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STEADY-STATE GRAIN GROWTH IN UO_2

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Introduction

According to Rhines and Craig (1), steady-state grain growth is characterized by an increase of the grain size without variation ***in*** the grain form. It is also observed that the form of the grain size distribution remains constant, whereas it is just displaced to larger sizes. One of us (2) has proposed a model for grain growth based on the tendency for minimization of the interfacial energy of the grain aggregate (which is stored in grain boundaries, triple lines, and quadruple points), and the tendency for uniformity in its spatial distribution. ***According*** **to this model, a combination of two polyhedra (the dodecahedron and the γ-tetrakaidecahedron, which is a polyhedron with 12 faces of 5 sides and 2 faces of 6 sides) is taken as the ideal form for grains, such that the number of quadruple points, triple lines, and grain boundaries are minimized. The steady-state size distribution is bimodal and can be described by**

$$\Delta V_i/V = n_o^g \cdot d_i^3 \cdot exp[\lambda_g \cdot (d_o - d_i) \cdot d_i^2] + n_o^p \cdot d_i^3 \cdot exp[\lambda_p \cdot (d_o/1.15 - d_i) \cdot d_i^2] \tag{1}$$

where g and p designate the components of the distribution **corresponding, respectively, to the γ-tetrakaidecahedra and dodecahedra.** $\Delta V_i/V$ is the ***volume*** fraction of size d_i, and n_o^g, n_o^p, λ_g, λ_p, and d_o are parameters. ***n_o*** **is related to the number of grains and λ to the dispersion of each component of the distribution.** In steady-state grain growth the p-component achieves 18% of **volume** fraction.

The topological path in steady-state grain growth is taken in this model as proposed by Rhines and Craig (1). Consequently, the grain growth law proposed by these authors should also be observed

$$1/N_v = (1 + a.t)N_v^o \tag{2}$$

where N_v is the specific number of grains (number of grains per unit volume), a is a constant related ***to*** the grain aggregate, and t is the time. N_v^o is the specific number of grains at $t = 0$.

To measure N_v avoiding the difficult technique of serial sectioning, one should make an assumption ***with regard to*** the grain form. In the ***derivation*** of equation (1) it was assumed that the grain sizes can be described by d_i, which was taken as the three dimensional grain diameter (given a grain of volume v_i, $d_i = (6v_i/\pi)^{1/3}$). Both d_i and N_v can be estimated by the Saltykov's method (3).

TABLE 1
Porosity Variation and Grain Size of Sintered and Re-sintered Samples

Re-sintering time (hours)	Porosity (% of volume	Mean intercept (μm)	***Total*** number of grains (10^6/mm^3)
0	6.43	2.7	50.309
0	6.64	3.1	36.784
1	4.81	4.2	15.110
1	4.79	4.2	15.000
1	4.72	3.9	18.509
2	4.35	3.6	23.112
2	4.37	3.8	21.498
2	4.38	3.9	20.527
3	3.94	5.6	6.395
3	4.38	5.7	6.155
5	3.67	5.8	5.898
5	3.58	6.7	3.907
5	3.60	6.0	5.906
8	3.48	6.7	3.622
8	3.65	6.7	3.945
8	3.47	7.0	2.948

Experimental Procedure

Nuclear grade UO_2 powder was pressed in the form of cylindrical pellets of 10 mm diameter and 10 mm height at 400 MPa. The pellets were sintered at 1973 K/2h/H_2. Some sintered pellets were re-sintered at 2003 K under H_2 ***for*** different times: 1 h, 2 h, 3 h, 5 h, and 8 h. To bring out the grain structure, the pellets were cut, polished, and thermal treated at 1523 K under CO_2 of technical purity ***for*** 1 h.

The grain size distributions and the mean intercept length were measured with the image analyzer Quantikov (4), which employs optical microscopy and the Saltykov's method. 340 to 1,940 grain sections per sample were taken. Very small grains should always exist because of the grain growth process. The experimental technique we have employed cannot reveal these grains. So, the measured ***total*** number of grains should be smaller than the real one. Moreover, the consideration of grains as spheres made by the Saltykov's method increases the dispersion of the measured size distribution in relation to the real one. As the grains of the samples are equiaxed, we expect this error should be small.

The calculated mean intercept length based on the size distribution is in agreement with the measured one. This indicates that the grains have a small deviation from the spherical geometry.

The porosity of the sintered and re-sintered pellets ***was*** measured through the Archimedes' method in xylol.

Results

Table 1 shows the results of the measurement of the porosity, the mean intercept length, and the specific number of grains.

Figure 1 shows some grain size distributions. ***These are*** typical of the steady-state grain growth, where the size distribution at different instants can be overlapped by a scale change. ***The data also show*** that the pellets have achieved a steady-state grain growth during the sintering.

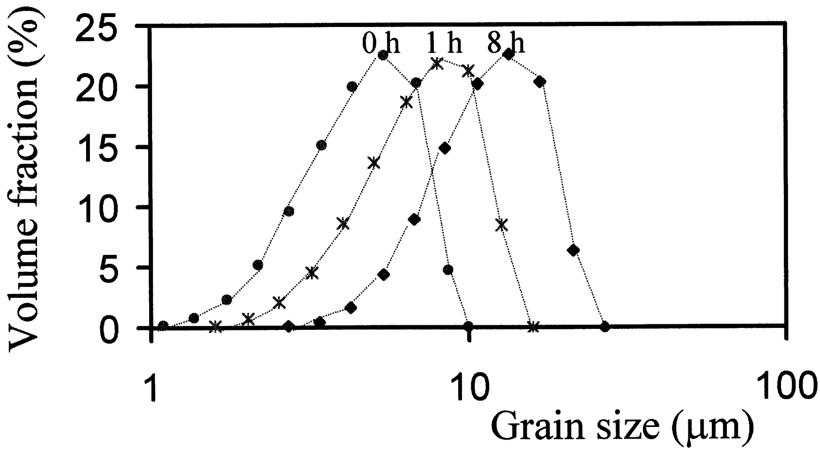

Figure 1. Grain size distribution at different re-sintering times.

Figure 2 shows the ***best*** fit of equation (1) to the 8 h re-sintering case. Similar fits were ***also obtained*** for ***the other*** samples. As expected by the model, the volume fraction for the *p*-component remained at 18%, as shown in Figure 3.

Figure 4 shows that equation (2) can describe the evolution of the specific number of grains, as proposed by Rhines and Craig.

As shown in Table 1, the pellets had a residual porosity that decreased during the ***sintering.*** **This porosity can influence the steady-state grain growth as expected by the model in two ways: by creating additional interface elements that are not considered in the model (e.g. pore-grain interface), and by space filling which is assumed in the derivation of equation (1). However, the results suggest that the steady-state grain growth was not disturbed by this porosity.**

One interesting feature of equation (1) is that the number of grains does not vanish as $d_i \rightarrow 0$, as in

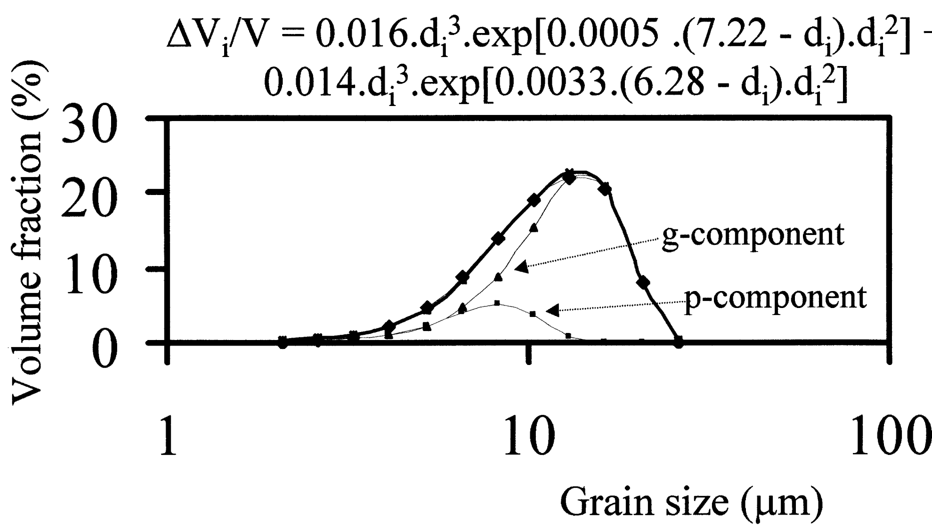

Figure 2. **Best fit** of equation (1) to the data of the 8h re-sintering case.

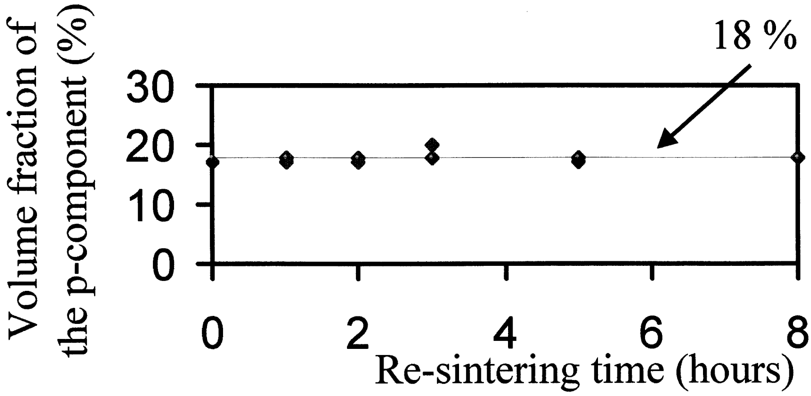

Figure 3. **Volume** fraction of the p-component according to the fit of equation (1) to experimental data.

the case of the lognormal distribution and theoretical distributions based on the LWS theory. As already pointed out, very small grains should always exist because of the growth process. The ***number density*** of these grains can be estimated through **equation (1)** ***by fitting it*** to experimental data. It is the sum of n_o^g and n_o^p. Figure 5 shows how $n_o^g + n_o^p$ decreases with the evolution of the steady-state grain growth.

The mean intercept length, λ, is usually employed as ***a measure of*** the grain size. It was observed that the following equation can describe its evolution

$$\lambda^{2.5} - \lambda_o^{2.5} = k.t \tag{3}$$

where $\lambda_o = 3.070$ μm is the initial mean intercept length and $k = 13.84\ \mu m^{2.5}/h$. **Since the mean intercept length is related to the surface to volume ratio of the grain aggregate, equation (3) should not be compared to equation (2).**

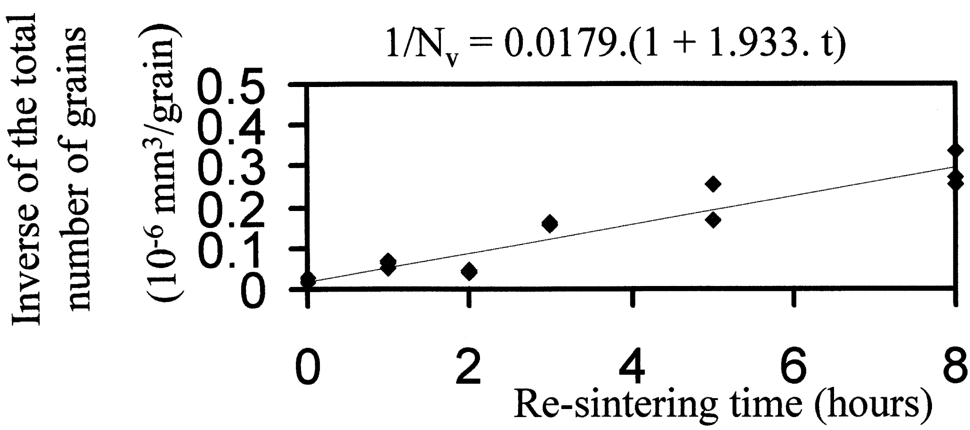

Figure 4. **Best fit** of equation (2) to the experimental **data.**

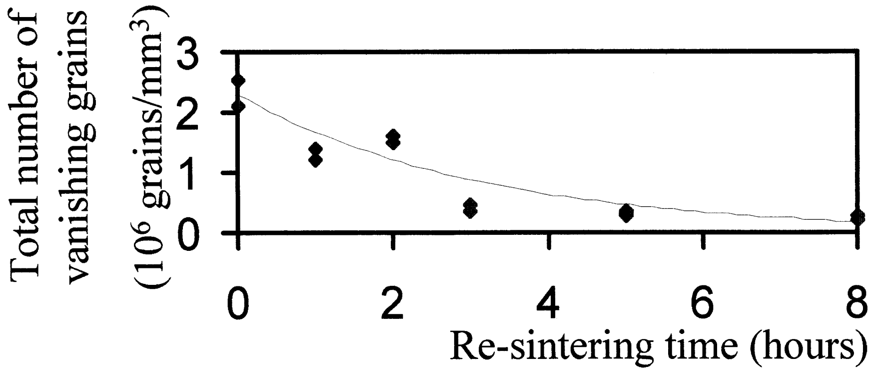

Figure 5. **Change in the total number of vanishing grains, as predicted from equation (1).**

Summary

We have observed steady-state grain growth in sintered UO_2 pellets of nuclear purity at 2003 K under H_2. The behavior of the grain size distribution at different ***instants is consistent with the grain growth model*** proposed by one of us. The ***total*** number of grains was estimated **using** the Saltykov's method, and ***the evolution is in accordance*** with the model proposed by Rhines and Craig. The ***parabolic*** growth law ***was*** observed for the mean intercept length with $n = 0.4$.

Acknowledgments

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