JOURNAL OF MATERIALS SCIENCE 7 (1972) 1351–1362

The effect of particle content, particle distribution and cold deformation on the recrystallization of low oxide Al-Al_2O_3 products

N. HANSEN
Metallurgy Department, Danish Atomic Energy Commission, Risö, Denmark

B. BAY
Materials Department, Danish Academy of Mechanical Engineering, Lyngby, Denmark

The recrystallization of dispersion-strengthened Al-Al_2O_3 products containing 0.6 and 1.2 wt % Al_2O_3 was followed by optical and transmission electron microscopy and by hardness measurements. The recrystallization was retarded compared to aluminium and the important structural parameters were the oxide content (proportional to the reciprocal particle spacing) and the distribution of oxide particles either as a uniform distribution or as a regular three-dimensional network. From the microstructures after cold work and after recovery it is suggested that particle-retarded recrystallization may be caused by pinning, during the recovery stage, of sub-boundaries and of individual dislocations. The hypothesis of retardation of recrystallization as due to particle-enhanced homogenization of dislocation structures during deformation is not supported by the microstructural observations. In the product containing 0.6 wt % Al_2O_3 the recrystallization was markedly retarded after 50% cold reduction, whereas the retardation was small after 80 and 90% reduction. An increase in the degree of cold deformation may reduce the critical size of the recrystallization nuclei, and thus the retarding effect of particles during nucleation may be reduced or disappear. It is therefore suggested that the degree of cold deformation and the particle spacing may be interdependent parameters when determining the recrystallization behaviour of dispersion-strengthened products.

1. Introduction

The microstructure of dispersion-strengthened products strongly influences the strength properties, and therefore the structural stability of desirable microstructures at high temperatures is of considerable interest. Empirically it has been found that recrystallization is retarded in products containing small, closely spaced particles, e.g. in Al-Al_2O_3 [1, 2] Cu-SiO_2 [3], and Ag-MgO [4], in all of which the particle size was below 0.1 μm and the particle spacing was 0.5 μm or less. In contrast to this behaviour, accelerated recrystallization has been observed in products containing coarse and widely spaced particles, e.g. in Al-$FeAl_3$ [5], where the particle size was about 1 μm and the particle spacing was 5 to 50 μm. Only products in the first category are dealt with in this paper.

In the products examined in this study the particles were either distributed uniformly in the metal matrix, or were arranged in a regular three-dimensional network [6]. It was the aim of the present work to study the recrystallization behaviour of these two types of structures. The alloy system chosen was Al-Al_2O_3. Two compositions were examined, namely 0.6 and 1.2 wt % Al_2O_3. These products were designated:
MD 201-uniform (uniform distribution of 0.6 wt % Al_2O_3 particles), MD 201-network (network distribution of 0.6 wt % Al_2O_3 particles), R400-uniform (uniform distribution of 1.2 wt % Al_2O_3 particles), R400-network (network distri-

bution of 1.2 wt % Al_2O_3 particles).

A supplementary parameter studied was the degree of cold deformation, since it has been found [7] that this parameter may affect the critical size of the recrystallization nuclei; the degree of cold deformation and the particle distribution may therefore be interdependent parameters in determining recrystallization characteristics. The recrystallization behaviour was studied by isochronal annealing followed by microscopy and hardness measurements.

2. Experimental Techniques

Al-Al_2O_3 products were manufactured by consolidation of atomized aluminium powder. The manufacturing procedures for products containing the oxide particles in a uniform distribution and in a three-dimensional network are given respectively in [8] and [6]. The purity of the aluminium phase in Al-Al_2O_3 was about 99.5% and commercial aluminium of about the same purity has been included for comparison. The chemical analysis and the structural parameters of the materials are given in Table I. There is a higher iron content in the Al than in the Al-Al_2O_3, but judging from recrystallization experiments on Al-Fe alloys [5] this difference should be unimportant. After manufacturing (extruding), the materials were recrystallized to a grain size of several hundred microns (see Table I).

The specimens were reduced by wire-drawing about 50, 70, 80 and 90% reduction in cross-sectional area. The cold-drawn products were heat-treated for 1 h at temperatures from 250 to 630°C. The temperature was controlled to $\pm$ 2 to 3°C. Hardness testing was carried out on a Vickers hardness tester. The values given are each the mean of six indentations; the standard deviations of the means are from one to three

TABLE I Chemical composition and structural parameters.

Material	Grain size (mm) Dia.	Length	Al_2O_3 (wt %)	Fe (wt %)	Si (wt %)	Thickness of Al_2O_3-plates (Å)	Dia. of Al_2O_3 plates (Å)	Particle spacing (μm)	Mesh size in oxide network after reduction §, ‖ (μm)	
MD201-uniform*	0.24	0.3–10	0.6	0.20	0.17	150	460	0.39	—	—
MD201-network*	0.62	0.3–10	0.6	0.20	0.17	150	460	—	1.4(50%)	1.2(90%)
R400-uniform*	0.14	0.3–10	1.2	0.25	0.18	64	450	0.17	—	—
R400-network*	0.28	0.3–10	1.2	0.25	0.18	64	450	—	0.6(50%)	0.5(90%)
Al†		0.28‡	—	0.47	0.11	—	—	—	—	—

*Other impurities: 0.03% Cu, 0.02% max each of Mn, Mg, Zn, Ti.
†Other impurities: 0.09% Mg, 0.05% Mn, 0.02% max each of Cu, Zn, Ti.
‡Equiaxed grains.
§Numbers in brackets are percentage reduction in area in the drawing operation preceding recrystallization.
‖The standard deviation has been estimated at less than 10%.

TABLE II Subgrain sizes after cold deformation and after recovery.

Material	Subgrain sizes μm*					
	50% reduction in area			90% reduction in area		
	Deformed	Recovered	Annealing† temperature (°C)	Deformed	Recovered	Annealing† temperature (°C)
MD201-uniform	0.40	0.48	300	0.29	0.56	250
MD201-network	0.45	0.54	300	0.33	0.43	250
R400-uniform	0.38	0.33	600	0.29	0.36	600
R400-network	0.37	—	—	0.27	—	—
Al	0.48	—	—	0.37	0.46	250

*The standard deviation has been estimated at less than 10%.
†Recovery heat-treatment: 1 h at temperature.

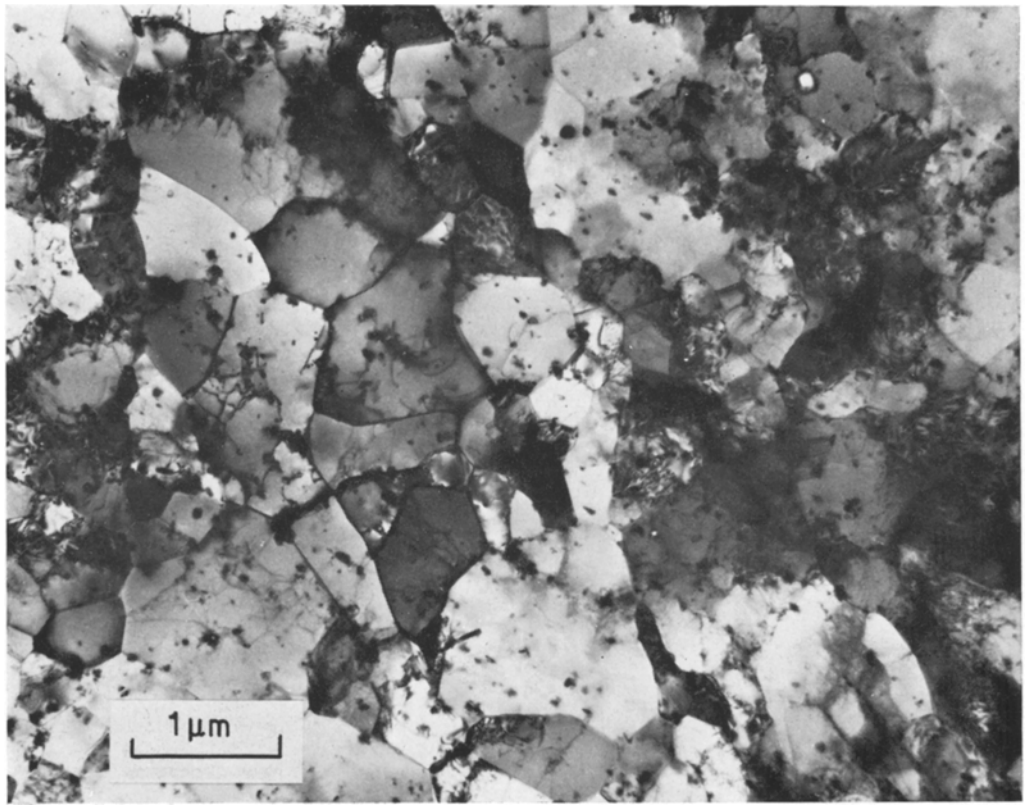

Figure 1 Microstructure of MD 201-uniform, reduced 90% by drawing.

units, the largest standard deviation being found in partly recrystallized products.

The preparation of specimens for optical microscopy is described in [9]. The foils for transmission electron microscopy were prepared by grinding wires on SiC paper to a thickness of 0.2 mm and electropolishing in a solution of 20 vol % perchloric acid in absolute alcohol. A jet technique of thinning was used and the electrolyte was kept at −20°C. The foils were examined with a 100 kV JEM 6A microscope.

3. Results

3.1. Microstructures after cold drawing

After drawing, the microstructures of the Al-Al_2O_3 products were quite similar to that of the Al. Subgrains were present after 50% reduction in area and further deformation reduced the subgrain size and made the subgrain boundaries appear sharper. In Table II are given the subgrain sizes after 50 and 90% reduction. Only small differences between the materials were observed, in agreement with previous results [10]. In the products containing uniformly distributed particles, the pinning of subgrain boundaries by particles was not very pronounced although quite a number of boundaries contained particles and there was a tendency for triple points to form at large particles (Figs. 1 and 2). However, subgrain-boundary pinning by particles was prevalent in the products containing a network structure of particles; most of the oxide boundaries have subgrain boundaries superimposed upon them (Figs. 3 and 4).

A distinct difference in the character of the sub-boundaries is noticed between R400-network and the rest of the products, in that the subgrain boundaries in R400-network appear more tangled and less clearly defined (Fig. 4).

The hardness increment is comparable in all the materials. This is illustrated in Table III by the hardness increment after 90% reduction in area.

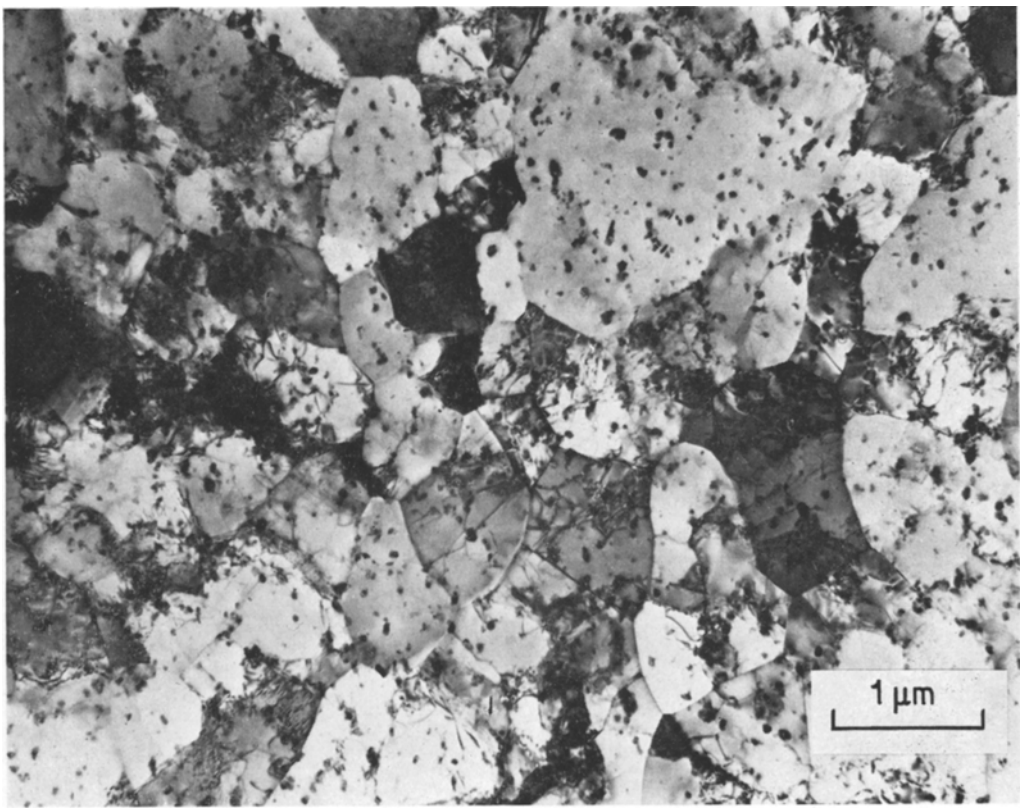

Figure 2 Microstructure of R400-uniform, reduced 50% by drawing.

TABLE III Hardness increment by cold-drawing.

Material	Initial hardness	Hardness after 90% reduction in area	Hardness increment
MD 201-uniform	31	53	22
MD 201-network	28	52	24
R400-uniform	40	59	19
R400-network	37	60	23
Al	22	49	27

3.2. Isochronal recrystallization

The results are plotted in Figs. 5 to 8 as hardness versus annealing temperature. Full recrystallization was obtained in Al and in MD 201 but not in R400. In Fig. 9, the temperature T_r, which corresponds to a hardness equal to the mean of the hardnesses at temperatures for start and completion of recrystallization (taken at 250 and 450°C) is plotted as a function of the reduction in area. In comparing the two MD 201 materials with Al a marked retardation was observed after 50% reduction, whereas a much smaller effect was observed after 80 and 90% reduction. An effect of the oxide distribution is clearly noticeable since the products having a uniform particle distribution recrystallized at a temperature well above that for Al after 50 and 70% reduction, whereas for the network distribution this is the case only after 50% reduction. The recrystallization of R400 is very much retarded compared to MD 201 and the recrystallization curves do not allow determinations of T_r. The effect of the oxide distribution was barely noticeable in R400. Optical microscopy revealed that Al-Al_2O_3 recrystallized with a much larger grain size than Al (Fig. 10). This is in agreement with previous results [9] and data obtained elsewhere [1, 11]. Equiaxed grains were formed in Al whereas the recrystallized grains in Al-Al_2O_3 were cylinder-shaped with a large length-to-diameter ratio and the cylinder axis parallel to the cold drawing (longitudinal) direction (Fig. 11). The recrystallized grain boundaries

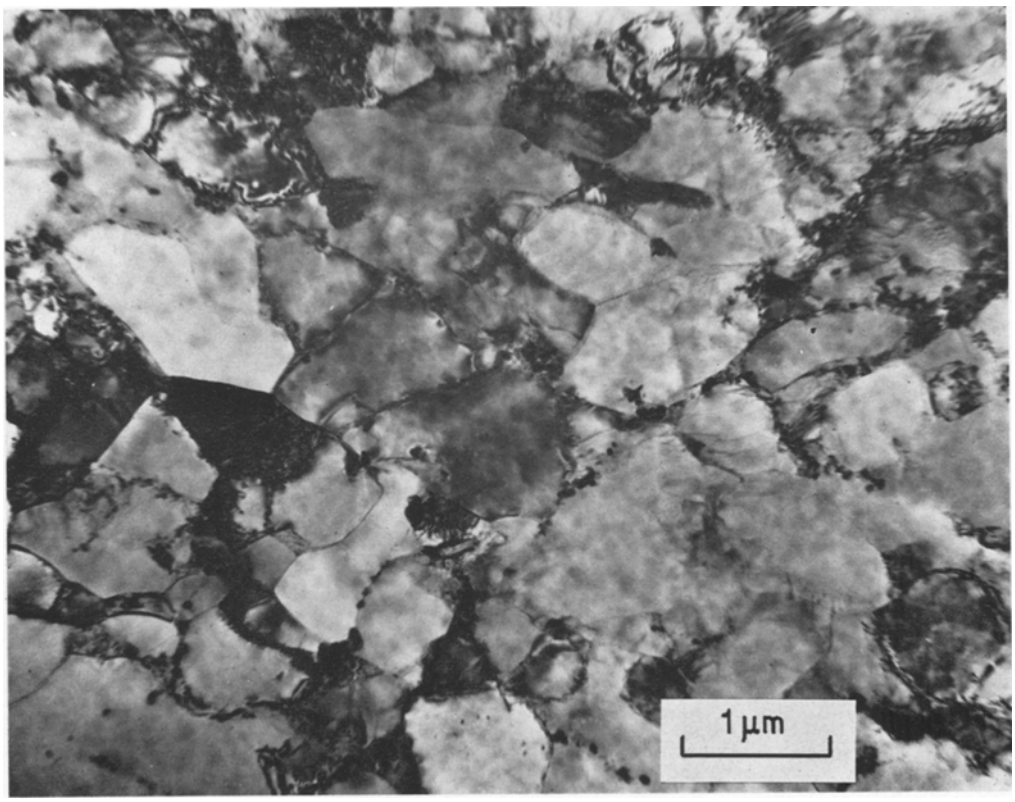

Figure 3 Microstructure of MD 201-network, reduced 50% by drawing.

were fairly smooth and their interaction with particles was not very pronounced; however, pinning of boundaries was occasionally observed.

3.3. Microstructures after recovery

To study structural changes before recrystallization, cold-worked Al and MD 201 were heat-treated for 1 h just below the recrystallization temperature, i.e. 250 and 300°C for products cold-drawn respectively 90 and 50%. R400 products were heat-treated for 1 h at 600°C. As a result of the heat-treatment, the subgrain boundaries became more clearly defined and the dislocation density within the subgrains decreased Recrystallized areas were occasionally observed but the structure normally consisted of subgrains only slightly larger than those observed after cold-working, as shown in Table II. Interactions between sub-boundaries and oxide particles have not changed much (Figs. 12 to 14) although the rows of oxide particles within subgrains in MD 201-network (Fig. 12) imply that the subgrain boundaries originally superimposed upon these oxide particles have become unpinned and have moved away.

4. Discussion

Al_2O_3 particles in Al retard the recrystallization and the present data for low-oxide products together with published results [1, 2] for products containing 2 to 5 wt% Al_2O_3 show enhanced retardation when the content of Al_2O_3 is increased. In the products containing Al_2O_3 particles in a uniform distribution an increase in the content of oxide results in a decrease in the particle spacing which is 0.4 μm and less. A similar retardation of recrystallization with decreasing spacing has been found [3, 12] in Cu-SiO_2 crystals having a particle spacing of 0.5 μm or less. The retarding effect of dispersed particles on the recrystallization may be due to particles affecting the nucleation and growth of recrystallization nuclei, which are taken as small volumes of material entirely or partly surrounded by high angle boundaries. The present data, together with previous observations [1, 11], show

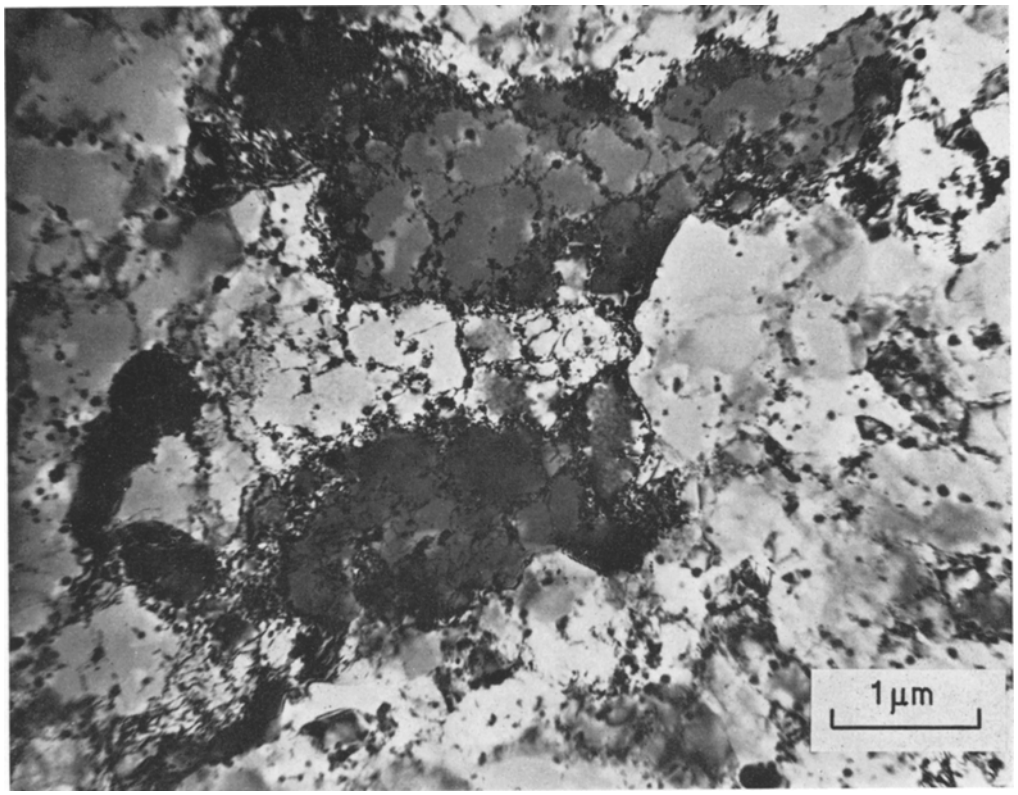

Figure 4 Microstructure of R400-network, reduced 50% by drawing.

that Al-Al_2O_3 products recrystallize with a very coarse grain size compared with aluminium and it is therefore assumed that the retardation of recrystallization is a retardation of nucleation rather than of growth.

The particles may affect the nucleation in different ways. Direct pinning of sub-boundaries by particles may retard or eliminate subgrain growth, thus hindering the formation of larger subgrains or recrystallization nuclei. The effect of particles may also be an indirect one in homogenizing the deformation substructure, thus eliminating regions of high lattice misorientation, suggested to be preferred nucleation sites. These retardation processes will be discussed in the following for Al-Al_2O_3 products.

Pinning of sub-boundaries as a cause of retarded nucleation was originally proposed by Doherty and Martin [13], who observed in Al-Cu alloys after recovery that sub-boundaries were held up at θ-$CuAl_2$ particles. The arrested subgrain size was of the same order as the particle spacing, approximately 1 to 2 µm, and it was suggested that unless the nuclei had become mobile before being pinned by particles, nucleation would be very difficult. In Al-Al_2O_3 products with the particles in a uniform distribution, the subgrains after recovery are larger than the particle spacing, and although sub-boundaries attached to particles were frequently observed, the large majority of sub-boundaries were smooth and unassociated with particles. In Al-Al_2O_3 products with the particles in a network, sub-boundary pinning is observed in the deformed structures, but the occurrence after recovery of rows of oxide particles within subgrains (Fig. 12) shows that unpinning must have taken place. These observations do not directly support the hypothesis by Doherty and Martin. As sub-boundary pinning, however, is frequently observed, it follows that this process *per se* cannot be ruled out as a cause of retarded nucleation.

Retarded recrystallization as due to a more homogeneous dislocation distribution after deformation has been proposed by Brimhall *et al*

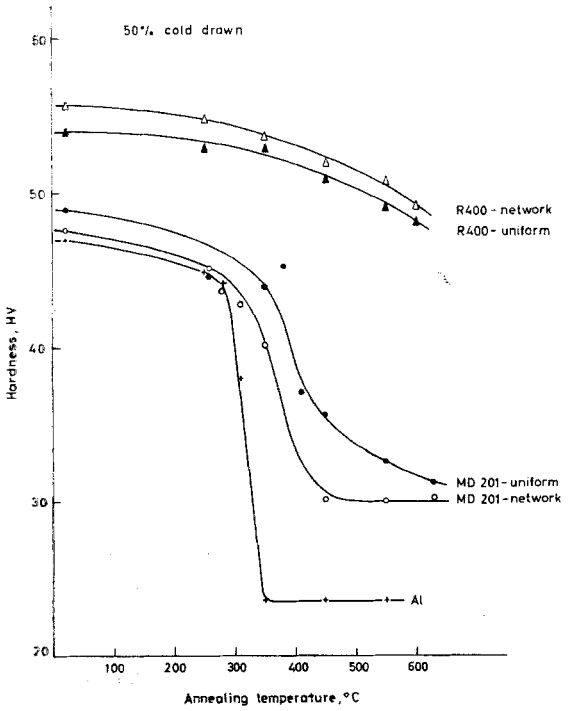

Figure 5 Curves of hardness versus annealing temperature; 50% reduction.

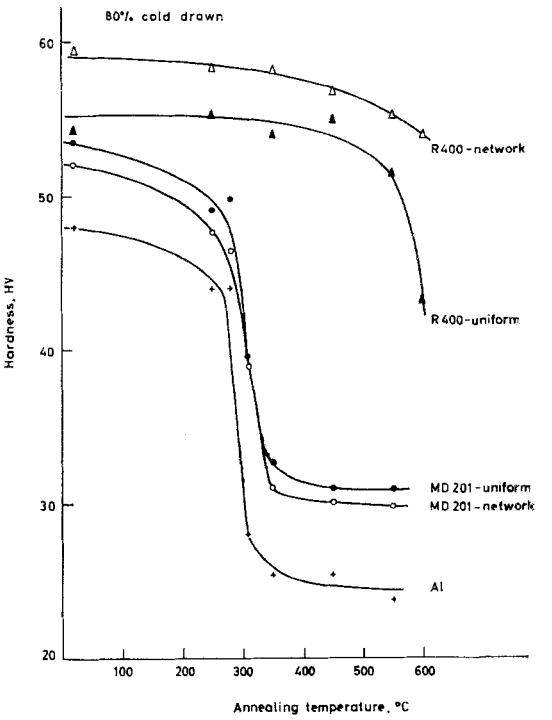

Figure 7 Curves of hardness versus annealing temperature; 80% reduction.

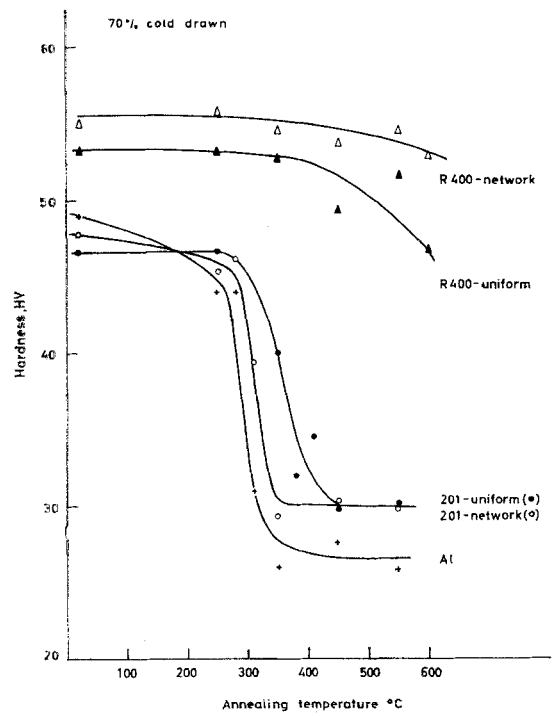

Figure 6 Curves of hardness versus annealing temperature; 70% reduction.

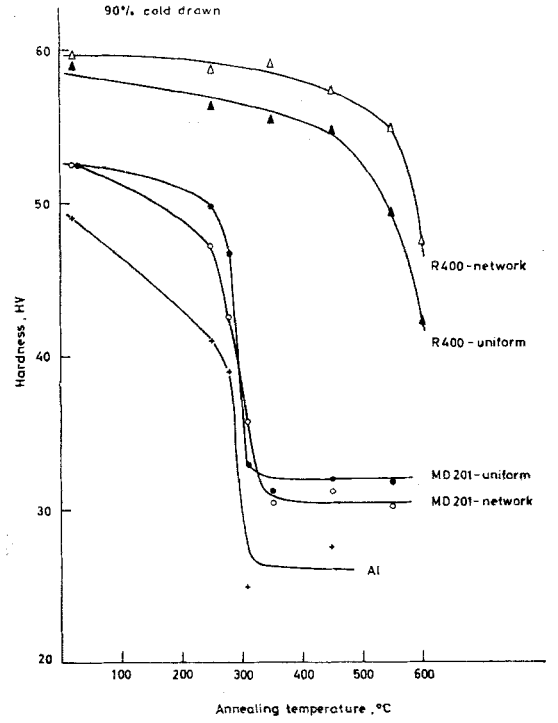

Figure 8 Curves of hardness versus annealing temperature; 90% reduction.

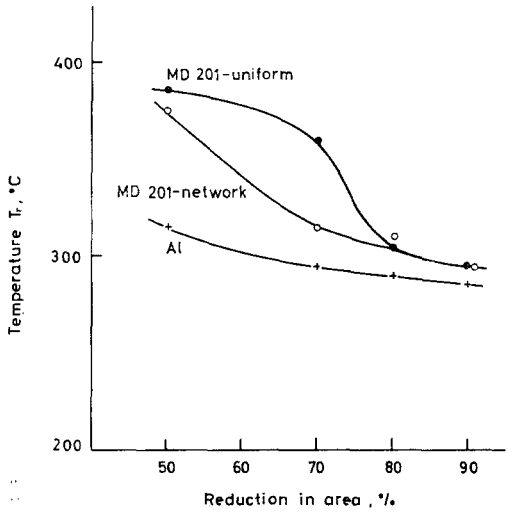

Figure 9 Recrystallization temperature versus degree of cold drawing.

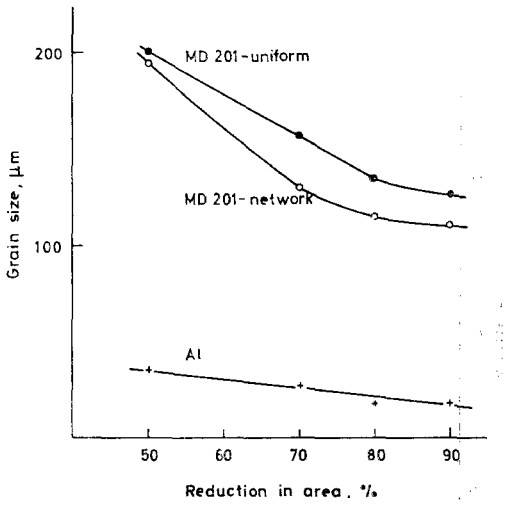

Figure 10 Recrystallized grain diameter (MD 201) and recrystallized grain size (Al) versus degree of cold-drawing.

[4] for Ag-MgO products. The lattice misorientation is reduced, thus a decrease in nucleation rate is expected. This hypothesis is supported by observations in Cu-SiO_2 crystals [12] and Ni-ThO_2 products [14]. The present observations show that Al-Al_2O_3 products behave differently since the deformation structures in both MD 201 products and in R400-uniform consisted of well defined subgrains similar in size and appearance to those found in Al. Also the misorientation across sub-boundaries is little affected when particles are introduced into aluminium; measurements on cold-drawn [10]

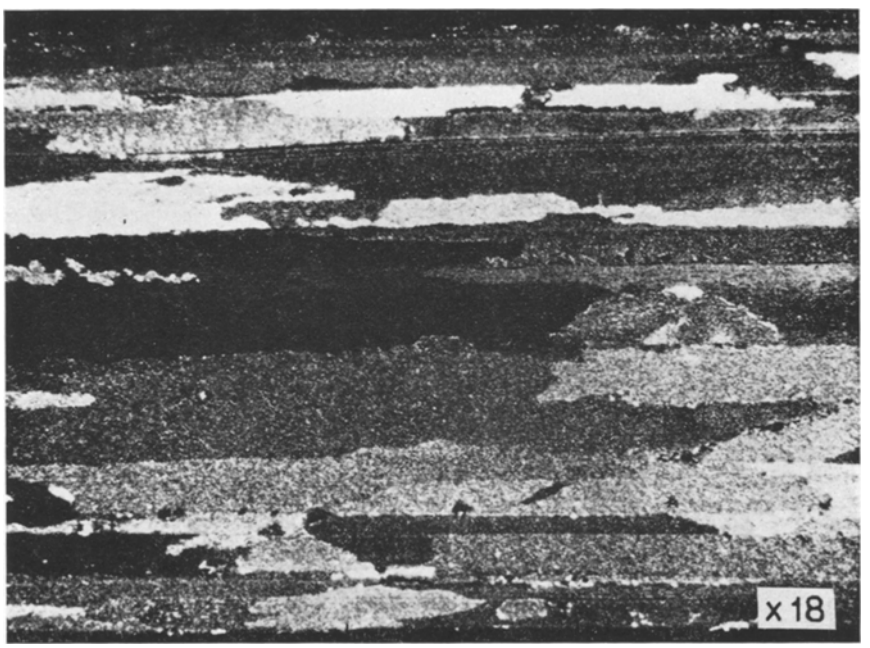

Figure 11 Microstructure of MD 201-uniform, reduced 50% by cold-drawing and recrystallized. Longitudinal section. Etched, magnification × 30.

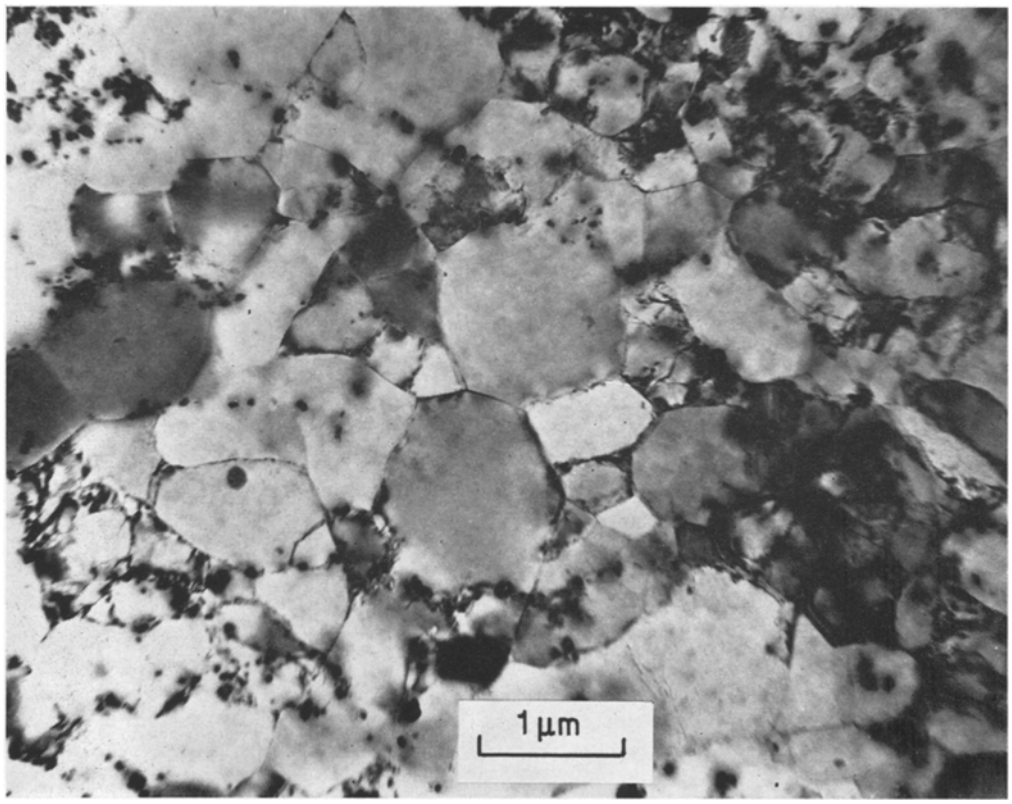

Figure 12 Microstructure of MD 201-network, reduced 90% by cold-drawing and heat-treated for 1 h at 250°C.

and cold-rolled [2] Al-Al_2O_3 products give misorientation angles respectively of the same magnitude, and twice as large, as those found in Al. An indication of a homogenizing effect of particles was found, however, in R400-network which showed more tangled sub-boundaries than the rest of the products. The difference in deformation structures observed between Al-Al_2O_3 and Ag-MgO, Ni-ThO_2 and Cu-SiO_2 cannot be explained unambiguously; differences in stacking-fault energies of the matrix metals, variations in degree and type of cold deformation and different dispersed-phase parameters may all be factors of importance.

In considering other ways in which dispersed particles may affect nucleation, a possibility exists that the movement of *individual dislocations* during recovery is limited owing to pinning by particles. Individual dislocations are present within the subgrains after deformation and the particles may limit their movement to the sub-boundaries, thereby reducing the speed at which the sub-boundary misorientation increases during recovery. Another possibility is that subgrain growth during recovery involves the movement of individual dislocations from a sub-boundary to neighbouring boundaries, and that this movement is limited by the particles. In MD 201 the dislocation density within the subgrains is fairly small after deformation and the latter process may be the most important. In comparison, the subgrains in R400 after deformation and after recovery contain a larger number of dislocations (Figs. 2 and 14) and both processes may be effective. The mechanism of particles acting as barriers to moving dislocations may not only influence the recrystallization behaviour but also the high-temperature strength of dispersion-strengthened products. In this context it is of interest to notice that a decrease in particle spacing, which is the most effective way to increase the high-temperature strength, strongly retards recrystallization.

Cold deformation may be an important parameter when studying the recrystallization of dispersion-strengthened products. The deforma-

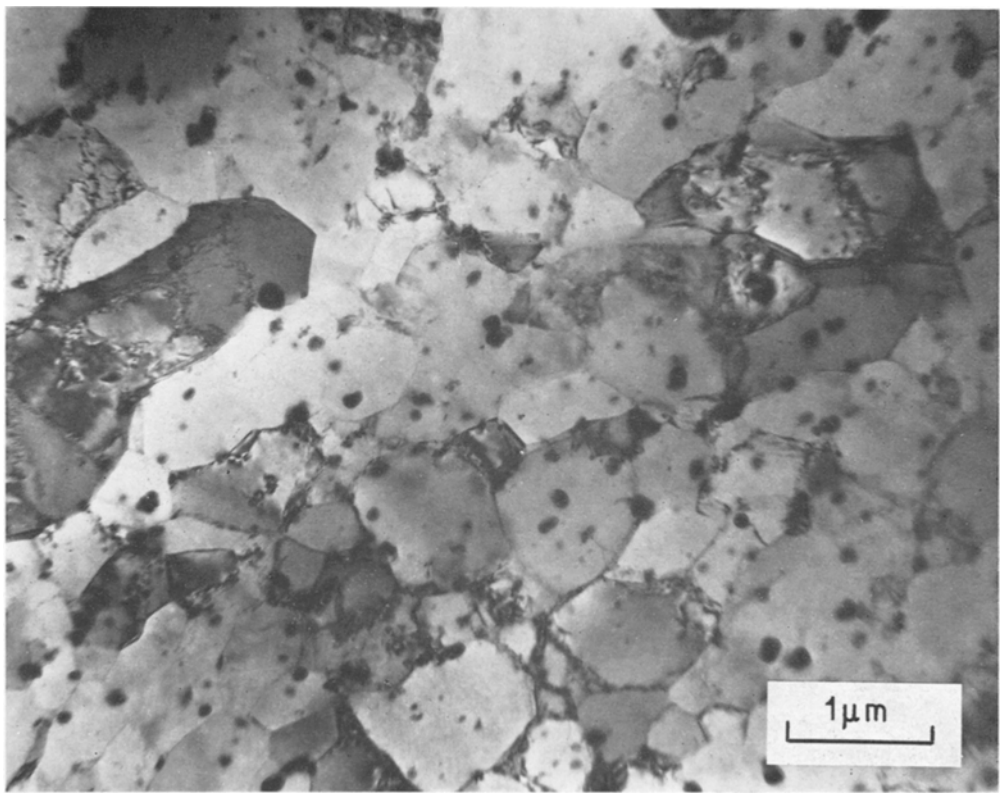

Figure 13 Microstructure of MD 201-uniform, reduced 90% by cold-drawing and heat-treated for 1 h at 250°C.

tion affects the misorientation across sub-boundaries [15], thus the critical size of the recrystallization nuclei may also be affected. This has been found [7] in pure aluminium (99.99%), annealed to the point of recrystallization, in which the size of "newly created recrystallized grains" decreased from about 30 to about 5 μm when the reduction by cold-rolling was increased from 30 to 98%. In assuming a similar effect in particle-containing materials a correlation between the roles of the degree of cold deformation and the particle spacing may exist, i.e. the effect of particles on nucleation may be largely reduced or disappear if by increasing the cold deformation the critical size of the nucleus can be reduced to be of the same order or smaller than the particle spacing. Measurement of nucleus sizes in dispersion-hardened products have to our knowledge not been carried out. In Al-Al_2O_3 the nucleus size may, however, be of the same order or smaller than in Al, since it has been observed that (a) the misorientation across sub-boundaries in Al-Al_2O_3 is equal to or larger than the misorientation found in Al, and (b) grains approximately 1 μm in size surrounded by high-angle boundaries are present after recovery [16]. The amount of stored energy introduced by deformation is assumed to be quite unaffected by the presence of oxide particles, as the reduction in subgrain size and the hardness increment at increasing degrees of deformation is practically the same in all the materials; it therefore follows that if the critical size of the recrystallization nuclei is assumed to be of the same order or slightly larger than the particle spacing in MD 201 the retardation of recrystallization in these products compared to Al would be expected to decrease when the degree of cold deformation is increased. This is supported by the experiments showing that after 50% reduction, the recrystallization temperature is 60 to 70°C higher than in Al, whereas after 90% reduction this difference has decreased to approximately 10°C. A correlation between the effects of the degree of cold work and the particle spacing may explain for MD 201 that network structures show less

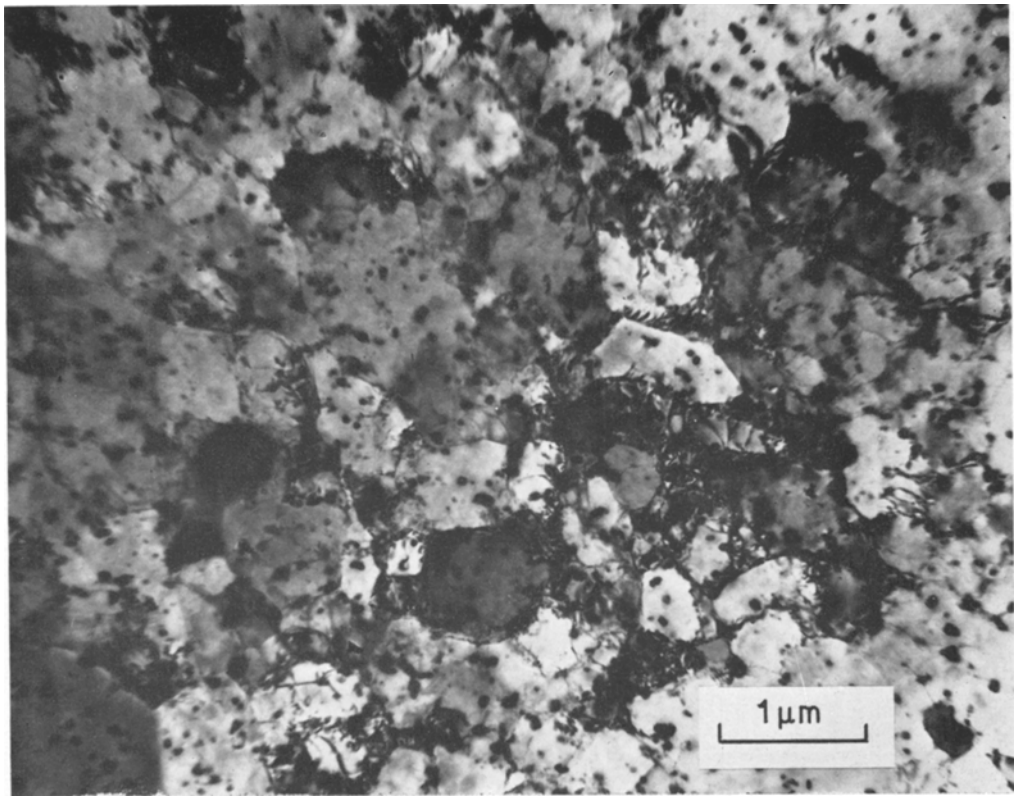

Figure 14 Microstructure of R400-uniform, reduced 50% by cold-drawing and heat-treated for 1 h at 600°C.

resistance to recrystallization than uniform structures; oxide-free areas in the network structure having an average mesh size of 1 to 2 μm may facilitate the formation of recrystallization nuclei compared with the uniform structures where the particle spacing is below 0.5 μm. This structural difference may also explain the observation (Fig. 9) that the major reduction in recrystallization temperature takes place at a lower degree of cold deformation in the network structures than in the uniform structures. In R400 the retarded recrystallization at all degrees of cold deformation and for both types of structures indicate that the size of the recrystallization nuclei may be large compared to the particle spacing, and to the mesh size.

5. Conclusions

(1) The recrystallization of Al-Al_2O_3 containing 0.6 and 1.2 wt % Al_2O_3 is retarded compared with Al. The important structural parameters involved in this retardation process are the oxide content (equivalent to the reciprocal particle spacing) and the distribution of Al_2O_3 particles either uniformly or in a network.

(2) The subgrain size after cold drawing 50 and 90% is less than 0.5 μm and is not affected to a great extent by either the content or the distribution of Al_2O_3 particles. Recovery near the recrystallization temperatures produces only slight increases in the subgrain size. From the microstructures after cold work and after recovery it is suggested that particle-retarded recrystallization may be caused by pinning during the recovery stage of sub-boundaries and of individual dislocations. The hypothesis of retardation of recrystallization as due to particle-enhanced homogenization of dislocation structures during deformation is not supported by the microstructural observations.

(3) In Al-Al_2O_3 containing 0.6 wt % Al_2O_3 and cold-drawn 50% the recrystallization is markedly retarded compared to Al, whereas little retardation is observed after either 80 or 90% reduction. An increase in the degree of cold deformation may reduce the critical size of the recrystalliza-

tion nuclei, thus the retarding effect of particles during nucleation may be reduced or disappear. It is therefore suggested that the degree of cold deformation and the particle spacing may be interdependent parameters when determining the recrystallization behaviour of dispersion-strengthened products.

Acknowledgements

The authors are grateful to J. Bachmann, J. Lindbo, P. Nielsen and L. Tankmar for discussions and assistance in the experimental work.

References

1. E. J. WESTERMAN and F. V. LENEL, *Trans. Met. Soc. AIME* **218** (1960) 1010.
2. D. NOBILI and R. DE MARIA, *J. Nucl. Mat.* **17** (1965) 5.
3. F. J. HUMPHREYS and J. W. MARTIN, *Acta Metallurgica* **14** (1966) 775.
4. J. L. BRIMHALL, M. J. KLEIN, and R. A. HUGGINS, *ibid* **14** (1966) 459.
5. P. R. MOULD and P. COTTERILL, *J. Mater. Sci.* **2** (1967) 241.
6. N. HANSEN, *Acta Metallurgica* **17** (1969) 637.
7. B. BAY, *Scripta Met.* **4** (1970) 489.
8. N. HANSEN, *Acta Metallurgica* **18** (1970) 137.
9. *Idem, Trans. Met. Soc. AIME* **245** (1969) 1305.
10. *Idem, ibid* **245** (1969) 2061.
11. R. J. TOWNER, *ibid* **230** (1964) 505.
12. F. J. HUMPHREYS and J. W. MARTIN, *Phil. Mag.* **17** (1968) 365.
13. R. D. DOHERTY and J. W. MARTIN, *J. Inst. Met.* **91** (1962) 332.
14. M. A. CLEGG and J. A. LUND, *Met. Trans.* **2** (1971) 2495.
15. P. B. HIRSCH and J. N. KELLAR, *Acta Cryst.* **5** (1952) 162.
16. G. F. MISSIROLI, D. NOBILI, and F. ZIGNANI, *J. Nucl. Mat.* **21** (1967) 199.

Received 22 February and accepted 6 March 1972.