

V. SUMMARY

The addition of foreign atoms into nominally pure Al produces changes in the recovery of the residual electrical resistivity following low-temperature irradiation with either electrons or neutrons. These changes, in conjunction with the observations of other experiments, have given some indication as to the nature of the interaction of solute atoms with interstitials.

The addition of solute atoms in pure Al causes a suppression of the recovery normally observed in the pure material. This suppression is about 20–27% of the recovery observed in the pure material and appears to take place mainly in a temperature region above 35°K.

Stage II_A (60–100°K) is a region in which defect identification is uncertain. Some evidence leads to the conclusion that an intrinsic defect migrates in this region while other data appear to indicate that impurity detrapping is the prominent process. The bulk of stage II, however, is rather clearly due to the release of interstitials, trapped during migration in stage I, from impurity-trapping sites. Based on the number of

substages observed for different solute atom additions by several investigators, it appears that, in Al, “electronic effects” are more important in determining the strength of the solute atom interstitial interaction than are “size effects” and that the ionic radius may be a reasonably valuable factor to use when correlating the interaction to some atomic parameter. However, there are still some unresolved discrepancies in the results of different investigators, and more work will be needed in order to justify the above generalities.

High effective frequency factors and varying measured energies may be accounted for with an impurity-trapping model. Thus the energies measured represent the sum of the actual migration energy of interstitials plus some fraction of a binding energy of interstitials to an impurity or impurity-interstitial cluster. The fractional order of reaction which was observed is understood to indicate a complex annealing process.

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Recovery of Electron-Irradiated Aluminum and Aluminum Alloys. III. Stage III

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The recovery of the residual electrical resistivities of pure Al and the Al alloys (nominally 0.1 at.%) Al-Mg, Al-Ga, and Al-Ag have been investigated following 1-MeV electron irradiation near 4°K. Analysis of the data for nominally pure Al in stage III (170–300°K) discloses substructure. (A very small substage occurs at lower temperatures and appears to be influenced by the residual impurities.) The main portion of the stage shows a variation in the observed activation energy with purity: The higher the purity of the “as-received” material, the lower the observed activation energy. A range of 0.46 to 0.59 eV was found for the materials investigated. The effective frequency factor for these materials showed a concurrent systematic variation. These effects are believed to be due to the interaction of migrating defects with residual impurities in the material. The recovery of the alloys in the stage-III region is more complex than in the pure material. There is more substructure in the recovery spectrum of the alloys, and the observed activation energies in stage III are higher. Al-Ag showed a resistivity decrease followed by an increase which is attributed to the clustering of Ag atoms. The observations are interpreted in terms of interstitial migration, restricted by impurities, in the earlier portion of stage III; vacancy migration becomes important in the latter portion of stage III. Stated differently, interstitials migrate in stage III and vacancies migrate in stage IV, but in Al these stages overlap appreciably.

I. INTRODUCTION

THE role of impurities in stage-III recovery in metals remains unclear, and some apparently contradictory results are present in the literature,^{1–8}

despite rather extensive investigation. At one extreme, the study of Sosin and Rachal³ of the recovery of

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¹ T. H. Blewitt, R. R. Coltman, C. E. Klabunde, and T. S. Noggle, *J. Appl. Phys.* **28**, 639 (1957).

² D. G. Martin, *Phil. Mag.* **6**, 839 (1961); **7**, 803 (1962).

³ A. Sosin and L. H. Rachal, *Phys. Rev.* **130**, 2238 (1963).

⁴ S. Ceresara, T. Federighi, and F. Pieragostini, *Phys. Letters* **6**, 152 (1963).

⁵ S. Ceresara, T. Federighi, and F. Pieragostini, *Phil. Mag.* **10**, 893 (1964).

⁶ C. L. Snead and P. E. Shearin, *Phys. Rev.* **140**, A1781 (1965).

⁷ F. Dworschak and J. S. Koehler, *Phys. Rev.* **140**, A941 (1965).

⁸ D. A. Grenning and J. S. Koehler, *Phys. Rev.* **144**, 439 (1966).

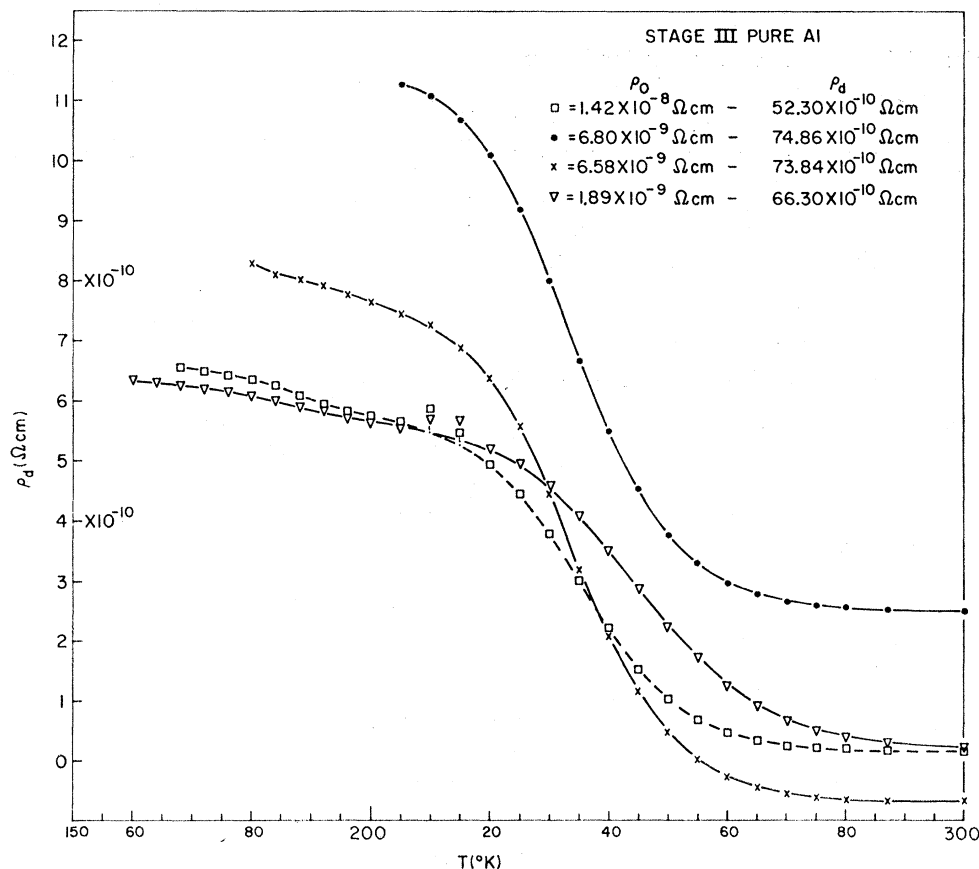


FIG. 1. Isochronal plot of stage III for Al of various purities. The higher the purity of the samples, the lower the value of the residual resistivity (ρ_0). Solid lines are for samples irradiated and annealed simultaneously. The dashed line (square symbol) is for the pure Al sample used with the alloys. The data for the sample with $\rho_0 = 6.58 \times 10^{-9} \Omega \text{ cm}$ should be viewed with caution. Post-experiment investigation clearly indicated inadvertent deformation. The cause of the failure of the sample with $\rho_0 = 6.80 \times 10^{-9} \Omega \text{ cm}$ to fully recover at 300°K is unknown.

Al-0.1 at. % Zn after low-temperature electron irradiation showed that stage III in this alloy and in pure Al were basically the same; that is, stage III was found to be second order in nature and characterized by the same activation energy in both cases. The differences in the temperature at which the recovery rate was a maximum could be accounted for by the different defect concentration in the two cases. Toward the other extreme, Martin² has reported that stage III becomes broadened and almost indistinguishable in Cu alloyed with ~ 0.82 at. % Ag or with ~ 0.5 at. % Cd.

The purpose of this paper is to examine in some depth the role of impurities in aluminum on stage III. As in the preceding paper⁹ (hereafter designated as II), the work has consisted of two parts. In one portion, dilute alloys, consisting of about 0.1 at. % Mg, Ag, or Ga, were investigated; in the other portion, nominally pure samples were used. In the latter case, however, the residual electrical resistivity of the samples differed

among each other, presumably indicating somewhat varying impurity content. Even these rather slight variations give rise to significantly different recovery characteristics.

An examination of the role of impurities in stage III is motivated by two primary purposes. First, it is to be hoped that such an examination would provide needed information concerning the nature of the interaction between selected solute atoms and the defects which are mobile in stage III. A full description of the nature of the interaction can be realized only when the identification of these defects is definitely established. This identification, using foreign atoms as atomic probes, is sought as a second purpose.

II. EXPERIMENTAL CONSIDERATIONS

All of the experimental methods which were used in this work are discussed in Paper II. In most instances, the same samples were used to investigate stage III as well as stage II. The methods of analysis, developed in Paper II, are employed here also.

⁹ K. R. Garr and A. Sosin, preceding paper, Phys. Rev. **162**, 669 (1967).

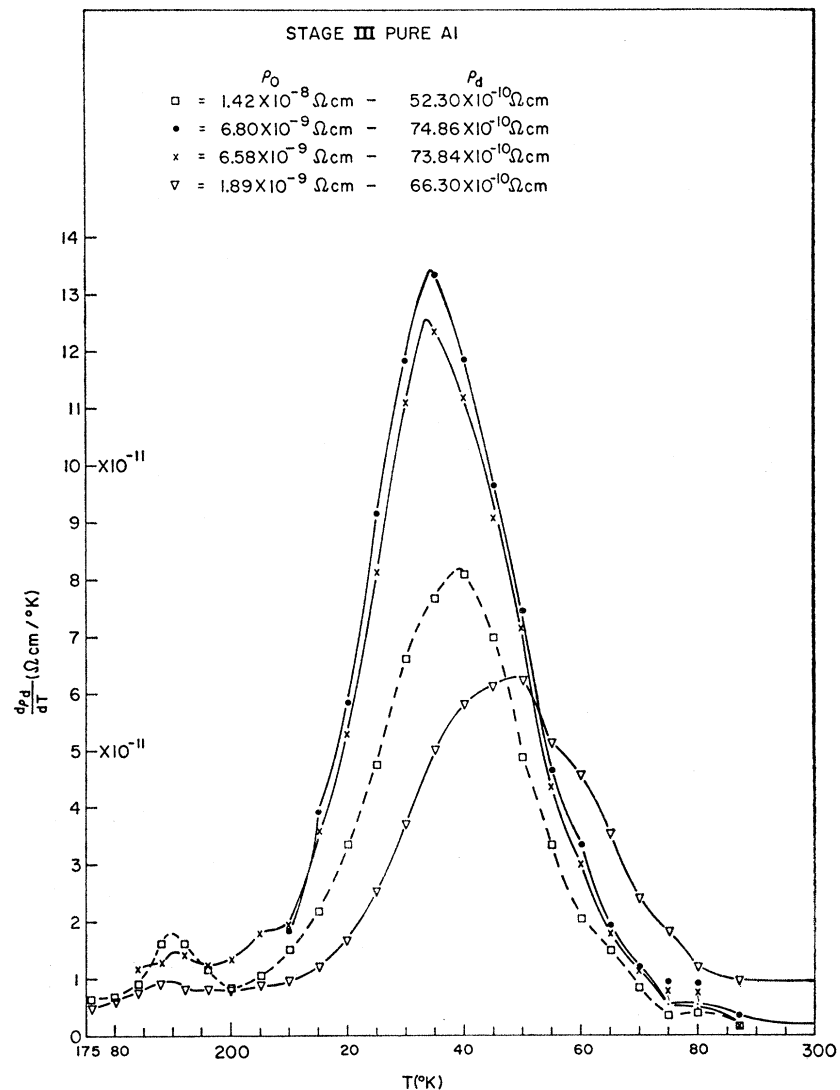


FIG. 2. Derivative plot of the isochronal shown in Fig. 1.

III. RESULTS

A. Pure Aluminum

The results of the recovery in stage III for pure Al are shown in Fig. 1. The different purities (as well as differences due to annealing treatment) are indicated by the values of ρ_0 , as mentioned earlier.⁹ The pure Al sample which was used with the alloys is shown as a dashed line (square symbol) and is included for easier comparison. The main features are more evident in the derivative plot, shown in Fig. 2. A small substage centered at 190°K is seen to decrease as the purity of the sample increases.

Figure 3 shows the variation in the activation energy of stage III with variation in ρ_0 . The data were analyzed assuming second order kinetics for all samples except the 4N5 pure Al sample used with the alloys. The kinetics for this latter sample were specifically investi-

gated during this study. The isothermal recovery curve for this pure Al sample is shown in Fig. 4. Analysis of the data using Eq. (II4)¹⁰ resulted in a value of $\gamma=2.07$ (see Fig. 5); a plot of $1/(\Delta\rho)$ versus t (time) shows that the data fit well to second-order kinetics (see Fig. 6). This analysis is in agreement with the results of Sosin and Rachal³ who found second-order kinetics in stage III using 99.9999% (6N) pure Al. These results serve as a basis for using second-order kinetics for the activation energy analysis of the other pure Al samples investigated.

The energy of the 4N5 pure Al sample used with the alloy was also calculated by the Meehan-Brinkman (MB) method [Eq. (II6)]. The results of the energy determination for this sample are shown in Fig. 7. We note the good agreement between the MB method and

¹⁰ This notation means Eq. (4) of Paper II.

TABLE I. Summary of results of stage-III annealing.^a

Sample	Temperature of stage (°K)	Iso-thermal temperature (°K)	Order of reaction (γ)	Activation energy E_e isochronal method (eV)	Temperature range of E_e (°K)	Activation energy E_e MB method (eV)	Temperature range of E_e (°K)	Effective frequency factor ^b ν_e
Al 0.06 ₀ at. % Mg	235	230	2.1 ₅	0.63±0.03	220-260	0.65±0.03	230-255	2.9×10 ¹⁸
Al 0.08 ₅ at. % Ga	210	230	2.3 ₀	0.61±0.03	230-260	0.64±0.03	230-260	1.2×10 ¹⁸
Al, pure:								
$\rho_0 = 1.42 \times 10^{-8} \Omega \text{ cm}$	240	230	2.0 ₀ -2.0 ₇	0.59±0.03	225-260	0.61±0.03	235-260	6.0×10 ¹⁵
$\rho_0 = 6.80 \times 10^{-9} \Omega \text{ cm}$	234	0.53±0.03 ^a	220-255	2.0×10 ¹⁴
$\rho_0 = 6.58 \times 10^{-9} \Omega \text{ cm}$	233	0.55±0.03 ^a	220-255	2.0×10 ¹⁵
$\rho_0 = 1.89 \times 10^{-9} \Omega \text{ cm}$	248	0.46±0.03 ^a	220-255	2.0×10 ¹²

^a Calculated assuming second-order kinetics.

^b Value calculated from isochronal method of energy determination.

the approach using Eq. (II5). The isochronal plot used for the MB analysis is shown in Fig. 8.

The effective activation energy for the various samples as seen in Fig. 3 and summarized in Table I is seen to vary in a systematic manner. As the residual resistivity of the material decreases, the energy and the effective frequency factor both decrease. The temperature range over which the activation energy is most representative is, however, not grossly affected by these changes, although the character of the recovery is altered. These variations among the different samples of nominally pure aluminum are consistent with

Eqs. (II12) and (II13) and as shown in Fig. 9. Thus the main conclusion to be reached is that the residual impurities in nominally pure aluminum play a significant role in stage III, particularly in the determination of the activation energy. Simson and Sizmann¹¹ have observed similar effects in the recovery of nickel of different purities following plastic deformation.

B. Alloys

The isochronal recovery plot for the alloys is shown in Fig. 10. As is readily seen from the figure, the alloys retained approximately twice the amount of resistivity

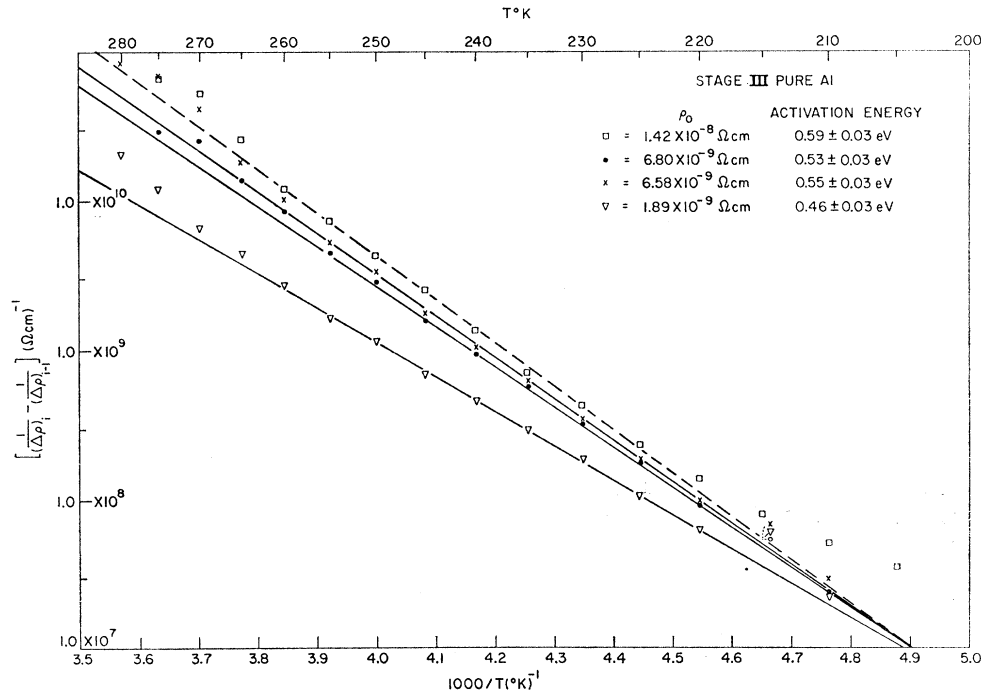


FIG. 3. Determination of the activation energies for Al of various purities using the $1/\Delta\rho$ method. The solid lines are for samples irradiated and annealed simultaneously. Second-order kinetics was assumed in the analysis of these samples. The dashed line (square symbol) is for the pure Al sample used with the alloys. Second-order kinetics was determined for this latter sample during this investigation.

¹¹ P. Simson and R. Sizmann, Z. Naturforsch 17a, 596 (1962).

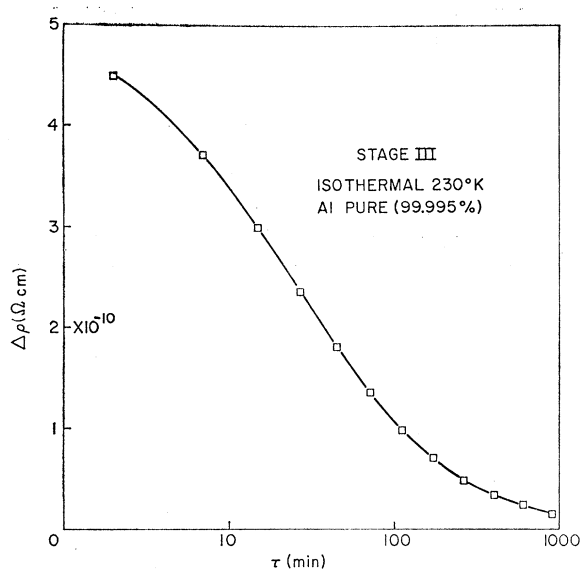


FIG. 4. The 230°K isothermal curve for the pure Al sample irradiated concurrently with the alloy samples.

increment of the pure sample at 170°K. Figure 11 shows the derivative of the isochronal plot shown in Fig. 10. From the derivative plot, it appears that the recovery in the alloys is somewhat more complex than in the pure Al. There appears to be appreciable substructure, particularly in the lower temperature region (i.e., below 230°K).

As may be seen from Table I, the energies for the two alloys, Al-Mg and Al-Ga, in stage III are not significantly higher, considering the increased concentration of impurity atoms, than the energy in the nominally pure material used in their fabrication. The temperature

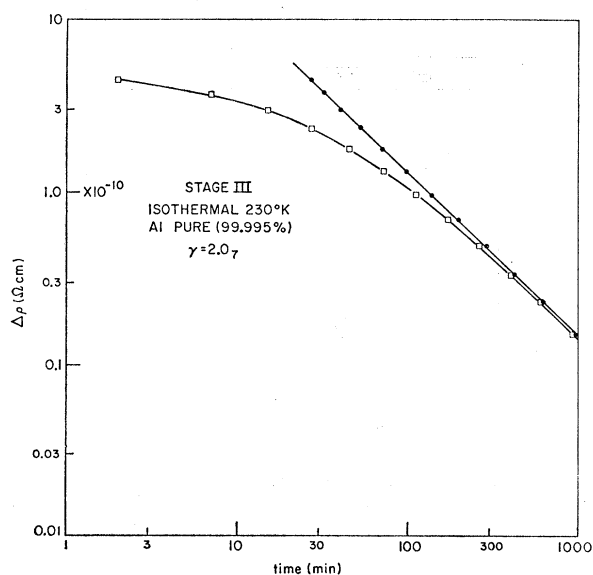


FIG. 5. Determination of the order of reaction (γ) for stage III with $M = 25$ min for the pure Al sample of Fig. 4.

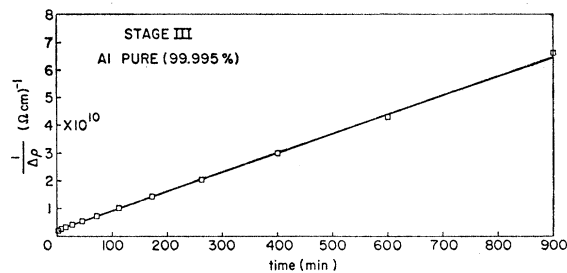


FIG. 6. A second method to determine the reaction coefficient (γ) for stage III for the pure Al sample of Fig. 4. The linearity of this plot indicates second-order kinetics.

range for which these energies are felt to be valid is about the same as that of the pure Al samples. It should be noted, moreover, that the effective frequency factor for these alloys is considerably higher than for the pure material, being of the order of 10^{18} sec^{-1} . Further, in the case of Al-Mg, the effective frequency factor obtained from the analysis of stage III is larger by almost a factor of 10^4 than that obtained for stage II (Paper II) where the evidence for a detrapping process is very strong.

Calculations using Eq. (II5) for the stages centered at 210°K (Al-Ga), 212°K (Al-Ag), 235°K (Al-Mg), and 240°K (pure Al) give the following results. For the pure Al sample, the theoretical and experimental recovery curves are in fair agreement, indicating that this stage is

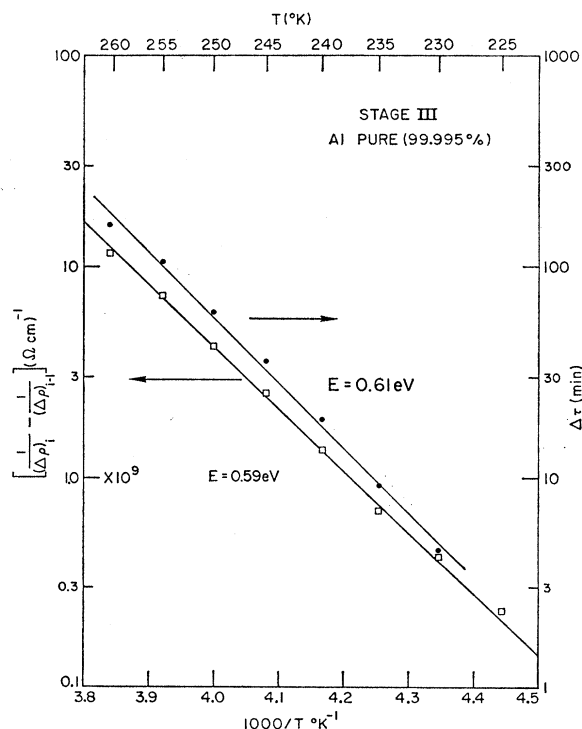


FIG. 7. Determination of the activation energy for the pure Al sample of Fig. 4 in the stage-III region by the $1/\Delta\rho$ method (square symbol) and by the Meehan-Brinkman method (dots).

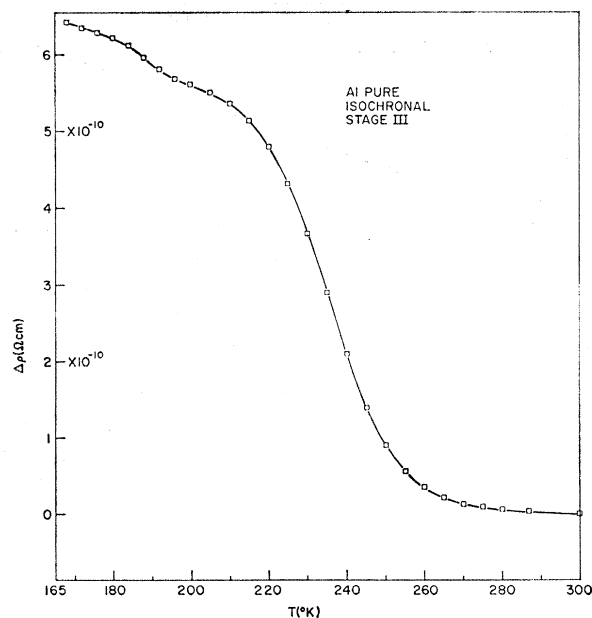


Fig. 8. Isochronal plot of stage III for the pure Al sample of Fig. 4.

not of a particularly complex nature. For the alloys, the highest peak in the experimental curve and the theoretical curves show a fair fit, but tails on either side of these peaks make the stages appear too broad to be single stages from 170 to 280°K. This, coupled with the observation that the peak at 190°K in pure Al is influenced by impurities and the considerations concerning the energies and effective frequency factors mentioned above (see Table I), leads to the conclusion that in this stage more than one process may be taking place.

The recovery of the Al-Ag alloy sample is of particular interest. Contrary to previous anneals where a resistivity increase is apparent (see Fig. 1 of Paper II), this alloy exhibited rather normal recovery behavior (Fig. 10), except that a large amount of damage remains at 300°K. This change in the recovery pattern may be explained by assuming that clustering of Ag atoms occurs during annealing but is significantly reduced after several irradiations and annealing treatments. That is, after several anneals the concentration of Ag atoms in solution is nearer an equilibrium (or pseudo-equilibrium) concentration and clustering becomes negligible. We believe, however, that the concentration of Ag atoms in solution was not altered drastically, since the amount of suppression in stage I did not change detectably.

IV. FURTHER DISCUSSION

There are several models proposed to explain the recovery of point defects after irradiation. Concentrating mainly on the stage-III recovery, these may be classified into four categories. (1) The vacancy model:

In this model, vacancies (or vacancy aggregates) are assumed to be the mobile defects in stage III. (2) One-interstitial model: In this model, interstitials are assumed to be the predominantly mobile species in stage III. However, the interstitial is taken to have been mobile in stage I and trapped prior to stage III by one or more possible processes: formation of di-interstitials or higher interstitial aggregates, trapping by impurities, trapping by dislocations, etc. (3) The conversion two-interstitial model¹²: In this model, one

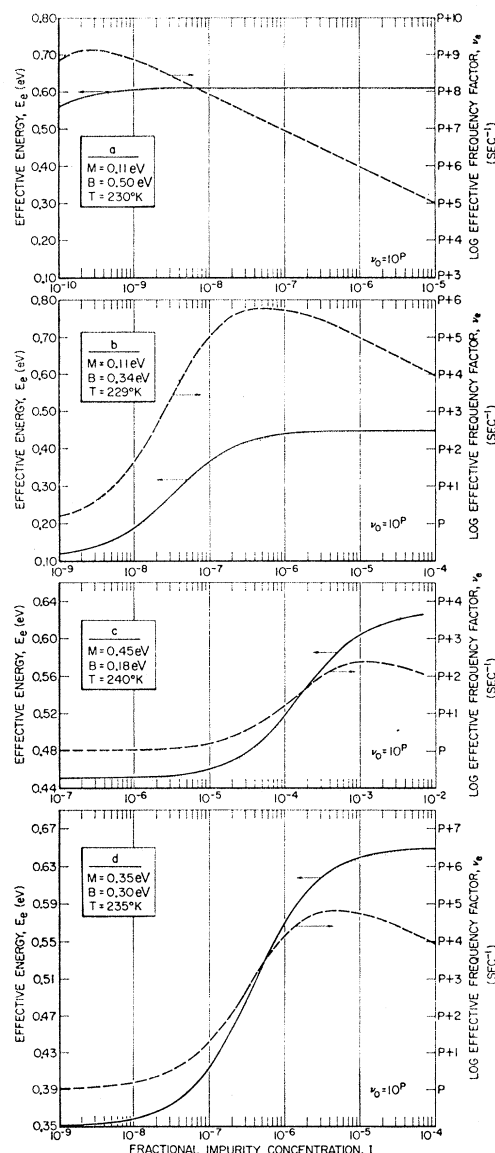


Fig. 9. Effective activation energy E_e and effective frequency factor ν_e calculated for different impurity concentrations I . The temperatures and values of migration energy M and binding energy B were chosen to yield results which might correspond to stage-III observations.

¹² W. Bauer, A. Seeger, and A. Sosin, Phys. Letters 24A, 195 (1967).

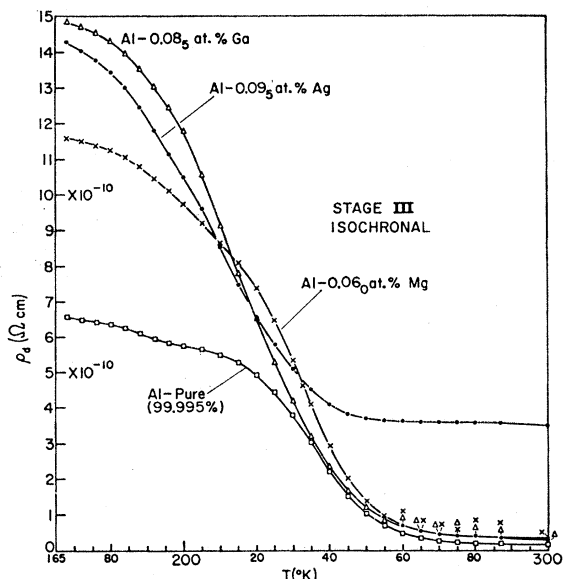


FIG. 10. Isochronal plot of stage III for Al and the Al alloys investigated. All samples were irradiated and annealed simultaneously.

form of interstitial is particularly mobile, migrating in stage I. The interstitial which migrates in stage III is a geometrically distinct specie but was formed during stage-I annealing. (4) The von Jan model¹³: In this model, free interstitial migration occurs initially in stage III. Previous recovery is due to annihilation of bound interstitial-vacancy pairs.

Federighi and co-workers have been the main proponents for vacancy migration in Al in stage III. Their evidence for this interpretation is based on quenching,¹⁴ plastic deformation,¹⁵ and neutron-irradiation experiments,^{16,17} and consists primarily of three observations. First, the activation energy they measure, about 0.6 eV, is close to the migration energy for vacancies found by De Sorbo and Turnbull.¹⁸ More direct evidence offered by Panseri and Federighi¹⁹ concerns the tendency toward segregation in various aluminum alloys following quenching, where interstitials are not expected to play a role. Also, it is well known²⁰ that mechanical recovery may occur at temperatures in the stage-III region, or even below this region, in high-purity aluminum.

An argument against assigning vacancy migration to stage III based on activation energies can be formulated

in the following manner. If in the pure Al sample all of the resistivity increment ρ_d remaining in stage III is assumed to be due to vacancies, then a hypothetical quenching temperature, to yield an equivalent value of $(\Delta\rho)_0$, may be calculated by

$$(\Delta\rho)_0 = A \exp[-E_f/kT_Q] = \rho_d, \quad (1)$$

where $(\Delta\rho)_0$ is the quenched-in resistivity, A is a constant, E_f is the formation energy for a vacancy, and T_Q is the quenching temperature. Using the values obtained by De Sorbo and Turnbull,¹⁸ $A = 3 \times 10^3 \mu\Omega \text{ cm}$, $E_f = 0.79 \text{ eV}$, and, from Fig. 1, the value of $(\Delta\rho)_0 = \rho_d = 5.5 \times 10^{-4} \mu\Omega \text{ cm}$ at 210°K , the calculated value of $T_Q = 590^\circ\text{K}$ is obtained. From the isochronal plots of Panseri and Federighi,¹⁴ their Fig. 2, or Doyama and Koehler,²¹ their Fig. 1, it is estimated that for a value of $T_Q \approx 590^\circ\text{K}$, vacancy migration does not become appreciable until approximately 320°K or higher. In order for appreciable vacancy migration to be observed in the stage-III region, T_Q would have to be about $825\text{--}875^\circ\text{K}$. This quenching temperature would give a defect concentration approximately fifteen times that observed. Note that by assuming the maximum value possible for $(\Delta\rho)_0$, the highest value possible for T_Q is calculated, and this results in the lowest possible value for the temperature of migration of vacancies. Nevertheless, this temperature for the migration of vacancies is near the end of the observed recovery in stage III. Thus the observed stage III following irradiation in Al is too low in temperature to be due solely to vacancies. Similar conclusions have been reached by Budin *et al.*²² for the case of stage III in copper.

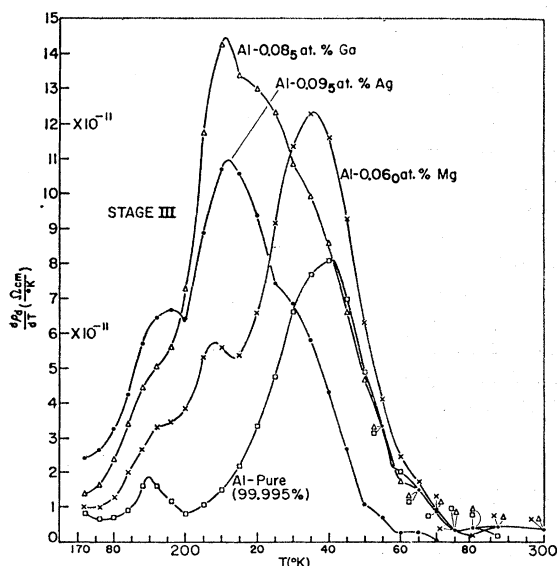


FIG. 11. Derivative plot of the isochronal shown in Fig. 10.

¹³ R. von Jan, *Phys. Status Solidi* **17**, 361 (1966).
¹⁴ C. Panseri and T. Federighi, *Phil. Mag.* **3**, 1223 (1958).
¹⁵ C. Panseri, S. Ceresara, and T. Federighi, *Nuovo Cimento* **29**, 1223 (1963).
¹⁶ S. Ceresara, T. Federighi, D. Gelli, and F. Pieragostini, *Nuovo Cimento* **29**, 1244 (1963).
¹⁷ T. Federighi, S. Ceresara, and F. Pieragostini, *Phil. Mag.* **12**, 1093 (1965).
¹⁸ W. De Sorbo and D. Turnbull, *Phys. Rev.* **115**, 560 (1959).
¹⁹ C. Panseri and T. Federighi, *Acta Met.* **8**, 217 (1960).
²⁰ C. Frois and O. Dimitrov, *Mem. Sci. Rev. Met.* **61**, 753 (1964).

²¹ M. Doyama and J. S. Koehler, *Phys. Rev.* **134**, A522 (1964).
²² C. Budin, P. Lucasson, and A. Lucasson, *J. Phys. (Paris)* **26**, 9 (1965).

In carrying through the analysis in the preceding paragraph, we have ignored any considerations of sink distribution or density. Since stage III is sensibly second order in nature, we may conclude that the distribution of vacancies and interstitials is effectively random throughout this stage. Thus we may consider merely sink density. In quenched samples, dislocations are the main sinks for vacancies; in irradiated samples, interstitials are presumably the most important sinks if we postulate vacancy migration in stage III. Taking reasonable estimates of sink concentrations for these two cases, the estimated annealing temperature would be lowered from 320°K by $\lesssim 50^\circ\text{K}$, in order to make the comparison. This new estimate, $\sim 270^\circ\text{K}$, is still near the end of stage III. Note that it is at the center of the increase observed in Al-Ag (see Fig. I-1 in paper II).

While the energy of 0.46 eV reported here is too small to be attributed to vacancies, it is, however, close to the migration energy attributed to divacancies (0.50 eV) by Doyama and Koehler.²¹ Divacancy migration in stage III after 1-MeV electron irradiation may be discounted because the concentration of divacancies available for stage III would be too small to account for the amount of recovery observed. Although a small number of divacancies may be formed during the irradiation, the annealing in stages I and II would reduce this to an insignificant amount by stage III.

These arguments do not preclude any vacancy migration in stage III. The derivative plot of this stage, Fig. 2, shows a change in the shape of the curves occurring in the latter region, around 250 to 265°K in particular. Thus, vacancies appear to move in this latter region of the recovery.

Recent Young's-modulus measurements by Keefer²³ on Al irradiated with 1-MeV electrons to a dose of 1.8×10^{17} electrons/cm² give further indication that vacancies migrate in the temperature range slightly above stage III. Specifically, Keefer observes a dislocation pinning stage which starts at about 260°K and is centered at 360°K. That the observed peak is due to vacancy migration is verified by further results of Keefer which show a pinning after quenching in the same range as the stage following irradiation. The dose used by Keefer was a factor of ten lower than in the present experiments. Taking this into account would lower slightly the observed temperature of migration, which would place the start of vacancy migration near the end of stage III, as in the present experiments.

It is evident that the discussion concerning ascribing vacancy migration to stage III revolves heavily about the measured activation energies and a discrepancy is generally present. The discrepancy in Al is, we believe, aggravated by the effects of impurities, as evidenced in this work by the dependence of activation energy of residual resistivity, and by the proximity of stages III

and IV. In this unfortunate situation, added importance is given to evidence which may be less quantitative but more direct in its implications. The evidence of Frois and Dimitrov²⁰ concerning mechanical recovery could be cited as such an example. In our opinion, the observation that mechanical recovery may occur even well below stage III in sufficiently pure Al or well above stage III in less pure Al makes such observations suspect as evidence for vacancy migration.

It is appropriate, then, to turn to models in which interstitial migration accounts for stage III. Experimental evidence may be cited which poses apparent difficulties for any interstitial model. Some of these have been reviewed recently by Corbett.²⁴ Accordingly, we will not attempt to discuss any model in detail but, rather, limit discussion mainly to features related to the present work.

It has been proposed that stage III consists of either migration of di-interstitials, formed in an earlier stage, to vacancies; of breakup of such di-interstitials and subsequent rapid migration of single interstitials; or of release of interstitials from traps formed by impurities or dislocations. Di-interstitial models suffer from the observation that stage III is present in samples irradiated at temperatures above stage I, where, presumably, interstitial migration occurs over long distances during irradiation. Under these conditions, it is difficult to understand how di-interstitials may be formed by the direct encounter of two migrating interstitials since the instantaneous concentration of single interstitials is always essentially zero. Furthermore, it has been shown²⁵ that the annealing above stage I in copper is essentially the same in samples irradiated above or below stage I—a result which is inconsistent with a di-interstitial model. Release of interstitials from dislocations appears unlikely based upon the observation that dynamic elastic modulus measurements²⁶ indicate a net arrival of point defects in stage III in Cu, not a release.

Finally, release of interstitials from impurities is ruled out as a primary mechanism for stage III, in Al, Cu, Ag, and Au, by the observations of Burger *et al.*²⁷ which show that stage III is essentially the same in samples in which the defect concentration was varied over one-hundredfold; in the high-dose cases, the defect concentration was well in excess of the impurity concentrations.

The next model we consider is one proposed recently by von Jan.¹³ As stated previously, in this model long-range migration occurs only in stage III. The recovery, in pure aluminum, in stages I and II is restricted to close

²⁴ J. W. Corbett, *Electron Radiation Damage in Semimetals and Metals* (Academic Press Inc., New York, 1966).

²⁵ W. Bauer and A. Sosin, *Phys. Letters* **24A**, 193 (1967).

²⁶ D. W. Keefer, J. C. Robinson, and A. Sosin, *Acta Met.* **13**, 1135 (1965).

²⁷ G. Burger, H. Meissner, and W. Schilling, *Phys. Status Solidi* **4**, 267 (1964).

²³ D. W. Keefer (private communication).

pair annihilation. An important factor in von Jan's model is the assumption that two forms of interstitials are created by irradiation but that conversion of one type to the other occurs near the end of stage I. In the context of recovery in aluminum alloys, it is difficult to see how this model can account for the existence of the large recovery substages generally introduced in stage II. It is even more difficult to account for the complex kinetics of these substages. Since long-range interstitial migration is presumed to occur at more elevated temperatures, any impurity-induced substructure in stage II must result from local motion in the general region of a vacancy, an interstitial, and an impurity atom. The resulting kinetics should be accurately first order, contrary to the observations reported in the previous paper.⁹

Finally, we consider the "conversion-two-interstitial model."¹² This model differs from its predecessor,^{3,6} the "two-interstitial model," in its further inclusion of conversion of crowdions to dumbbell interstitials in stage I by other defects, including impurities. With respect to ability to account for the observations in this paper and the preceding one, both models appear adequate. The complex kinetics in the stage-II substages can be ascribed to interstitial-vacancy recombination and interstitial trapping by impurities following release of interstitials (crowdions) from shallow impurity traps. The activation energy in stage III and its dependence on residual resistivity, the general shape of stage III, and the tendency toward second order reaction all are consistent with interstitial (dumbbell) vacancy recombination, disturbed by the lingering effects of impurities. Any criticism of the conversion-two-interstitial model would evidently arise from considerations not related to this paper or the preceding one. Such criticisms have been discussed in the introduction of this model.¹²

V. SUMMARY AND CONCLUSIONS

The addition of impurity atoms in pure aluminum introduces substantial modification in the recovery

pattern of stage III following electron irradiation. Pure aluminum displays two recovery stages. The first is small in size and centered at 190°K and is influenced by the residual impurities. The second and main stage is centered between 235 and 245°K depending on the purity of the sample and the integrated electron flux. The observed activation energy for the recovery varies with the purity of the material for nominally pure aluminum. The higher the purity of the material, the lower the activation energy. A variation from 0.46 to 0.59 eV was observed for the materials used in this investigation. The effective frequency factor also varied with the purity, being lower for the higher-purity material. These variations on the energy and frequency factors may be explained on the basis of an impurity-trapping model, as discussed in the previous paper.

The recovery in alloyed samples was found to be more complex in the stage III region, as evidenced by the greater substructure in the alloys compared to the pure material. The activation energy governing the bulk of stage III is only slightly higher than in some of the nominally pure aluminum samples, being about 0.62 eV. All alloy samples, with the exception of Al-Ag, showed essentially complete recovery by 300°K.

The Al-Ag sample showed considerably different recovery in the stage-III region. Starting at about 170°K, there was a decrease in the resistivity which continued to about 235°K, after which there was an increase which lasted until about 290°K, followed by another decrease. This pattern persisted for several successive investigations of the recovery and then apparently ceased. This behavior can be explained on the basis of a clustering of Ag atoms.

From the analysis of the data, it is concluded that vacancies are not the main defect migrating in stage III in Al, particularly the earlier portion of the stage. The evidence in this article and the two previous ones indicates long-range interstitial migration in stages I, II, and III, with impurity interaction playing a significant role in each stage.