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FURTHER STUDIES ON THE EFFECTS OF IONIZING RADIATION, HEATING CYCLES AND QUENCHING ON THE TEMPERATURE DEPENDENCE OF THE ELECTRONIC CONDUCTIVITY OF DIFFERENT DILUTE Al–Si AND Al–Fe ALLOYS

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ABSTRACT

Wires of pure Al, dilute Al–Si, and more dilute Al–Fe alloys were carefully prepared. Comprehensive measurements of the direct voltage electrical conductivity were made on several specimens of these materials. Furthermore, conductivity measurements were followed as a function of temperature, heat recycling, quenching, Si or Fe content, and after a chosen absorbed gamma dose. Values of activation energies for conduction, energy gap and the activation energy for the process of carrier liberation were established and discussed in detail. In the higher temperature region, the electrical resistivity and activation energy for conduction were found to decrease with heat recycling. This was explained by the annealing of the alloy and dissolution of the precipitated phases.

The results obtained were all in agreement since the values of σ, ΔE and Φ were all considerably increased by quenching, Si or Fe content and γ-irradiation. These were highly ascribed to, and proved to be due to the formation of precipitated phases and lattice imperfections caused by γ-radiation damage. This may lead to the propagation of defective crystal lattices.

Finally, conductivity measurements are a useful and sensitive (non-destructive) tool for controlling either Si or Fe content in the desired Al–Si and Al–Fe alloys to the required quality.

Gamma radiolysis at a selected absorbed dose could also be used to replace either Si or Fe dopants for controlling the electrical properties of pure Al metal in the Al industries.

INTRODUCTION

It is of interest to note that the nucleation [1] of precipitates in Al–Si alloys was enhanced by pre-ageing quenched specimens near room tempera-

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ture. Such an enhancement can be attributed to dislocation loops formed by the condensation of quenched-in vacancies. On the other hand, it was found that [2] Si precipitates became visible after the dislocation loops had disappeared and that the number of loops was smaller than the number of precipitates, which indicated that dislocation loops did not act as preferred nucleation sites for Si precipitates. Ozama and Kimura [3,4] showed that the Si precipitates nucleated on vacancy clusters, but their formation, and the subsequent nucleation, took place within a few seconds after quenching.

Electrical resistivity provides a means for continuous measurements to be made during heat treatment or deformation, and is successfully used in the detection, formation and migration of irradiation defects, and cold work defects [5–11].

The present work was undertaken in an attempt to follow the behaviour of the temperature dependence of direct current electrical conductivity in pure Al (supplied by Al-Co., Egypt), and dilute Al–Si alloys before and after both quenching and an absorbed gamma dose (5×10^7 rad). The results obtained were analysed and interpreted in terms of quenched-in defects and γ-radiation damage.

EXPERIMENTAL

Pure Al metal was used. Dilute Al–Si and Al–Fe alloys were made from pure, high purity Al, Si and Fe from BDH (> 99.9%). Wires of pure Al, Al–Si (0.01–0.37% Si), and Al–Fe (0.0005–0.0030% Fe), 82 mm long and 0.4 mm diameter, were used.

The DC electrical conductivity circuit used enabled continuous measurements to be made during various heat treatments using a 16 OB Keithley digital multimeter with an accuracy of 10^{-8} Ω cm. A vertical furnace (10 cm diameter, 20 cm long) was used for heating the specimens. Samples were quenched from 220 °C in an iced water tank.

For gamma irradiation, the specimens were exposed in air to a chosen absorbed energetic gamma dose of 5×10^7 rad at room temperature using a ^{60}Co γ-source (300 Cis).

Measurements were conducted before and after irradiation, as well as after quenching and heat recycling.

RESULTS AND DISCUSSION

Effects of quenching

Alloys based on the Al–Si system are called "silumin alloys", which are distinguished by their good castability (high fluidity and low shrinkage), good corrosion resistance, high ductility and low specific gravity (2.57 g cm^3).

Iron is a harmful impurity in Al–Fe alloys, since it considerably reduces the strength and ductility of the alloy. Therefore, iron content should never exceed 0.0030%.

The room temperature electrical conductivity of annealed pure Al is $2.6 \times 10^4\ \Omega^{-1}\ cm^{-1}$ (see Fig. 1) which is in agreement with that previously given ($34 \times 10^4\ \Omega^{-1}\ cm^{-1}$) by Lakhtin [12].

The results depicted by selected representative graphs (Figs. 1–7) indicate that the electrical conductivities of both pure Al and Al alloys decrease linearly with temperature. This behaviour is mainly attributed to lattice vibrations and the increased free electron density, along with their decreased carrier mobility upon cooling (Figs. 1–7). The conductivity decreases for pure Al and Al alloys, which is correlated with further annealing of the specimens and the removal of internal strains. The electrical conductivity (room temperature value, 20 °C) of pure Al was found to decrease with its alloying either with silicon or iron, and as a function of their content. This is most probably related to the alloying influence. This influence appears to be ageing, causing precipitation, which is in agreement with observations of previous investigators [11,13,14].

Quenching of pure Al (Fig. 1) from 70–220 °C in iced water has no significant effect on the final electrical conductivity values obtained, indicat-

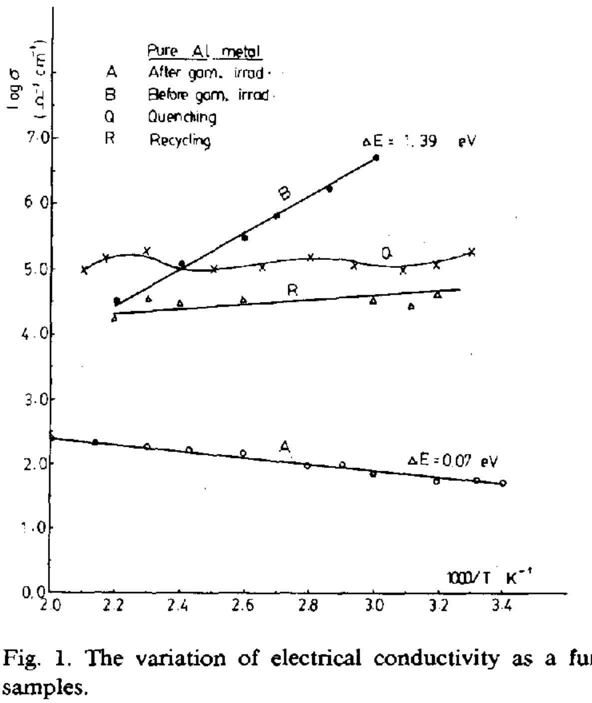
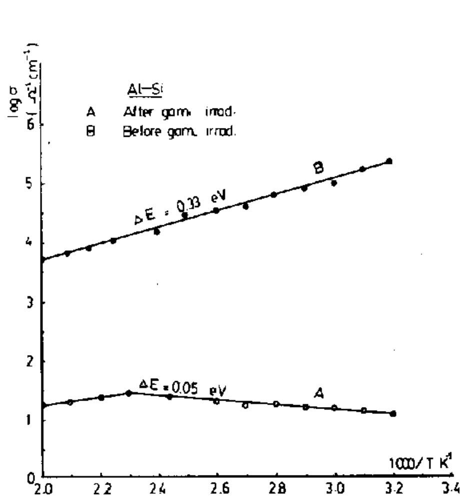


Fig. 1. The variation of electrical conductivity as a function of temperature for pure Al samples.



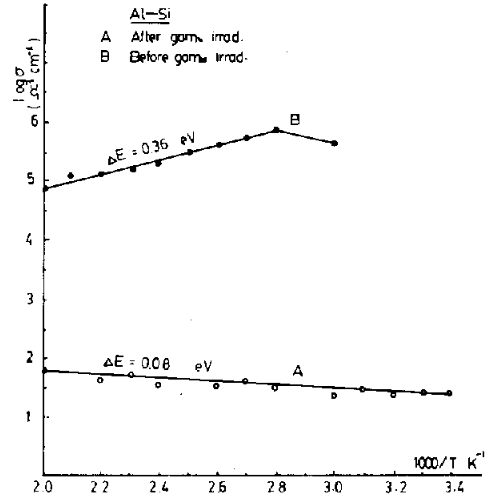
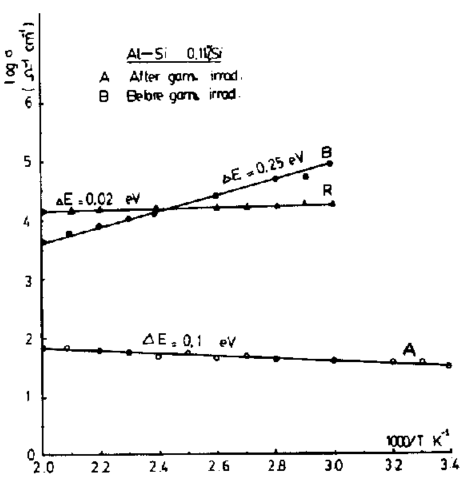


Fig. 2. The variation of electrical conductivity as a function of temperature for Al–Si (0.01% Si) samples.

Fig. 3. The variation of electrical conductivity as a function of temperature for Al–Si (0.02% Si) samples.



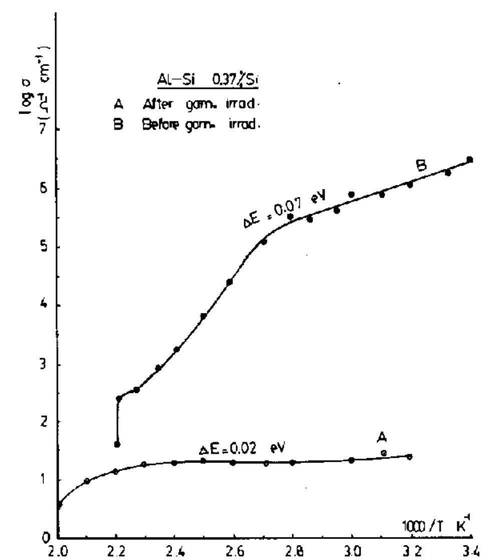
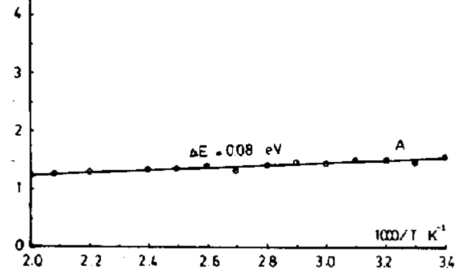


Fig. 4. The variation of electrical conductivity as a function of temperature for Al–Si (0.11% Si) samples.

Fig. 5. The variation of electrical conductivity as a function of temperature for Al–Si (0.37% Si) samples.

ing that the quenched-in defects cannot exist therein. On the other hand, quenched-in defects are considerable in Al–Fe alloys (see Fig. 7) under the same quenching conditions as for pure Al, causing a considerable decrease in the electrical conductivity obtained after quenching. This was found to be consistent with the results of Flynn et al. [15], Feller-Kniepmeier and Gobrecht [16] and Madden and Cottrell [17]. Alternatively, quenched-in defects might be in the form of free vacancies, vacancy agglomerates or stacking fault tetrahedra [16], depending upon quenching temperature, size of wires, speed of quenching and quenching medium. Quenching tetrahedra were reported [16,18] to form in gold quenched from 900 °C in iced water. Chik [19] indicated that the building rate of these tetrahedra is highest at room temperature and practically zero at 100 °C. The results obtained from Figs. 8 and 9 show that saturated and supersaturated states are attained with prolonged and relatively high temperatures for Al–Si (0.1% Si) and Al–Fe (0.0015% Fe). These effects are attributed to the interaction of quenched-in vacancies with screw or mixed dislocations [20].

Furthermore, in accordance with previous investigators [21,22], GP zones are formed during the quenching of the present alloys which are controlled by the enhanced diffusion of Si or Fe atoms due to quenched-in vacancies. The formation of these zones can cause an increase in the final obtained electrical resistivity values.



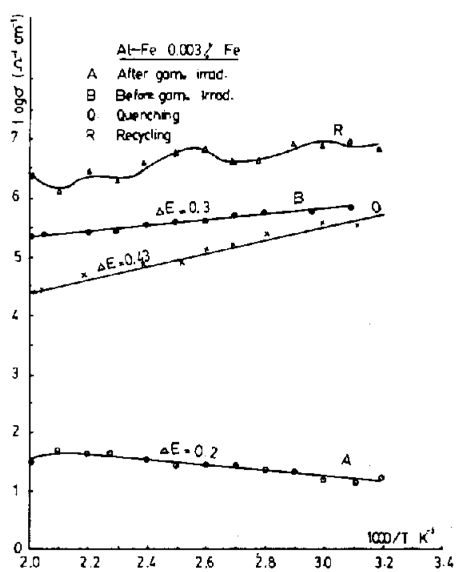


Fig. 6. The variation of electrical conductivity as a function of temperature for Al–Fe (0.0015% Fe) samples.

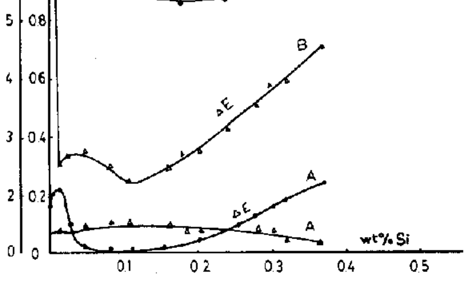
Fig. 7. The variation of electrical conductivity as a function of temperature for Al–Fe (0.0030% Fe) samples.

Effect of heat recycling

Figures 1, 4 and 7 show the effect of heat recycling on the temperature dependence of electrical conductivity. It can be seen that heat recycling increases the final electrical conductivity obtained and progresses with the increasing number of heat cycles. This could probably be correlated with the role of heating in the dissolution of the present precipitated phases and anihilation of the defects formed and/or lattice strains in both Al–Si and Al–Fe alloys. This effect is more pronounced in Al–Fe alloys due to the harmful effect of Fe content in comparison with Si in Al–Si alloys.

Effect of γ-irradiation

Figures 1–9 and Table 1 illustrate the various changes in the electrical parameters of both Al–Si and Al–Fe alloys which are collaborated or induced after an absorbed gamma dose (5×10^7 rad). It can be seen throughout that the electrical conductivities decrease whereas activation energy for conduction, gap width and the activation energy for the process of carrier liberation are all reversed from metallic to semiconducting activation energy for both Al–Si and Al–Fe alloys. These changes were more pronounced in Al–Si alloys (Figs. 1–4) than for Al–Fe alloys (Figs. 6 and 7)



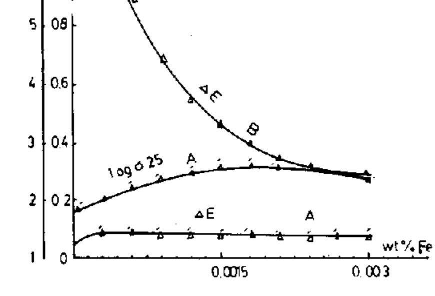


Fig. 8. The variation of room temperature electrical conductivity, log σ_{25}, and activation energy for conduction, ΔE (eV), as a function of Si content in dilute Al–Si alloys. (B) Before irradiation, (A) after irradiation.

Fig. 9. The variation of room temperature electrical conductivity, log σ_{25} (by extrapolation), and activation energy for conduction, ΔE (eV), as a function of Fe content in very dilute Al–Fe alloys. (B) Before irradiation, (A) after irradiation.

which could be due to the greater interaction of Al with Si content and the harmful effect of the Fe impurity due to the greater diffusion of iron (1.26 Å) to that of Si (1.32 Å).

From Fig. 8, it can be easily seen that: (a) both silicon doping and γ-radiolysis decrease the electrical conductivity of Al–Si alloys with the tendency to rise again at higher Si contents. The latter is ascribed to the formation of a more conductive precipitated phase; (b) the activation energy for conduction continuously decreases after irradiation even for high Si contents (0.37%).

From Fig. 9 it can be noted that: (a) Fe content has a greater influence on the activation energy values than γ-radiation; (b) γ-radiation damage becomes constant at an early stage with respect to % Fe due to the harmful effect of iron. Accordingly, with the exception of a higher Si content (0.37%) in Al–Si alloys and a lower Fe content (0.0015%) in Al–Fe alloys, the dose of γ-radiation absorbed induced semiconductivity in both Al alloys investigated.

The activation energy for carrier liberation [23] could be calculated for the materials investigated using the relation

$$\Phi = \Phi_2 - \Phi_1$$

where Φ_2 is the activation energy for conduction after the absorbed gamma dose (5×10^7 rad), and Φ_1 is the activation energy for conduction before irradiation (see Table 1).

From Table 1 and Figs. 8 and 9, it is clear that although the Fe content in Al–Fe alloys is very small, it consistently decreases the activation energy for conduction and, hence, the activation energy for the process of carrier

TABLE 1

Values of the activation energy obtained before (Φ_1) and after (Φ_2) the absorbed gamma dose, and the activation energy for the process of carrier liberation (Φ) for dilute Al–Si and very dilute Al–Fe alloys

Al–Si alloys				Al–Fe alloys			
Wt.% Si	Φ_1 (eV)	Φ_2 (eV)	Φ (eV)	Wt.% Fe	Φ_1 (eV)	Φ_2 (eV)	Φ (eV)
0.00	1.38	0.05	1.33	0.000	1.38	0.05	1.33
0.01	0.30	0.08	0.22	0.0003	1.10	0.10	1.0
0.05	0.35	0.10	0.25	0.0006	0.90	0.09	0.81
0.10	0.25	0.11	0.14	0.0009	0.70	0.08	0.62
0.15	0.28	0.090	0.19	0.0012	0.55	0.07	0.48
0.20	0.35	0.08	0.27	0.0015	0.47	0.08	0.39
0.25	0.47	0.07	0.40	0.0018	0.40	0.09	0.31
0.30	0.57	0.06	0.51	0.0021	0.35	0.06	0.29
0.35	0.68	0.04	0.64	0.0024	0.32	0.07	0.25
				0.0027	0.29	0.04	0.22
				0.0030	0.30	0.06	0.24

liberation, unlike Si in Al–Si alloys. These observations could well be explained by the greater mobility of Fe than for Si in the investigated alloys.

The relatively low activation energy for carrier liberation of the Al–Fe alloy (see Fig. 9) compared to that for the Al–Si alloy (see Fig. 8) could be regarded as evidence for the extent of radiation damage. Thus, Al–Fe alloys have a greater resistance to γ-radiation damage than Al–Si alloys. This could be correlated with the greater mobility of Fe than Si in Al–Fe and Al–Si alloys, respectively, and the large lattice defects initially present in Al–Si alloys.

In summary, Figs. 1–9 illustrate the effects of γ-radiolysis on pure Al, dilute Al–Si alloys and very dilute Al–Fe alloys. Gamma radiolysis is thought to have two different effects [24]: (a) pining of dislocation, thus producing a yield point effect; (b) producing in the volume of the crystal new obstacles to the motion of dislocations. Pining might be due to isolated point defects which migrate to the dislocation lines or are possibly directly created there by the action of radiation [25]. These isolated point defects create dislocation loops. These impede the motion of mobile dislocations, giving rise to a high electrical resistivity, which is fairly temperature independent. According to Seeger [26], the present radiation-induced damage might be due to small clusters of point defects which form in irradiated materials as observed black dots [27]. These results are attributed to the formation of precipitation nuclei, due to the increased diffusion caused by the presence of newly formed vacancies and interstitial atoms.

Accordingly, since irradiation increased the density of lattice defects, interactions with the impurity atoms reached higher levels, leading to the further observed increase in the measured parameters. The electrical resistivity increase after irradiation agrees well with the literature [23,28].

Finally, all results obtained and their stimulated discussions were in conformity, leading to the establishment of a nearly full account of various factors affecting and controlling the behaviour of the electrical properties of pure Al, dilute Al–Si and very dilute Al–Fe alloys for aluminium in electricity and industry.

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