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THE INTERPRETATION OF HRTEM IMAGES OF PARTIALLY AMORPHIZED PYROCHLORE STRUCTURE TYPES.

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ABSTRACT

MASTER

The results of image simulations on partially amorphous microlite ($\text{Ca}_2\text{Ta}_2\text{O}_7$, pyrochlore structure) are presented. Results indicate that HRTEM images are not sensitive to the position of amorphous layers within a crystalline matrix. In addition, it is observed that the limit of detection of amorphous material within a crystalline matrix is dependent upon the total thickness of the sample. In thin crystals ($<150 \text{ \AA}$), up to 75 volume percent crystalline material can give rise to aperiodic images, yet the addition of a small amount of crystalline material (80 volume percent crystalline) produces a periodic image. Images calculated for isolated spheres of amorphous material distributed within crystalline microlite suggest that isolated domains of amorphization are observable at sample thicknesses less than three times the diameter of the feature. The image contrast of amorphized domains is enhanced by imaging at defocus settings significantly different than Scherzer focus. These results indicate that interpretation of HRTEM images of partially amorphized crystalline materials should be undertaken with caution, and estimates of the volume of damage considered only qualitative.

INTRODUCTION

The interpretation of high resolution transmission electron microscopy (HRTEM) images of structurally damaged materials has been based on the assumption that the mottled image contrast is caused by the loss of long range periodicity within the crystal and that the interface between periodic and aperiodic regions in the image corresponds spatially to the boundary between the crystalline and amorphous regions in the crystal. Neither assumption has previously been demonstrated [1]. The edge of the area of mottled image contrast may not correspond spatially to the interface between crystalline and amorphous regions due to interference between electron beams originating within the two regions, thus causing uncertainty in the interpretation of HRTEM images. This ambiguity has prevented unequivocal identification of isolated α -recoil tracks and complicates estimates of the volume of material in damaged regions.

In order to test the assumption that the mottled HRTEM image contrast is caused by a loss of long range periodicity, a series of simulated HRTEM images have been calculated for a series of partially amorphized crystal models. All calculations are done using the full multislice procedure [2,3], as implemented in SHRLI (Simulated High-Resolution Lattice Images) by Michael O'Keefe [4], and make use of the method of periodic continuation [5] to approximate the interaction of the electron beam with aperiodic features. All images presented here are calculated for the [110] zone of microlite ($\text{Ca}_2\text{Ta}_2\text{O}_7$; pyrochlore structure).

HRTEM images of the pyrochlore [110] zone were calculated for a series of $371.23 \text{ \AA}$ "crystals" which contained amorphous material, distributed in continuous layers, and which comprised 5, 10, 20, and 30 volume percent of the total crystal thickness. As expected, the HRTEM image becomes increasingly aperiodic in appearance with increasing amorphous volume. However, amorphous contents as low as 25 volume percent were found to give rise to aperiodic HRTEM images in thin crystals. In thicker sections, images of materials with as much as 60 volume percent amorphous content appear to be essentially crystalline.

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THE SENSITIVITY OF HRTEM IMAGES TO POSITION OF APERIODIC FEATURES

The sensitivity of HRTEM images to the placement of amorphized regions within a crystalline matrix has been investigated by calculating images of crystals in which a constant 37.123 Å thick amorphous layer is distributed at varying positions in the 100 slice multislice sequence (see Figure 1). Geometries include amorphous slices placed at: 1. the bottom surface of the crystal (corresponding to the damage expected in a back-thinned ion-milled sample); 2. the center of the crystalline matrix (corresponding to a buried α -recoil track or a buried amorphous layer caused by ion implantation); 3. the top surface of the crystal (a top-thinned ion-milled sample); 4. on both surfaces (as is the case of a normal ion-milled sample or a sample with oxidized surfaces). Regardless of the configuration, the exit images are indistinguishable, *i.e.* the character of HRTEM images is largely independent of the position of the amorphous slice within the stacking sequence.

IMAGING OF ISOLATED AMORPHOUS DOMAINS

Images of spherical amorphized domains were calculated in order to determine the limit of detection for aperiodic features within a crystalline matrix. The configurations presented here include: 1. a single sphere having a 15 Å radius and a maximum atom displacement of 2 Å; 2. two slightly overlapping spheres with 12 Å radii and a maximum atom displacement of 1 Å; 3. six randomly positioned 12 Å spheres located within a 50 Å zone in the near-surface region of the crystal (approximating a partially amorphized buried layer resulting from single-energy ion-implantation). Amorphization was accomplished by displacing all atoms within the specified radius of the center by a random displacement (limited in magnitude to the specified maximum, but not direction). This procedure results in a peripheral zone (with thickness equal to the specified maximum displacement) containing interstitials originating within the sphere of complete amorphization.

The resulting images (Figure 2), suggest that, with the exception of thin crystals (< 100 Å), single collision cascades (of 30 Å radius) are not observable in HRTEM images (for larger cascades, the maximum crystal thickness is likely to increase by an amount proportional to the increase in the amorphized domain size). Therefore, the aperiodic domains observed in HRTEM images of thick crystals represent overlapping or superimposed amorphous domains. If these interpretations are correct, the volume of damage due to ion-implantation or α -decay, as determined from the areal extent of aperiodic contrast in HRTEM images, are underestimated.

CONCLUSIONS

1. A minimum of 10 volume percent of amorphous material is required in order to be observed above background in HRTEM images. Amorphous contents as low as 25 volume percent may give rise to aperiodic images in thin crystals; and in thicker sections, as little as 40 volume percent crystalline material may produce periodic images (within the normal expected level of background noise).
2. Material which is as much as 25 volume percent crystalline may yield images that appear to be amorphous (See Figure 1; column 3, row 4 and column 4, row 2).
3. The HRTEM image is insensitive to the position of aperiodic regions within crystalline materials.
4. When observed in thin crystals (< 2 diameters), the spatial extent of a feature in an image corresponds well to the feature itself; however, in thicker sections there is no direct correspondence.
5. Improved aperiodic image contrast is obtained at defocus conditions significantly different from Scherzer focus.
6. Care must be taken when assigning significance to estimates of damage obtained from HRTEM images.

Figure 1. The effect of amorphous slice placement on image periodicity. Stippled areas represent amorphous slice positions. Images are output at 10 slice increments (1 slice = 3.71 Å). The similarity of image 10 in each series demonstrates the insensitivity of HRTEM images to the placement of the 10 slice amorphous layer within the crystalline sequence, and that the image resulting from a 10 percent amorphous material appears crystalline within the expected levels of background. Notice that image 4, column 3 and image 2, column 4 are calculated for materials composed of 75 volume percent crystalline material, yet the images are essentially aperiodic.

REFERENCES

1. G. Lumpkin and R.C. Ewing, *Phys Chem Minerals* 16, 2 (1988)
2. P. Goodman and A.F. Moodie, In: *Proc. Int. Conf. on Electron diffraction and the nature of defects in crystals*, Melbourne (Australian Acad. of Science, 1965).
3. P. Goodman and A.F. Moodie, *Acta Cryst. A* 30, 280 (1974).
4. M. A. O'Keefe, *SHRLI Users Guide*, (distributed with the SHRLI programs, 1980).
5. M.A. O'Keefe and S. Iijima, in: *Electron Microscopy Vol. I*, Ed. J.M. Sturgess (Microscopical Soc., Toronto, Canada, 1978).

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Figure 2. Calculated HRTEM images for isolated spherical amorphized domains. Configurations modeled include: a) a single sphere, (radius 15 Å, maximum displacement 2 Å, center located 22.3 Å below surface), b) two 12 Å spheres (1 Å maximum displacement) located at (0.4, 0.4, 0.3) and (0.6, 0.6, 0.5) relative to the tetragonal supercell ([100] = 5*[1-10], [010] = 7*[001], [001] = 5*[110]); and c) six 12 Å spheres (maximum displacement: 1 Å) located at (0.5 0.5 0.3), (0.25 0.25 0.35), (0.75 0.75 0.35), (0.2 0.5 0.5), (0.2 0.8 0.5), and