

AGGLOMERATION OF VACANCIES IN PURE FAST-QUENCHED ALUMINIUM<sup>\*)</sup>E. GIRT, B. LEONTIĆ<sup>\*\*)</sup> and A. KURŠUMOVIĆ*Metal Physics Laboratory, Department of Natural Sciences, University of Sarajevo, Sarajevo*

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*Abstract:* In order to observe the migration of vacancies in pure aluminium the metal was fast-quenched (»splat«-cooled) and immediately upon forming stowed in liquid nitrogen. Samples thus conserved were subjected to resistometric observation from 77 K to room temperature. They were then cooled once more to 77 K and the measurement of electrical resistance repeated.

The results indicate that in the interval of 77 K to  $170 \pm 5$  K the values of electrical resistance in the two sets of measurements differ, being larger in the first.

The effect is tentatively interpreted as the agglomeration of monovacancies into clusters since during the first set of measurements the electrons were scattered by a greater number of centers while during the second set of measurements these centers decreased in number. The results suggest that all monovacancies probably disappear already at about 170 K.

### 1. Introduction

According to a number of author's<sup>1-4)</sup> monovacancies do not exist in pure aluminium above 220 K. In order to verify this assertion we developed a new experimental technique which confirmed it but indicated that the temperature at which monovacancies disappear is lower.

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The basic idea of the experiment is that the samples must be «frozen» as they emerge from the process of fast quenching at which point they contain a large concentration of vacancies. Such «frozen» samples are then subjected to resistometric measurements while being slowly warmed to room temperature. Following this first set of measurements the samples are cooled to 77 K and analogous measurements repeated. The difference between the two sets of data will furnish

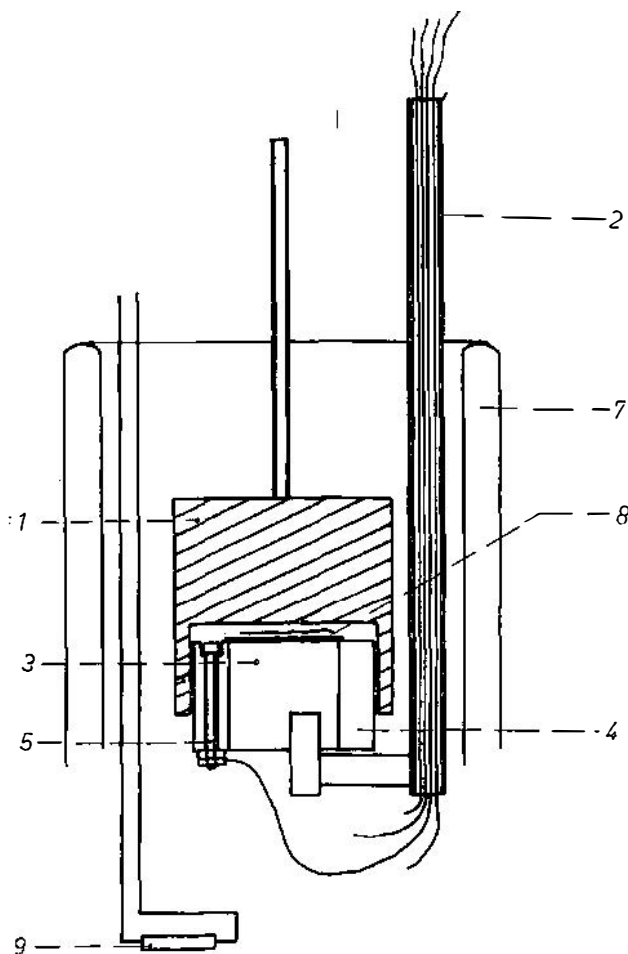


Fig. 1. Cross-sectional view of the apparatus used for measurements. The numbered components are described in the text.

information on the reduction in the number of scattering centers. Experimentally the problem is reduced to two main tasks. The first is to preserve the quenched sample in the state as formed by cooling it as fast as possible to 77 K. The second is to solve the problem of microwelding the electrical leads to the cold sample without heating it. Thus the process of microwelding has to be carried out in liquid

nitrogen. The sample can be brought very quickly to the temperature of liquid nitrogen by the use of the »rotating mill« quenching device<sup>5</sup>). As soon as it is formed and ejected from between the rollers it enters a vessel containing liquid nitrogen. By suitably shaping the vessel and bringing the liquid nitrogen close to the rollers the sample can be brought to 77 K in less than 1 ms upon being formed. It thus retains most of the properties it had at the instant it was quenched. Two contacts are then welded on the sample while submerged in the liquid nitrogen using a thyristor device<sup>6, 7</sup>). In practice no difficulty was experienced due to the presence of liquid nitrogen.

In order to facilitate the manipulation of the samples and the measurements of their resistivity an appropriate apparatus was constructed.

## 2. Description of the apparatus

The apparatus (Fig 1.) consists of a sample holder with a massive lid 1. The holder itself consists of a stainless steel tube on which a copper platform 3 is mounted. A layer of teflon 4 surrounds the platform. Four bolts 5 traverse the teflon layer. These bolts are connected at the bottom to four leads while at the top they are connected to aluminium foil (6, Fig. 2). In addition a thermocouple also traverses the teflon layer (Fig. 2).

When the samples are made they are transferred together with the liquid nitrogen. At the bottom of this dewar a mesh holder is placed so that the samples can be raised close to the surface. A selected sample can then be grabbed with a special precooled tool and the other samples lowered back into the dewar. The special sample holder is now lowered into the liquid nitrogen in the dewar and precooled.

The selected sample is now placed on the holder so that its two ends slip under the aluminium foils (6, Fig 2). The welding proceeds with a precooled electrode the copper platform serving as the other electrode. After the welding, a Mylar foil (Fig. 1) is slipped under the sample and the latter is covered with the lid 1, which is lined inside with Mylar to insulate it from the sample. All the steps just described are carried out under liquid nitrogen so as not to raise the temperature of the sample.

The measurements are started at 77 K and then the nitrogen is driven off from the dewar by means of a heater (9, Fig. 1). Owing to its large mass the holder together with the sample heats up very slowly. The resistance is measured every five degrees while the current through the sample is held to 500  $\mu$ A. The potential is measured with an accuracy of  $\pm 30$  nV. When the desired temperature is reached the dewar is refilled with liquid nitrogen and the operation just described is repeated.

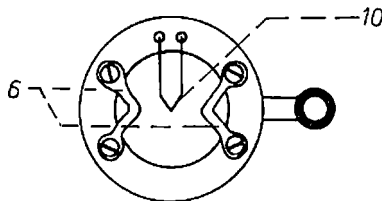


Fig. 2. A detailed view of the sample holder. The thermocouple (10) is situated at the sample center.

### 3. Results

The results of electrical resistance measurements in the temperature range from 77 K to 223 K are graphically shown in Fig. 3 and Fig. 4. Both graphs show the same change but Fig. 4 shows in more detail the interval within which the process of combination of vacancies into clusters is most pronounced. The values of resistance in the first series of measurements i. e. those started with the assumption that the sample contains »frozen« vacancies are shown on the graph with circles. Repeated measurements are shown with »X«-S.

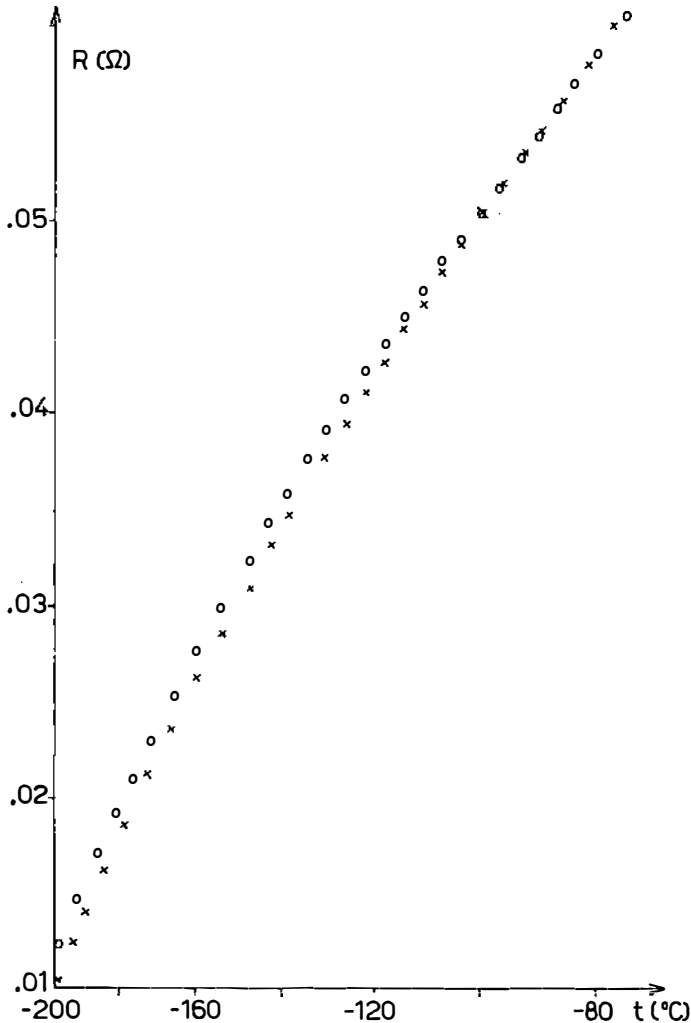


Fig. 3. The overall resistivity curve as obtained with a sample showing a fairly large amount of vacancy »freezing«.

The effect expected is evident since the repeated measurements show a 16% decrease in electric resistance with respect to the values first measured.

The results suggest that there is a strong migration of vacancies possibly monovacancies, and that it takes place intensively, because at around  $-120^{\circ}\text{C}$  this migration practically ceases. A large number of measurements on many samples were taken and the effect was observed in all samples, although its magnitude varied from case to case between 5% and 25%. We believe this variation is due to the fact that a large number of monovacancies migrate toward »condensation« centers during the short time the sample spends at room temperature (from the mo-

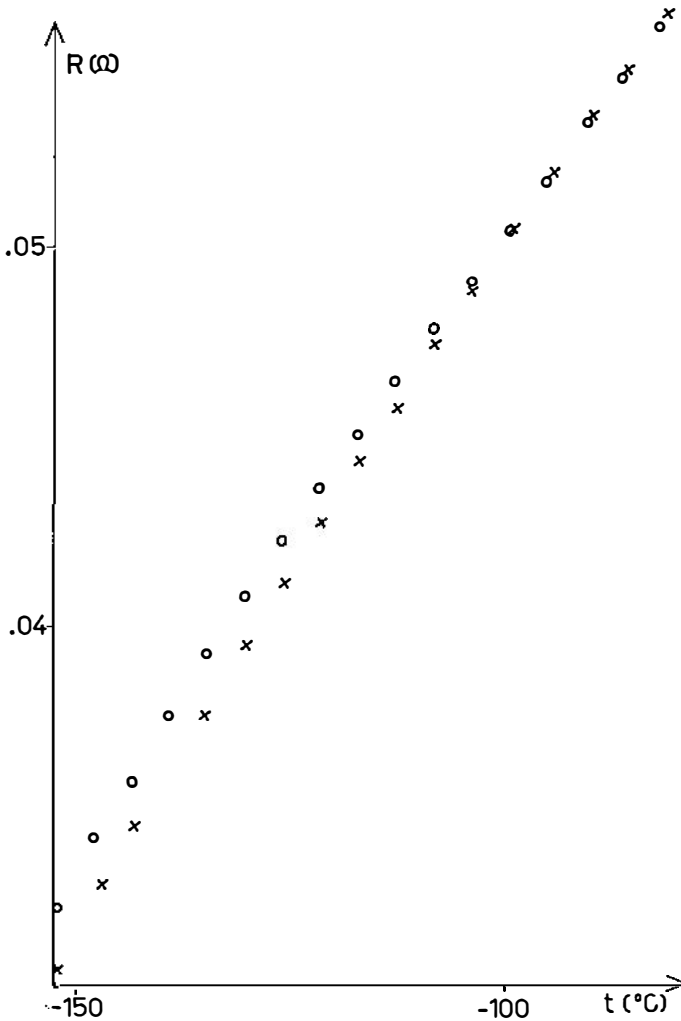


Fig. 4. A more detailed curve showing the region where massive vacancy agglomeration is believed to take place.

ment of formation until it reaches the liquid nitrogen). In a system quenched directly from the melt<sup>3)</sup> such centers are very numerous.

If we interpret these results to be due to rapid migration of monovacancies to the closest nucleation centers then these monovacancies disappear at  $-100 \pm \pm 5^\circ\text{C}$  since all our measurements converge to the same value at  $-100^\circ\text{C}$ . This result is at variance with that of several authors who quote this temperature to be  $-50^\circ\text{C}$ .

Another interpretation of the results described may be that the very large number of nucleation centers that are formed by the high temperature quenching serve as intermediate »stations« where some particularly high free energy clusters form before the sample is »frozen«. These clusters have very low activation energies and they readily diffuse to create larger centers below  $-100^\circ\text{C}$ . Those monovacancies that are »left over« then remain free to migrate at higher temperatures since their activation energies seem to be of the order of 0.7 eV.

It should nevertheless be stressed that we are dealing with very rapidly quenched systems which are known to be very disordered and differ in some properties from »conventionally« prepared samples.

#### Acknowledgements

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## NAKUPLJANJE VAKANCIJA U ČISTOM ULTRABRZO KALJENOM ALUMINIJU

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#### Sadržaj

Migracija vakancija u čistom brzo kaljenom aluminiju proučavana je tako da su uzorci »zamrznuti« u tekućem dušiku odmah po izlasku iz mlina za kaljenje.

Uzorci su podvrgnuti rezistometrijskom ispitivanju dok se griju na sobnu temperaturu da bi zatim ponovno bili vraćeni na 77 K i mjerenja ponovljena.

Rezultati pokazuju da u intervalu 77 K do  $170 \pm 5$  K električni otpori pokazuju odstupanja između prvog i drugog niza mjerenja time što je u prvom nizu mjerenja otpor veći.

Ovaj rezultat se interpretira kao efekt grupiranja monovakancija u grupice za vrijeme prvog grijanja pa se u drugom nizu mjerenja elektroni raspršuju na manjem broju nezavisnih središta. Vjerojatno je da iznad 170 K monovakancije u čistom kaljenom Al nestaju potpuno.