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SELF DIFFUSION IN MAGNESIUM OXIDE

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Quantitative measurements were made of the shrinkage rate of dislocation loops, $b = \frac{1}{2}[101]$, which were near the center of the foil. The effect of loop to sink distance on the rate of climb has been directly confirmed. A value of $110,000 \pm 4,200$ cal/mole for the activation energy for bulk diffusion and $(1.37 \pm 0.26) \times 10^{-2}$ cm²/sec for the pre-exponential factor were obtained. Comparison of these results with tracer and electrical conductivity measurements suggest that oxygen ion diffusion is the rate controlling process for dislocation climb.

INTRODUCTION

Self diffusion studies on MgO have been reported by tracer diffusivity measurements.^{1,2} For cation self diffusion, a layer of Mg²⁸ (21.3 hr. half life) is deposited on one side of an MgO single crystal and heated to high temperature. On heating, the radioactive tracer atoms are distributed throughout the crystal. The final distribution and consequently the diffusion coefficient is evaluated by sectioning. Oxygen self diffusion coefficients have also been determined by measuring the rate of exchange between a limited volume gas phase enriched with the isotope O¹⁸ and MgO powder.² One uncertainty in this

kind of tracer diffusivity experiment is caused by the presence of dislocations which can act as fast diffusion pipes. Dislocation climb involves diffusion of both anion and cation. The rate of diffusion will be controlled by the slower moving species. By studying the shrinkage of small dislocation loops due to transport of material to or from the surface of a thin foil it should be possible to obtain quantitative information on the diffusion of the slower diffusing ion. By selecting loops that are relatively isolated from other loops, the mechanism would be unambiguously volume diffusion. The only experimental work on dislocation climb in MgO available in the literature is that of Groves and Kelly.³ However, since their observations were based on thin foils prepared from bulk samples after different annealing treatments and the climb of individual dislocations was not observed it was impossible to separate volume diffusion and pipe diffusion. By developing techniques of annealing thin foils outside the electron microscope and of photographing the same area under identical diffraction conditions, we have been able to study volume and pipe diffusion separately. Measurements of the shrinkage rate of the same dislocation loops were made at more than one temperature to eliminate errors in the calculation of activation energy due to poorly known pre-exponential factors.

EXPERIMENTAL

Large grained polycrystalline MgO was purchased from Muscle Shoals Electro-chemical Corp., Tuscumbia, Alabama. Semi-quantitative spectrographic analysis revealed the following impurities (in PPM):

Al-200, Si-200, Fe-30

Na and K were undetected up to 2 PPM, Ag undetected up to 1 PPM, all other elements also undetected.

Single crystal specimens in the form of thin sheets ($\sim 0.50 - 0.25$ mm thick) were obtained by cleaving along $\{100\}$ planes. The surface damage introduced during cleaving was removed by chemical polishing in hot orthophosphoric acid ($150-160^\circ\text{C}$) to a thickness of about 0.3 mm. These sheets were then bent plastically backwards and forwards (± 5 cm radius) about 200 times until they were full of slip bands. Following the deformation the specimens were thinned further, after applying masking lacquer around the edges. Final thinning to obtain electron microscope foils was done by a jet polishing technique.⁴ After cold working the thinning was done primarily from one side because plastic deformation was maximum near the surfaces.

In thin sheets bent along the $[010]$ axis, dislocation dipoles are introduced on $(101)[\bar{1}01]$, $(\bar{1}01)[101]$ and on $(1\bar{1}0)[\bar{1}\bar{1}0]$, $(\bar{1}\bar{1}0)[110]$ slip systems. Dipoles on $(101)[\bar{1}01]$ and $(\bar{1}01)[101]$ are of primary interest as dipoles on $(1\bar{1}0)[\bar{1}\bar{1}0]$ are seen edge-on along $[001]$ which is perpendicular to the surfaces of the foil.

Annealing Technique: An MgO (98%) crucible with a tight platinum lid was fired at 1500°C for 48 hours. Then an MgO tube 1/16 in. internal diameter and 14 in. long was fitted in the crucible so that the atmosphere inside the crucible could be isolated from the furnace atmosphere and could be controlled from outside. This was also given a firing treatment similar to that of the crucible before connecting it to the crucible. The thin foil was kept between two pure MgO crystals. One of these crystals had a spherical cavity, made by an ultrasonic drill. The thin foil was placed in the cavity in such a way that the area of interest did not touch the enclosing crystals. A calibrated Pt-10% Rh thermocouple was connected to the crucible to measure the temperature inside the crucible. The temperature could be controlled to better than $\pm 5^{\circ}\text{C}$. For temperatures above 1400°C, there was some indication that the specimens became slightly non-stoichiometric. This was easily detected as a change in transmitted intensity inside the electron microscope. The problem was overcome by introducing some oxygen in the crucible atmosphere. Care was taken not to turn the foil over during electron microscope observations.

Electron Microscopy: All the foils were examined in a Siemens 100 kV electron microscope. For a particular set, the same diffraction conditions were used. Stereomicroscopy was done along the 200 Kikuchi band and around the 001 pole at each stage in order to be able to locate positions of all loops. Objective lens current was measured at each step of the picture making, keeping intermediate and projector lens currents fixed. The microscope was calibrated in small intervals

of objective lens current for the same projector and intermediate lens currents. The measured positions were always taken as the average of g_{+ve} and g_{-ve} diffraction conditions.

All the quantitative measurements were made directly from the electron micrograph plates. Electron micrographs were observed in a Nikon enlarger at 20 $\times$ to make measurements. The thickness of the foil (2L) was determined to $\pm 10\text{\AA}$ by measuring the parallax of surface reference points (ends of screw dislocations running across the foil or dirt particles on the foil surfaces) in the stereo pairs. The following relation was used:

$$\text{Thickness of foil (2L)} = \frac{\text{Parallax}}{2 \sin(\theta)}$$

where ' θ ' is the angle of tilt between stereo pairs.

THEORY

The influence of the sink to loop distance on the climb process was demonstrated by observing the climb behavior of loops near surfaces of the foil or near dislocations. Figure 1 shows the same area after various annealing treatments at 1195°C. The dislocation loop at 1, $b = \frac{1}{2}[101]$, is near one of the surfaces of the foil; and the loop at 2, $b = \frac{1}{2}[101]$, is near a larger dislocation loop. Both these loops annealed out faster than the very small dislocation loop with $b = \frac{1}{2}[101]$ at 3 which is near the center of the foil. The latter has the greater driving force for shrinkage because of its small size. These observations show that the climb rate is controlled by diffusion of vacancies between the loop and the surface rather than emission or absorption of vacancies at the loop. In fcc metals this is also found to be true when

dislocations are not widely split into shockley partials.^{5,6}

In the case of diffusion-controlled climb it is not necessary to make any detailed assumptions about the state of the dislocation core other than that it can maintain the vacancy concentration in the lattice surrounding the loop at its local equilibrium concentration. The rate of climb is then determined by the vacancy flux between the lattice surrounding the loop and surfaces of the foil. The vacancy concentration in the vicinity of a dislocation loop is $C_0 \exp \left[\frac{F_c B^2}{kT} \right]$, where F_c is the driving force for climb, B^2 is the cross-sectional area of a vacancy and C_0 is the vacancy concentration at the surfaces of the foil. When the radius of the dislocation loop (r) is small compared with the foil thickness, the loop can be considered as a sphere of radius r and the diffusion equation is solved for spherical symmetry, with the boundary conditions that vacancy concentration:

$C = C_0 \exp \left[\frac{F_c B^2}{kT} \right]$ at $x = r$ and $C = C_0$ at $x = L$. Here x is measured from the center of the dislocation loop, and $2L$ is the thickness of the foil. The rate of shrinkage, dr/dt (following Dobson et al.⁵) for this case is given by

$$\frac{dr}{dt} = - \frac{2D}{b} \exp \left[\frac{F_c B^2}{kT} - 1 \right] , \quad (1)$$

where D is the self bulk diffusion coefficient for the rate controlling species.

For large loop radii ($r > L$), one loop can be approximated by a straight line of length $2\pi r$ and the diffusion equation is solved for cylindrical symmetry with the boundary conditions that $C = C_0 \exp[F_c B^2/kT]$ at $x = b$ and $C = C_0$ at $x = L$, where x is measured from the dislocation

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line. The rate of shrinkage dr/dt for this case is given by

$$\frac{dr}{dt} = -2 \pi \frac{D}{b} \ln \left(\frac{L}{b} \right) [\exp(F_c B^2/kT) - 1] \quad (2)$$

Following Seidman and Balluffi,⁷ for small loops the diffusion geometry may be better represented by a toroidal source (radii r and r_o and concentration C_1 on the torus) situated at the center of a sphere of radius L . The concentration on the surface of the sphere is C_o . The shrinkage rate is given by

$$\frac{dr}{dt} \approx \frac{-2 \pi D}{b} \frac{(C_1 - C_o)}{\ln(8r/r_o)} \quad (3)$$

For large loop radii, the rate equation corresponding to Eq. (2) is

$$\frac{dr}{dt} = \frac{-2 \pi D}{b} \frac{(C_1 - C_o)}{\ln\left(\frac{4L}{\pi r_o}\right)} \quad (4)$$

Using the expression given by Bacon and Crocker⁸ for the elastic energy of a prismatic dislocation loop, F_c can be written as

$$F_c = \frac{\mu b^2}{4 \pi (1 - \nu) r} \left[\ln \left(\frac{8r}{r_o} \right) - 2 + \frac{3 - 2\nu}{4(1 - \nu)} \right]$$

where ν is the Poisson's ratio and r_o is the radius of the core of a dislocation.

Substituting for $\nu \approx 0.3$ in the range of temperature 1080-1450°C and $r_o \approx 1.5b$, F_c can be rewritten as

$$F_c = \frac{\mu b^2}{2.8 \pi r} \ln \frac{1.7r}{b} \quad (6)$$

Since $F_c b^2 \ll kT$, the Eq. (1) can be rewritten as

$$\frac{dr}{dt} = \frac{-2D}{b} \frac{F_c b^2}{kT}, \quad (7)$$

because $B^2 \approx b^2$, for $\frac{1}{2} \langle 101 \rangle$ prismatic edge dislocation loops.

By substituting for F_c in Eq. (7) and simplifying, we have

$$\frac{r dr}{\ln \frac{1.7r}{b}} = \frac{-\mu D b^3}{1.4 \pi kT} dt \quad (8)$$

Similarly Eq. (2) can be written as

$$\frac{r dr}{\ln \frac{1.7r}{b}} = \frac{-\mu D b^3 \ln(L/b)}{1.4 \pi kT} dt \quad (9)$$

By integrating numerically the left hand side of Eq. (8) and Eq. (9) from r_1 to r_2 corresponding to a given interval of time of annealing at a temperature T , D can be determined. Finding D values at different temperatures and recalling $D = D_0 \exp(-E_{act}/kT)$, the activation energy (E_{act}) can be determined. Using this value of activation energy the pre-exponential term D_0 was then calculated.

RESULTS AND DISCUSSION

Figure 2 is a set of pictures of the same area after various annealing treatments at four different temperatures (1202°C, 1291°C, 1368°C and 1243°C). The pictures represent the end points of annealing treatments at one temperature. Dislocation loops 1, 2, 3 and 4 ($b = \frac{1}{2}[101]$) remained roughly near the center of the foil. This was confirmed by stereopairs taken after each annealing treatment. Dirt particles on the surfaces of the foil provided convenient surface reference points. The set in Fig. 3 is at two different annealing

temperatures (1260°C and 1400°C) and another set in Fig. 4 is at two different temperatures (1400°C and 1427°C). Plots (r^2 vs t) for the loops which remained near the center of the foil are given in Figs. 5 through 7.

Using Eqs. (8) and (9) an average value of activation energy $110,000 \pm 4,200$ cal/mole was obtained in the temperature range 1100 to 1427°C. The value of D_0 in this temperature range was found to be $(1.37 \pm 0.26) \times 10^{-2}$ cm²/sec. In calculations of activation energy direct vacancy elastic interaction with the dislocation loop has been neglected. For a large dislocation loop, where the stress field can be approximated by the stress field of a straight dislocation line, the elastic interaction of Vacancies due to change in their size (because of lattice relaxation) and shear modulus is given in the Appendix. Hickman and Walker⁹ found that the nature of the dilation around an isolated Schottky defects in MgO is contraction and varies from 0.2 to 0.4 times the molecular volume. As shown in the Appendix, a dilation of 10 to 40% could increase the shrinkage rate as much as 50%.

The value of activation energy agrees with the activation energy which has been associated with intrinsic diffusivity of oxygen ions obtained from electrical conductivity measurements.¹⁰ For shrinkage of dislocation loops both ions Mg^{++} and O^{--} must diffuse at the same time, but the rate of diffusion is probably controlled by oxygen ion mobility. This is expected because of the much larger ionic radius of O^{--} (O^{--} (1.32 Å), Mg^{++} (0.66 Å)).

Oishi and Kingery¹ measured the oxygen self diffusion coefficient by measuring the rate of exchange between a controlled gas phase (oxygen gas at 150 mm pressure) enriched and isotope O^{18} and MgO

powder in the temperature range 1300-1750°C. Their value of activation energy for self diffusion of oxygen ion was 62,400 cal/mole and they attributed this to impurity controlled or structure sensitive diffusion. If we take this value as that corresponding to extrinsic or impurity controlled diffusion of oxygen, then it represents H_m , the enthalpy of motion of oxygen ions. Oxygen vacancies are created due to the presence of aliovalent impurities. The activation energy 110,000 $\pm$ 4200 cal/mole obtained in air experiments probably corresponds to intrinsic diffusivity of O^{--} and therefore, corresponds to $\frac{H_f}{2} + H_m$ where H_f is the enthalpy of formation of a Schottky defect. From this H_f is 3.6 $\pm$ 0.22 eV and H_m is 2.76 $\pm$ 0.11 eV. Harding et al.¹¹ did measurements of self diffusion of Mg^{++} in single crystals of MgO and determined $H_f = 3.4 \pm 0.2$ eV and the enthalpy of motion for cation vacancy (H_m, Mg^{++}) = 1.7 $\pm$ 0.1 eV. There is good agreement in the values of H_f . Both in anion and cation diffusion H_f corresponds to the enthalpy of formation of the complete Schottky defect. The value of H_m for oxygen ion migration deduced from electrical conductivity measurements is 2.7 eV.¹⁰ The higher value of H_m for oxygen ions (2.76 $\pm$ 0.11 eV) compared to that of Mg^{++} (1.7 $\pm$ 0.1 eV) is also expected from ionic radii considerations.¹²

Oishi and Kingery's value of activation energy, 62,400 cal/mole is also the same as the activation energy for pipe diffusion, reported by the present authors.¹³ Therefore instead of interpreting their results as extrinsic diffusion of oxygen they might also be interpreted as pipe diffusion. Oishi and Kingery probably used unannealed MgO powder in their experiment. These particles might have contained a high dislocation density. This has been found to be true for NaCl and KCl

powders as discussed by Barr et al.¹⁴ The relatively high values of the diffusion coefficient obtained by Oishi and Kingery support this idea.¹⁴ The equality of the activation energy for bulk diffusion under an impurity controlled diffusion condition to the activation energy for pipe diffusion along dislocations is just a coincidence.

Groves and Kelly³ measured the sizes of dislocation loops by transmission electron microscopy in thin foils prepared from cold worked samples of various annealing treatments at 1242°C, 1333°C and 1426°C. They determined the average growth rate at these temperatures and found the activation energy of $76,000 \pm 4600$ cal/mole. By photographing the same area in transmission electron microscope under identical diffraction conditions¹⁵ after various annealing treatments outside the electron microscope, the present authors have found that the primary mechanism of loop coarsening below about 1300°C is the self climb. Most of the close loop pairs, which result from breaking up of dipoles during the beginning stages of annealing in the cold-worked samples, coalesce by self climb below about 1300°C. Only very few big loops grow at the expense of smaller loops by bulk diffusion. In thin foils isolated loops can shrink only by bulk diffusion. Therefore, the apparent activation energy of $76,000 \pm 4600$ cal/mole obtained by Groves and Kelly³ probably represents contributions from both volume diffusion and pipe diffusion.

CONCLUSIONS

1. The shrinkage or growth of dislocation loops in MgO is controlled by diffusion of vacancies rather than emission from the dislocation.
2. The rate controlling ion during diffusion of Mg^{++} and O^{--} is the oxygen ion.
3. The activation energy of self-diffusion of oxygen ion has been interpreted to be $110,000 \pm 4,200$ cal/mole and the value of the pre-exponential was found to be $(1.37 \pm 0.26) \times 10^{-2} \text{ cm}^2/\text{sec}$. This method of loop-shrinkage for the measurements of self diffusion, using the technique developed by the present authors would prove very useful in studying precisely the effects of various impurities on self diffusion, as MgO is a very good host for many impurities.

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APPENDIX:

EFFECT OF ELASTIC INTERACTION ON THE RATE OF CLIMB

The elastic interaction arises from two effects (i) the size effect because of interaction between the stress fields of the vacancy and the dislocation, and (ii) the inhomogeneity interaction, which arises because of different elastic constants associated with the vacancy. After approximating vacancy-loop-interaction by vacancy-straight edge dislocation interaction and neglecting orientation dependence of the potential,¹⁶ the size effect interactions can be written as:

$$E_1 = \frac{-4\mu\epsilon b r_o^3}{r} = \frac{-W_1}{r}, \quad (1)$$

and inhomogeneity interactions as:

$$E_2 = \frac{-5\mu b^2 r_o^3 (1-\epsilon)^3}{2\pi (1-\nu)(7-5\nu)r^2} = \frac{-W_2}{r^2}, \quad (2)$$

where $r_o (1-\epsilon)$ is the radius of the vacancy and r_o the atomic radius.

This binding energy will increase the vacancy concentration in equilibrium with the loop; and while this will tend to increase the rate of climb, the increase in the apparent activation energy for diffusion will tend to decrease the climb rate. If the interactions were completely localized at the dislocation core the two effects would cancel out, but since the decreased climb effect occurs over relatively long ranges the net effect of the interaction is to increase the rate of climb.

Following Gibbs and Turnbull¹⁷ for the size effect the ratio of vacancy fluxes with and without interactions is given by:

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$$\begin{aligned} \left(\frac{dN}{dt} \right)_b / \left(\frac{dN}{dt} \right)_d &= \frac{\ln(L/b)}{\int_{r_0}^L \frac{1}{r} \exp \left(-\frac{W_1}{rkT} \right) dr} \\ &= \ln(L/b) / \left[\log \frac{1}{r} + \frac{a}{r} + \frac{a^2}{r^2 \cdot 2 \cdot 2!} + \frac{a^3}{r^3 \cdot 3 \cdot 3!} + \dots \right]_{r_0}^L \quad (3) \end{aligned}$$

$$\text{where } a = \frac{-W_1}{kT}.$$

Similarly for inhomogeneity interactions:

$$\begin{aligned} \left(\frac{dN}{dt} \right)_b / \left(\frac{dN}{dt} \right)_d &= \frac{\ln(L/b)}{\int_{r_0}^L \frac{1}{r} \exp \left(-\frac{W_2}{r^2 kT} \right) dr} \\ &= \ln(L/b) / \left[-\frac{1}{2} \left[\log \frac{1}{r^2} + \frac{c}{r^2} + \frac{c^2}{r^4 \cdot 2 \cdot 2!} + \frac{c^3}{r^6 \cdot 3 \cdot 3!} + \dots \right] \right]_{r_0}^L \quad (4) \end{aligned}$$

$$\text{where } c = -\frac{W_2}{kT}.$$

For core radius about $2b$ and maximum dilation of 0.4 the climb rate increases as much as 50% .

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FIGURE CAPTIONS:

- Fig. 1. Effect of loop to sink distance on the shrinkage rate of dislocation loops. The dislocation loops at 2 and 1 are shrinking at much faster rates than other loops of similar sizes because of being near to a very big loop and surface of the foil respectively. In all figures the length of the marker is 0.3μ .
- Fig. 2. Shrinkage of dislocation loops $b = \frac{1}{2} [101]$, due to lattice diffusion. Annealing treatments A $\rightarrow$ B, 42 min 5 secs at 1368°C ; B $\rightarrow$ C, 45 min 52 secs at 1291°C ; C $\rightarrow$ D, 150 min 8 secs at 1202°C ; D $\rightarrow$ E, 46 min 10 secs at 1243°C . Intermediate pictures taken at one temperature are not shown. In all plates to follow the indicated magnification applies to particular photograph, other photographs in the plate have slight variations.
- Fig. 3. Annealing of dislocation loops, $b = \frac{1}{2} [101]$. Annealing treatments: A $\rightarrow$ B, 64 min 0 secs at 1260°C ; B $\rightarrow$ C, 80 min 0 secs at 1260°C ; C $\rightarrow$ D, 20 min 10 secs at 1400°C ; D $\rightarrow$ E, 20 min 5 secs at 1400°C .
- Fig. 4. Climb of dislocation loops, $b = \frac{1}{2} [101]$. Annealing treatments: A $\rightarrow$ B, 11 min 0 secs at 1427°C ; B $\rightarrow$ C, 16 min 0 secs at 1427°C ; C $\rightarrow$ D, 24 min 35 secs at 1400°C ; D $\rightarrow$ E, 9 min 1 sec at 1400°C .
- Fig. 5. Annealing curves of loops in Fig. 2.
- Fig. 6. Annealing curves of dislocation loops in Fig. 3.
- Fig. 7. Annealing curves of dislocation loops in Fig. 4.

XBB 722-993

Figure 1.

XBB 721-118

Figure 2.

XBB 721-21

Figure 3.

XBB 721-119

Figure 4.

XBL 7111-4703

Figure 5.

XBL 7111-4702

Figure 6.

XBL 7111-4701

Figure 7.

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