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MODEL FOR GRAIN GROWTH IN DOP-26 IRIDIUM CLAD. R. J. Lipinski and V. Tikare, Sandia National Laboratories*, Mail Stop 0747, PO Box 5800, Albuquerque, New Mexico 87185, rjlipin@sandia.gov. *(Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.)

Introduction: The fuel in a General Purpose Heat Source (GPHS) used in radioisotope power systems is contained within an iridium based DOP-26 alloy (with 0.3 wt% tungsten and doped with thorium) clad [1]. Past testing has shown that to ensure that the clad remains strong and resilient, it is important that the number of grains through the clad thickness be sufficient to avoid brittle behavior. The nominal grain size for a fresh fueled clad is about 25 µm. This allows about 26 grain diameters through the clad nominal thickness of 640 µm. The grains will grow at elevated temperatures and the growth rate will increase with temperature. This report describes the growth rate and suggests an improved model for grain growth.

Past Model and Data: Measurements of grain growth in DOP-26 Ir with time and temperature were made by McKamey, et al [2, 3, 4]. To correlate the data, Reference 2 used the theoretically derived equation for normal curvature-driven grain growth, which is as shown below if p is set equal to 2:

$$d = \left(d_0^p + k_0 t \exp(-Q/RT)\right)^{1/p} \qquad (1)$$

where

- d is the average grain size after time t (µm)
- d_0 is the initial grain size across the thickness (µm)
- t is time at the annealing temperature (hr)
- k_0 is a constant (μm^p/hr)
- Q is the activation energy (kJ/mol)
- R is the gas constant = 8.314 J/mol/K
- T is the anneal temperature (K)
- p is the grain growth exponent (dimensionless)

Curvature-driven grain growth (p=2) occurs in a pure material in which small grains with greater average internal curvature are consumed by larger grains growing outward so as to reduce the overall grain surface energy in the material [5].

The assumption of curvature-driven grain growth may not be the best option for DOP-26 Ir because of the pinning effect of the Ir_5Th particles. These particles anchor the grain boundaries and slow the overall grain growth rate. This effect can be simulated by using a different growth exponent (p) which can be empirically determined. For example, Chaim [6] shows how yttria-stabilized zirconia follows a growth rate dependent on the cube root of the sum of diameters to the third power (p=3) rather than diameter squared.

To explore the options of different growth behaviors, the data in Reference 2 were analyzed with different values of p. All the data for the D2 process with annealing in a vacuum, in 1.3 mPa oxygen, and in 13.3 mPa oxygen from Reference 3 were used (i.e., from Tables II, III, IV, V, VI, and X). The values used were the average grain sizes for DOP-26 alloys in the short traverse (ST) direction, which is perpendicular to the rolling direction, and is through the clad thickness. For the cases with oxygen present, the effect of larger grains on the surface of the clad was estimated to be negligible for this analysis. For some of the data the reference did not explicitly identify the initial average grain size, so the first reported size was identified as the starting point for subsequent analyses. The initial report time (t_0) was subtracted from the subsequent times to obtain the incremental time for grain growth.

Figure 1 shows a plot of grain size predicted by equation (1) with p = 2 as done in Reference 2 vs. the measured grain size. These data are compared to the model given by equation (1), plotted as a line of slope = 1.0 going through the origin. The parameters Q and k_0 were optimized to obtain the best fit. Time zero was assumed to be the time of the first measured grain diameter. For this assumption, a Q of 563 kJ/mol and k_0 equal to 2.78×10^{18} μm^2/hr give the best fit to the data for all temperatures.

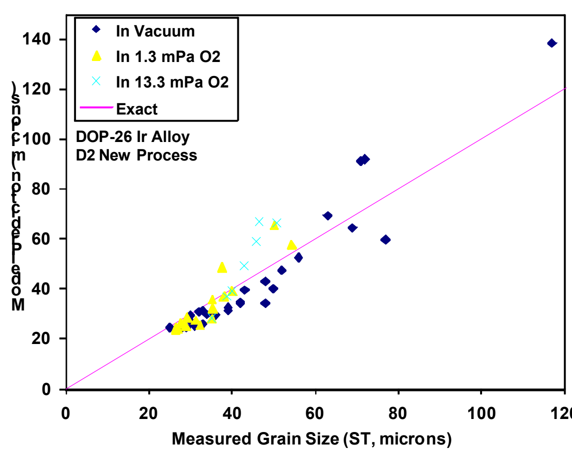

Figure 1. Model predicted grain diameter with p=2 vs. measured diameter

A detailed analysis of the statistical variation of the data is not available, but typically 100 to 600 grain-boundary crossings were included in each average (from multiple cross-sections). The plot shows that the measured grain sizes are not in good agreement with those predicted by the model. The measured grain size are consistently larger than predicted at small grain sizes and scattered at the larger sizes.

Improved Model: Recognizing that the Ir_5Th particles present in the iridium clad pin the grain boundaries and reduce their mobility, an improved model was developed. Figure 2 shows a plot, similar to that in Figure 1, in which the grain growth exponent in the equation (1) was optimized. The optimal fit was p=6 with Q = 734 kJ/mol and $k_0 = 6.13\times10^{30}$ μm^6/hr. The fit is better than with p=2. The growth coefficient (k_0) is of order 10^{30}, but the units are μm^6/hr, which is the reason for the large value. The goodness of fit parameter R^2 is 0.871 for Figure 1 and 0.936 for Figure 2. A value of p=6 may seem high compared with the previous model, but it does result in a better empirical fit. Furthermore, pinning particles are expected to severly retard grain growth resulting in a much higher grain growth exponent, p.

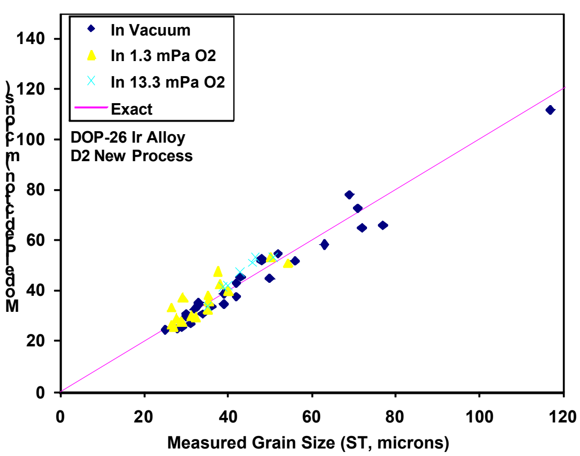

Figure 2. Model predicted grain diameter with optimized p vs. measured diameter.

Figure 3 shows the predicted and measured grain diameter vs. time for p=6. The grain diameter is normalized by the initial grain diameter so a variety of data can be plotted on the same graph. For reference, the initial diameter is in the range from 24 to 30 μm, which is not a large variation in initial conditions. Figure 3 shows good agreement between the measure grain sizes and the predicted sizes. This gives additional confidence in the grain growth model proposed in this work.

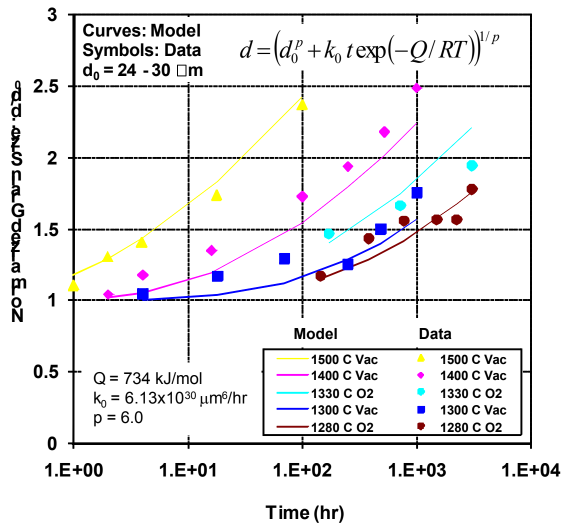

Figure 3. Model predicted grain diameter with p=6 vs. time.

If two or more heating events occur in succession, either Equation 1 or the figures must be used iteratively. The increment in grain size for each individual event starting from 25 μm cannot be simply added linearly. For example, for two heating events, the resulting grain diameter is:

$$d=\left(d_0^p+k_0t_1\exp\left(\frac{-Q}{RT_1}\right)+k_0t_2\exp\left(\frac{-Q}{RT_2}\right)\right)^{1/p} \quad (2)$$

where

T_1 is the temperature of the first heating event (K)

T_2 is the temperature of the second heating event (K)

t_1 is the duration at temperature T_1 (hr)

t_2 is the duration at temperature T_2 (hr)

The model proposed in this work will enable better prediction of grain growth under a variety of conditions.

References: [1] Skrabek, E., (2005) *Space Technology and Applications International Forum – STAIF 2005*, Albuquerque, NM, Feb. 13-17, 2005, CP246, pp 776-781. [2] McKamey, C. G., et al. (1996) *J. of Alloys and Comp.*, v244, pp 175-183. [3] McKamey, C. G., et al. (1998) *Grain Growth Behavior and High-Temperature High-Strain-Rate Tensile Ductility of Iridium Alloy DOP-26*, ORNL-6935, Oak Ridge National Laboratory, Oak Ridge, TN. [4] McKamey, C. G., et al, (2002) *Grain Growth Behavior, Tensile Impact Ductilitiy, and Weldability of Cerium-Doped Iridium Alloys*, ORNL/TM-2002/114, Oak Ridge National Laboratory, Oak Ridge, TN. [5] Humphreys, F. J., et al (1995), *Recrystallization and Related Annealing Phenom*ena, Elsevier Ltd, Kidlington, Oxford, UK. [6] Chaim, R., (2002) *Materials Sci. and Eng. A,* v. 486, pp 439-446.