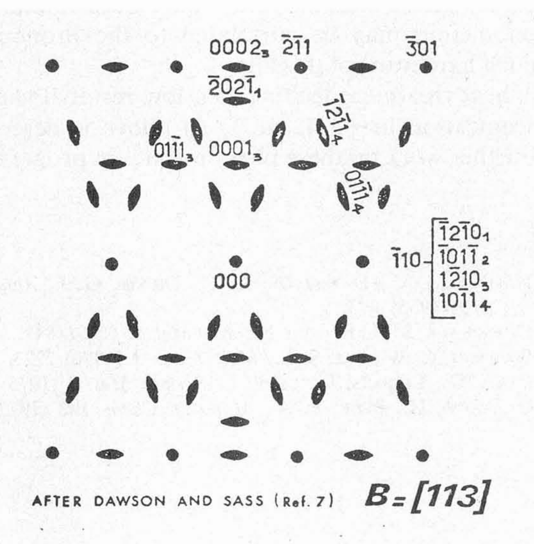
(a)

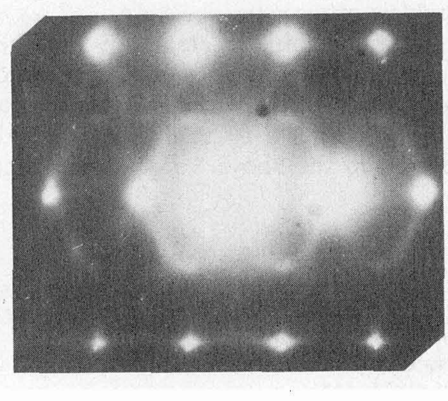


(b)



(d)





(c)

FIG. 10. — Electron diffraction patterns of a Fe 49 at % Al alloy (T_q = 1 000 °C in (a), 800 °C in (b) and 1 150 °C in (c), T_q = 1 000 °C, T_A = 438 °C 24 h in (d)). Diffuse intensities are observed between the lattice reflections.

Complementary methods. — A good correlation can be found between the vacancy concentration after quenching and the hardening observed on quenched FeAl alloys [1]. The results of the investigation on a sample quenched from 1 000 °C and annealed at 425 °C are shown in figure 9. The decrease in hardness fits in quite well with the elimination of vacancies, except, perhaps, for the lower vacancy concentrations.

The electron diffraction patterns of the quenched alloys exhibit diffuse intensities between the reciprocal lattice points (figure 10). The diffuse intensities are similar to these of the diffuse ω phase observed in Zr 22 wgt % Nb alloy [7]. For this reason, these diffuse intensities are thought to be due to the presence of a diffuse ω phase in the b.c.c. lattice.

The shape of the diffuse scattering varies with the vacancy concentration. This diffuse ω phase is thought to be responsible for the hardening mentionned above.

Field ion microscopy was performed on samples both, immediatly after quenching and after full annealing at 425 °C. Only iron atoms can be imaged by this technique in the FeAl alloys. Thus the vacancy sites imaged on the Fe sublattice can be either quenched — in vacancies, field-induced vacancies or Al antistructure atoms [8]. It is assumed that the number of field induced vacancies is the same in quenched and annealed samples. Let d be the total number of vacancies eliminated during the annealing and m the difference of vacant sites on the iron sublattice between quenched and annealed samples. Then it can be shown that the difference Δu of Fe antistructure atoms is

$$\Delta u = m - \frac{d}{2}.$$

Measurements were performed for three compositions (46, 49.5 and 51 at % Al) with samples quenched from 1 000 °C; Δu was found to be respectively 0, 2.5 $^{\circ}/_{00}$ and 4.5 $^{\circ}/_{00}$; it is therefore concluded that for high Al contents, vacancy elimination is concomitant with a small improvement in the degree of order.

The problem of the reference state. — Low vacancy concentration in simple metals can be studied by positron annihilation down to concentrations of 10^{-7}. The angular correlation of the annihilation radiation method has been used to study the residual defect concentration in the above FeAl alloys. It has been shown that slow cooling from high temperatures of these alloys is not a good method to eliminate vacancies; in this case, the residual concentrations are similar to those obtained after quenching from 700 °C in the two alloys. Even the special cooling treatment of Okamoto and Beck [9] has been tried and does not give good results.

Satisfactory results have been obtained in 33 and 42 at % Al alloys for specimens quenched from 1 200 °C and then annealed at 350 °C for three weeks.

In the equiatomic alloy, the results were significantly different, because in this alloy the vacancy concentration is high even at low temperatures. However decreasing the annealing temperature to 350 °C decreases substantially the positron annihilation parameters (figure 11).

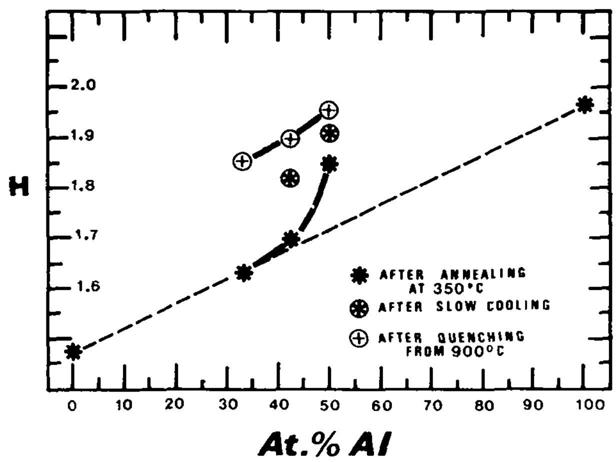


FIG. 11. — Angular correlation of positron annihilation : normalize peak counts H for different alloys and heat treatments.

Conclusion. — It has been shown that, during annealing, vacancy elimination in a quenched B_2 Fe 42 at % Al alloy occurs in localized regions by clustering of vacancies on { 111 } planes.

In Fe 49 at % Al alloy, diffuse intensities in electron diffraction patterns similar to those observed from the diffuse ω phase in ZrNb alloys have been found. This phenomenon may be correlated to the pronounced quench hardening of this alloy.

A heat treatment leading to a low residual vacancy concentration in Fe 42 at % Al alloys is described.

Further work on these phenomena is in progress.

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