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MECHANISM OF DIMENSIONAL INSTABILITY

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ABSTRACT

An explanation of the dimensional instability of alpha-uranium under irradiation is advanced, based upon the anisotropic diffusion of lattice imperfections to grain boundaries and free surfaces. An approximate solution of the diffusion equations for an ellipsoidal grain is obtained, which predicts growth rate as a function of pile flux, temperature, and grain size. The calculations are applied to polycrystalline material and some comparisons made between theory and experiment which confirm the feasibility of the suggested mechanisms of dimensional instability.

MECHANISM OF DIMENSIONAL INSTABILITY

By

L. L. Seigle

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I. Introduction

The dimensional instability of alpha-uranium under irradiation has generally been attributed to the action of internal stresses arising from the passage of fission fragments through the lattice (1,2). These stresses are caused by the thermal expansion of small heated regions in the wake of the fragments, and are imagined to produce an irreversible plastic deformation of the metal with extension in some preferred direction.

In addition to generating heat, however, fission fragments are also considered to produce large numbers of lattice defects, particularly interstitial atoms and lattice vacancies.* It is usually assumed that most of these defects recombine within the grains, but in the light of recent investigations, which are discussed in the following section, it seems likely that some return to the lattice by other processes, for example, diffusion to grain boundaries. If the diffusion of lattice imperfections is anisotropic this process could bring about changes in grain dimensions which might be large enough to account for the observed growth of uranium. The present work has been written in order to examine the feasibility of such a mechanism of dimensional instability.

II. Dimensional Changes Produced by the Diffusion of Lattice Imperfections

The idea that shape changes can be associated with the diffusion of lattice imperfections between free surfaces was advanced by Nabarro in 1948⁽⁴⁾. He considered changes in dimensions of a metal crystal as a result of the diffusion of vacancies from one surface to another under the action of external stresses. An unexpected experimental confirmation of Nabarro's ideas was obtained by Alexander^(5,6) while investigating the sintering of copper wire compacts. Alexander observed that once grain boundaries had been eliminated by grain growth, sintering of the copper wire bundles practically stopped. He also noted that voids were preferentially eliminated in the vicinity of grain boundaries. Herring pointed out^(7,8) that these observations could be explained on the basis of Nabarro's ideas, if it were assumed that lattice vacancies migrated from the voids to the nearby grain boundaries, under the action of internal surface stresses, and were there eliminated by a small movement of the grains together. In other words atoms were subtracted from the lattice at the grain boundary and migrated via a vacancy (or interstitial) mechanism to the void. Such a diffusional pro-

*It has been calculated by Seitz (3) that, after 10% burnup of the U^{235} in a natural uranium sample, each atom has been displaced about 17 times.

cess was also conceived by Herring to account for creep in polycrystalline metals at high temperatures, and experiments by Buttner, et al⁽⁹⁾, confirmed this point of view.

Finally direct observation of dimensional changes occurring upon the absorption of lattice defects at grain boundaries was made by Balluffi, et al, while studying porosity formation in dezincified alpha brass⁽¹⁰⁾. Porosity did not form upon dezincification of fine grained sheets, apparently since the lattice vacancies generated by the outflow of zinc were preferentially absorbed at the grain boundaries. In agreement with this hypothesis it was observed that the relative lateral shrinkage of fine-grained sheets was greater than that of coarse-grained sheets in which porosity did form. These experiments clearly proved that the absorption of vacancies at grain boundaries was accompanied by a shrinkage of the grains normal to the boundary.

III. Theory of Dimensional Instability

In the light of the above evidence it seems inescapable that some of the lattice imperfections generated in alpha-uranium during pile exposure must also diffuse to and be eliminated at grain boundaries and free surfaces. Interstitial atoms will add to the lattice, producing an extension, and vacancies subtract from the lattice, producing a contraction of the grain at its boundaries. If a sufficient number of defects migrate to the grain boundaries, and if the fluxes are unbalanced, this process would cause macroscopic changes in shape of the grains. In order to estimate the rate of growth due to the operation of such a process, it is necessary to calculate the rate and anisotropy of diffusion of the imperfections.

A) Rate of Diffusion of Imperfections to Grain Boundaries

The rate of flow of imperfections to the grain boundaries is proportional to the diffusion coefficient of the imperfection and the concentration gradient at the grain boundary. The concentration gradient is obtained from the steady state distribution of defects and this can be calculated by solution of the appropriate differential equations. Following Seitz⁽³⁾, we assume that atoms are displaced from lattice sites at a rate αn_0 where n_0 is the density of atoms in uranium and α is determined by the pile flux. It is also supposed that displaced atoms will find their way back onto the lattice by only two processes: (1) recombination with vacancies, and (2) diffusion to grain boundaries or free surfaces. The rate of recombination is calculated by noting that since there are

twice as many interstitial sites as atoms in α -uranium, (Appendix I), the probability that any one interstitial site is occupied is $\frac{n_i}{2n_0}$ where n_i equals the density of interstitial atoms. The probability that any one of the five surrounding lattice points is vacant equals $\frac{5n_v}{n_0}$ where n_v is the density of vacancies, and the probability factor for recombination at a given interstitial site is $5/2 \frac{n_i n_v}{n_0^2}$. If the jump frequencies of interstitial atoms and vacancies are ν_i and ν_v then the rate of recombination per unit volume of material will be

$$\left(\frac{5}{2} \frac{n_i n_v}{n_0^2} \right) (\nu_i + \nu_v) (2n_0) = \frac{5n_i n_v \nu}{n_0}$$

in which $\nu = \nu_i + \nu_v$. Taking diffusion into account, the rates of accumulation of vacancies and interstitial atoms are given as

$$\frac{\partial n_i}{\partial t} = D_{ix} \frac{\partial^2 n_i}{\partial x^2} + D_{iy} \frac{\partial^2 n_i}{\partial y^2} + D_{iz} \frac{\partial^2 n_i}{\partial z^2} + \alpha n_0 - \frac{5n_i n_v \nu}{n_0} \quad (1)$$

$$\frac{\partial n_v}{\partial t} = D_{vx} \frac{\partial^2 n_v}{\partial x^2} + \dots \dots \dots + \alpha n_0 - \frac{5n_i n_v \nu}{n_0} \quad (2)$$

and under steady state conditions

$$\frac{\partial n_i}{\partial t} = \frac{\partial n_v}{\partial t} = 0 \quad (3)$$

The coefficients $D_{ix}, \dots, D_{vx}, \dots$, are the diffusion coefficients of interstitial atoms and vacancies in the principal crystallographic directions, which are orthogonal in the orthorhombic lattice of α -uranium.

It appears that the above system of equations could be solved to obtain the steady state distribution of interstitial atoms and vacancies for given boundary conditions by successive approximations. The mathematics becomes very involved, however, and no serious attempt has yet been made to carry the solution beyond the first approximation.

As a first approximation it is assumed that the vacancy concentration is constant throughout the volume of a grain. The concentration of interstitial atoms varies and is maintained at the value n_e at the grain boundary, where n_e is the con-

centration of interstitial atoms in thermal equilibrium with the lattice. By making the changes of variables $\xi = x/\sqrt{D_{ix}}$, $\eta = y/\sqrt{D_{iy}}$ and $\zeta = z/\sqrt{D_{iz}}$, equation (1) becomes

$$\frac{\partial n_i}{\partial t} = \frac{\partial^2 n_i}{\partial \xi^2} + \frac{\partial^2 n_i}{\partial \eta^2} + \frac{\partial^2 n_i}{\partial \zeta^2} - 5\left(\frac{n_v \nu}{n_0}\right)n_i + \alpha n_0 = 0 \quad (4)$$

Transforming to spherical polar coordinates

$$\frac{1}{\rho^2} \frac{\partial}{\partial \rho} \left(\rho^2 \frac{\partial n_i}{\partial \rho} \right) + \frac{1}{\rho^2 \sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial n_i}{\partial \theta} \right) + \frac{1}{\rho^2 \sin^2 \theta} \frac{\partial^2 n_i}{\partial \phi^2} - 5\left(\frac{n_v \nu}{n_0}\right)n_i + \alpha n_0 = 0 \quad (5)$$

With the boundary condition $n_i = n_e$ when $\rho = \rho_0$, the solution to (5) is

$$n_i = \left[\frac{\rho_0 \left(n_e - \frac{\alpha n_0^2}{5n_v \nu} \right)}{\sinh(\rho_0 k)} \right] \frac{\sinh(\rho k)}{\rho} + \frac{\alpha n_0^2}{5n_v \nu} \quad (6)$$

In this equation

$$k = \left(\frac{5n_v \nu}{n_0} \right)^{1/2}$$

Equation (6) is the solution for an ellipsoidal grain whose axes are in the ratio $\sqrt{D_{ix}} : \sqrt{D_{iy}} : \sqrt{D_{iz}}$. The gradient of interstitial atoms along the x axis can be estimated by differentiation,

$$\begin{aligned} \left(\frac{\partial n_i}{\partial x} \right)_{y=z=0} &= \frac{dn_i}{d\rho} \left(\frac{\partial \rho}{\partial x} \right)_{y=z=0} = \frac{1}{\sqrt{D_{ix}}} \frac{dn_i}{d\rho} \\ &= \frac{\rho_0 \left(n_e - \frac{\alpha n_0^2}{5n_v \nu} \right)}{\sinh(K \rho_0)} \cdot \frac{K \rho \cosh(K \rho) - \sinh(K \rho)}{\rho^2} \quad (7) \end{aligned}$$

where $K = \left(\frac{5n_v \nu}{n_0 D_{ix}} \right)^{1/2}$

The rate of diffusion of interstitial atoms into the grain boundary at the point where it is pierced by the x axis, is therefore

$$\left(\frac{\partial n_i}{\partial t} \right)_{x_0} = -D_{ix} \left(\frac{\partial n_i}{\partial x} \right)_{x=x_0, y=z=0} = K D_{ix} \left[\frac{\alpha n_0^2}{5n_v \nu} - n_e \right] \left[\coth(K \rho_0) - \frac{1}{K \rho_0} \right] \quad (8)$$

and the rate of extension of the grain in the x direction at this point

$$\frac{\partial l_x}{\partial t} = \frac{1}{n_0} \left(\frac{\partial n_i}{\partial t} \right)_{x_0} = K D_{ix} \left[\frac{\alpha n_0}{5 n_v \nu} - \frac{n_e}{n_0} \right] \left[\tanh(K x_0) - \frac{1}{K x_0} \right] \quad (9)$$

The constant value of n_v for these calculations may be taken as the steady state value under irradiation assuming recombination but no diffusion to grain boundaries. This is obtained by equating the rate of generation of vacancies with the rate of recombination with interstitial atoms,

$$\alpha n_0 = \frac{5 n_i n_v \nu}{n_0} \quad (10)$$

$$\frac{n_v}{n_0} = \left(\frac{\alpha}{5 \nu} \right)^{1/2}$$

Also, at the relatively low temperatures under consideration, concentration of interstitial atoms in thermal equilibrium with the lattice is probably very small so that $\frac{n_e}{n_0} \ll \frac{\alpha n_0}{5 n_v \nu}$.

Therefore

$$\frac{\partial l_x}{\partial t} \approx \left(\frac{\alpha^3 D_{ix}^2}{5 \nu} \right)^{1/4} \left[\tanh(K x_0) - \frac{1}{K x_0} \right] \quad (11)$$

$$\text{where } K = \left(\frac{5 n_v \nu}{n_0 D_{ix}} \right)^{1/2} \equiv \left(\frac{5 \alpha \nu}{D_{ix}^2} \right)^{1/4}$$

In order to obtain a numerical estimate of $\frac{\partial l_x}{\partial t}$, Seitz's figure of $\alpha = 9.0 \times 10^{-7} \text{ sec}^{-1}$ for fission in 200 days of 10% of the U_{235} in natural uranium is taken⁽³⁾. Assuming that $\nu_i \gg \nu_v$, i.e., that interstitial atoms diffuse much more rapidly than vacancies, the jump frequency may be approximated by

$$\nu \approx \nu_i \sim 10^{13} e^{-\frac{Q_i}{RT}} \quad (12)$$

and the diffusion coefficient

$$D_i = \nu_i d^2 \sim 10^{-7} e^{-\frac{Q_i}{RT}} \quad (13)$$

where 10^{13} is the characteristic oscillation frequency of atoms in the lattice and d is the jump distance for interstitials, of the order of 10^{-7} cm .

In equations (12) and (13) Q_i is the activation energy for migration of interstitial atoms in alpha-uranium. The only information which could be found in the literature regarding the activation energy for the jump from one interstitial position to another in a pure metal lattice was the calculation of Huntington for copper. He estimated that the saddle point energy for the particular jump process in which an interstitial copper atom replaces a normally situated atom on its lattice position, and the replaced atom is pushed into a second interstitial site, is about 6,000 cal/mol (12). In the absence of any better information and since the melting point of uranium is close to that for copper it was decided to use this value for Q_i , which, since it is calculated for a close-packed lattice, may be on the high side for alpha-uranium. With this value for the activation energy,

$$D_i = 0.1e^{-\frac{6,000}{RT}} = 2 \times 10^{-4} \text{ cm}^2 \text{ sec}^{-1} \text{ at } 200^\circ\text{C} \quad (14)$$

Substituting in (11) one obtains for the rate of growth in the x direction at 200°C ,

$$\left(\frac{dx}{dt}\right) = 7.4 \times 10^{-10} \left\{ \text{ctnh} (1.2 \times 10^3 x_0) - \frac{1}{1.2 \times 10^3 x_0} \right\} \quad (15)$$

For values of the argument greater than 10, the function $(\text{ctnh} - \frac{1}{x})$ is approximately unity, which means that $\frac{dx}{dt} = 10^{-9}$ cm per

sec independent of grain size for all grain sizes in excess of about 100μ . An idea of what this number means in practical terms may be obtained by noting that the percentage increase in length in the x direction which could occur in 200 days is equal to $\frac{(3600)(24)(200)(10^{-9})}{100 \times 10^{-4}} \times 100 = 178\%$.

B. Anisotropy of Diffusion

The calculations given above suggest that the diffusional flow of lattice imperfections could easily be large enough to account for observed shape changes in alpha uranium, but in order for the mechanism to operate it is necessary that the flow of interstitial atoms and vacancies into the grain boundaries do not cancel each other. It has already been pointed out by Huntington (13) that the anisotropy of diffusion in an anisotropic crystal can be different for interstitial atoms and vacancies. As shown below, application of the "random walk" scheme to migration of imperfections yields a result of this nature for alpha-uranium.

The diffusion coefficient in the direction x is proportional to

$$D_x = \sum_j \Gamma_j (\delta l_j \cos \alpha_{jx})^2 \quad (16)$$

where Γ_j is the frequency of jump j , δl_j is the length of jump j , and α_{jx} is the angle between the jump direction and the x axis. There are two groups of interatomic distances in the alpha-uranium lattice, approximately 2.8 Å and 3.3 Å (18). Atoms within the corrugated sheets parallel to the (010) plane are separated by distances of 2.762 and 2.852 Å, and atoms between adjacent sheets by 3.261 and 3.322 Å. If it is assumed that vacancy-atom exchange takes place only between nearest and next-nearest neighbors, i.e., the frequency factor for the larger jumps is negligible, then the conclusion is immediately apparent that the diffusion coefficient for vacancies perpendicular to the corrugated sheets, i.e., in the $[010]$ direction, is zero. If jumps between nearest and next-nearest neighbors are considered to be equally likely then diffusion coefficients in the $[100]$, $[010]$, and $[001]$ directions will be in the ratio 1:0: .8.

In order to apply this scheme to interstitial diffusion it is necessary first to locate the interstitial positions in the alpha-uranium lattice. The calculations (Appendix I) indicate that there are two types of interstitial sites, one which accommodates a sphere 1.10 Å in diameter, and the other a sphere 1.36 Å in diameter. Confining attention to the larger interstitial site, there are eight of these per unit cell located at

$$\begin{array}{ll} (1/2, y, 1/2+z) & (0, 1/2+y, 1/2+z) \\ (1/2, y, -z) & (0, 1/2+y, -z) \\ (1/2, -y, 1/2-z) & (0, 1/2-y, 1/2-z) \\ (1/2, -y, z) & (0, 1/2-y, z) \end{array}$$

where $y = 0.085$, and $z = 0.050$. Each of these is at the center of a very slightly skew pyramid, whose base is a square of four atoms lying in one corrugated sheet and apex one atom in the next sheet. Since each of the large interstitial sites is surrounded by 5 atoms, and there are 8 interstitial sites and 4 atoms per unit cell, it is apparent that each atom must have 10 adjacent interstitial sites. Taking Huntington's picture of interstitial self-diffusion, if one of the interstitial sites is occupied, there are 9 ways in which a given adjacent atom can be pushed from its normal lattice position into one of its adjacent interstitial sites. With each interstitial site surrounded by 5 atoms there are hence 45 ways in

which a single interstitial atom can make a "jump". In addition, the pyramid of atoms surrounding each interstitial site may have one of four possible orientations; therefore 180 jumps can be distinguished. Symmetry considerations reduce the total number of jumps which must be considered to 33. Assuming that each possible jump is equally likely, application of equation (16) indicates that the diffusion coefficients for interstitial atoms in the a, b, and c directions will be in the ratios 0.9:0.9:1, i.e., the diffusion of interstitials is nearly isotropic, as might be expected from the large variety of possible jumps.

The above considerations indicate that the flow of vacancies and interstitial atoms to the grain boundaries are probably not equal in all directions. In particular, it appears that only interstitial atoms are likely to migrate in the $[010]$ direction and therefore a grain should tend to increase its length in this direction. The $[100]$ direction is slightly favored for vacancy migration, and therefore shrinkage. As shown by the experiments of Turkalo⁽¹⁴⁾, single crystals of alpha-uranium do apparently grow in the $[010]$ direction and shrink in the $[100]$ direction under irradiation. No dimensional changes in the $[001]$ direction were observed.

V. Discussion

The preceding calculations are for a grain free from subboundaries, dislocations or mechanical constraints. They predict that single crystals will grow in the $[010]$ direction at a constant rate which is a function of pile flux and temperature, but independent of size above a certain small size of the order of 100 μ . According to equation (11) the rate of growth will vary directly with the 3/4th power of the pile flux and the fourth root of the jump frequency for interstitial atoms.

The variation of growth rate with temperature assuming a value of 6,000 cal. per mol for the activation energy for the interstitial jump is shown in Fig. 1. Growth essentially stops at 100°K. This prediction has been recently confirmed by KAPL (16). On the other hand Fig. 1 indicates that the growth rate should approximately double upon increasing temperature from 200° to 500°C, and it has been reported that the growth rate of irradiated alpha-uranium decreases at temperatures near 500°C (17). It would be important to know if any structural changes occurred in the specimens irradiated at 500°C. The growth rate of a polycrystalline material should be a function of grain size, and possible grain growth during irradiation at elevated temperatures would tend to lower growth rates.

Although the individual crystallites may change shape, a polycrystalline mass will suffer no dimensional instability if the grains are randomly oriented. Under these circumstances the shape changes of the individual grains cancel each other out. If a preferred orientation exists, the components of growth will have a non-zero resultant in this direction, and the aggregate will change shape. Neglecting the constraints imposed by one growing grain upon another, the rate of fractional length increase of the polycrystal in the preferred direction, $\frac{dL_x}{dt}$, will be

$$\frac{dL_x}{dt} = \frac{2\gamma}{d} \cdot \frac{dl_x}{dt} \quad (17)$$

where $\frac{dl_x}{dt}$ is the growth rate of a single grain given by (11), d is the average grain diameter, and γ is a coefficient expressing the degree of preferred orientation. Since $\frac{dl_x}{dt}$ is approximately constant, for fairly coarse grained material the predicted rate of growth is inversely proportional to the grain size. This is a consequence of the fact that the smaller the grain size, the larger the grain boundary area in a given polycrystalline specimen to which imperfections can diffuse. The inverse relationship between grain size and growth rate is borne out by the experimental data presented by Turkalo⁽¹⁴⁾.

In reality the grains of a polycrystalline sample cannot grow freely and independently since, if the matrix is to remain continuous, the grain surfaces must remain in contact. Unless the growth directions are perfectly aligned, therefore, internal stresses will be built up by the continuing diffusion of imperfections which will either stop the flow to the grain boundaries or cause plastic deformation or fracture. The maximum stress which can be built up by the diffusional flow is the osmotic pressure, P , resulting from the difference in concentration of imperfections at the grain boundary, n_e , and at the grain center, n . Since the concentrations of imperfections are low,

$$P = \frac{RT}{V_0} \ln\left(\frac{n}{n_e}\right) \quad (18)$$

where V_0 is the specific volume of alpha-uranium. Using the value for n calculated from equation (10), with α and γ as previously given, and assuming a work of formation of interstitial atoms of 100,000 cal. per mol., P at 200°C is of the order of 10^7 psi. This is of the same order as the theoretical maximum strength of the perfect crystal and it therefore appears that stresses high enough to deform and even disrupt the grains are easily developed. On the other hand since the yield stress of the pure metal is a few orders of magnitude below the osmotic pressure at 200°C it does not appear that internal stresses will appreciably slow down the diffusion of interstitial atoms to the grain boundaries.

A factor which was neglected in the calculations is the existence of sinks for imperfections within the grains such as random dislocations and subgrain boundaries. The absorption of vacancies and interstitial atoms by such internal sinks might also cause shrinkage or growth of the lattice in preferred directions. Such a possibility takes on added importance when the occurrence of plastic deformation insures the presence of large numbers of dislocations in the matrix. With high supersaturation of interstitials these might also precipitate as stacking faults between the corrugated (010) sheets, as has been suggested for other materials (15).

V. Conclusions

On the basis of the foregoing calculations it appears possible that significant changes in the shape and structure of alpha-uranium samples under irradiation are produced by the anisotropic diffusion of lattice imperfections to grain boundaries and free surfaces. Although observed changes in alpha-uranium could be accounted for by this mechanism alone, it seems reasonable to suppose that stress relaxation and diffusional processes act together, one being the counterpart of the other. Experiments to determine the effect of temperature and grain size upon the growth rate of irradiated alpha-uranium should help to decide which is the dominant process.

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APPENDIX I

THE LOCATION AND SIZE OF INTERSTITIAL HOLES IN THE ALPHA-URANIUM LATTICE

The purpose of this appendix is to indicate the locations and sizes of various interstitial holes in the orthorhombic uranium lattice, based on a hard sphere model. First plane configurations of three and four atoms will be examined, and then the true interstitials surrounded by four atoms in space and by five atoms in space will be obtained. The data and notation contained in a report by Tucker⁽¹⁸⁾ will be used.

Plane Configurations

Figure 2 presents the orthorhombic uranium lattice made of spheres touching at the closest interatomic distance. The three light colored balls in the a b plane are illustrative of the first type of configuration to be examined. If one edge of the unit cell is taken as the origin, the three atoms can be represented by the vectors $[0i + 0j + 0k]$, $[ai + 0j + 0k]$ and $[a/2i + b/2j + 0k]$ where i , j , k are respectively parallel to a , b , c . This notation will in the future be shortened to $[0,0,0]$, $[a,0,0]$, $[a/2,b/2,0]$. A vector R $[r,s,t]$ describes the position of the hole surrounded by the atoms. Since R must be equidistant from the three atoms, $|R|^2 = r^2 + s^2 + t^2 = (r-a)^2 + s^2 + t^2 = (r-a/2)^2 + (s-b/2)^2 + t^2$. The solution of these equations is $R = [a/2, b/4 - a^2/4b, 0]$, or, evaluated, $[1.43, 1.12, 0]$. The distance from $[0,0,0]$ to $[1.43, 1.12, 0]$ is $1.82\text{\AA}$; thus the diameter of the hole, using $1.38\text{\AA}$ as the atomic radius of uranium, is $0.88\text{\AA}$. The data for this and other plane configurations similarly calculated are listed in Table I. For these plane configurations, only one location is calculated and no attempt will be made to place them in the unit cell.

Interstitial Surrounded by Four Atoms in Space

Figure 2 also illustrates the interstitial amid four atoms in space as the dark marble surrounded by the four light-colored balls. The results of a sample calculation are given in Table I. For counting purposes, this interstitial will be considered as lying between a base of three atoms in the a b plane and the near neighbor apex atom above (or below). There will be two tetrahedral configurations for each atom (one pointing in the general direction of $+C$ and the other, $-C$). Since each apex atom has two interstitials associated with it, and each interstitial is shared by four atoms, there will be eight interstitials surrounding a single atom. Table III lists the various interstitials around one atom, which is chosen for convenience as $[0,0,0]$. These interstitials

may be derived from the sample calculation (Table I) by translating the apex atom of the calculation through the lattice. However, when this is done, whenever a jump of $c/2$ is made the sign of the component when the apex is at $[0, 0, 0]$ must be reversed. This is the equivalent of saying that there are four differently oriented tetrahedral configurations, as can be seen by inspection of Figure 2. Finally, since there are two interstitials per atom and four atoms per unit cell, there are eight four-atom interstitials in the unit cell. Table II locates these interstitials in the unit cell given by Tucker(18).

Interstitial Surrounded by Five Atoms in Space

Figure 3 illustrates the five-atom interstitial as the dark marble amid the five light-colored balls. These atoms form a very slightly skewed pyramid, as is shown by the following calculation. Consider the four atoms lying in the corrugated plane; their center of gravity is $[a/2, -by, c/4]$ from Table I. Since the apex atom of the configuration will be at $[a/2, b/2-2by, c/2]$ the vector V joining the apex and the center of gravity of the base will be $V = [0, b/2-by, c/4]$. On the other hand, the normal to the corrugated plane is given by the cross-product of two vectors in the plane such as $[a, 0, 0]$ and $[0, -2by, c/2]$: $N = [0, -2by, c/a]$ or $N = [0, -2by, c/a]$. If the pyramid is regular, N and V will be parallel, or $N \times V = 0$. $N \times V = [ac^2/8 - ab^2y + 2ab^2y^2, 0, 0]$. For $N \times V = 0$, $ac^2/8 - ab^2y + 2ab^2y^2 = 0$ or $y = 0.116$. However, for uranium $y = 0.105 \pm 0.005$; therefore the pyramid is slightly skewed. The angle between N and V can be calculated from $N \cdot V$ or $N \times V$, and is approximately $1-1/2^\circ$.

Table I lists the results of the calculation of one five-atom interstitial. The data in Tables II and III follow the same arguments as for the four atom interstitial, except that ten interstitials surround one lattice point (each interstitial is now shared by five atoms). There are again four orientations of the pyramidal configuration.

Summary

The locations and sizes of various interstitial holes have been calculated, using a hard sphere model, for the orthorhombic uranium lattice. The largest interstitial, having a diameter of $1.36\text{\AA}$, is surrounded by five atoms. There are eight of these in the unit cell.

LS:CR:EW

TABLE I

LOCATION OF VARIOUS INTERSTITIAL HOLES IN THE URANIUM LATTICE
(THE ORIGIN IS AT THE EDGE OF THE UNIT CELL)

Type	Atoms Associated	Hole Location		Diameter _o of Hole A
		General Terms	Numerical	
3 atoms in ab plane	$\begin{bmatrix} 0 & 0 & 0 \\ a & 0 & 0 \\ a/2 & b/2 & 0 \end{bmatrix}$	$\begin{bmatrix} a/2 & b/4 - \frac{a^2}{4b} & 0 \end{bmatrix}$	$[1.43 \ 1.12 \ 0]$	0.88
3 atoms not in ab plane	$\begin{bmatrix} 0 & 0 & 0 \\ a/2 & b/2 & 0 \\ a/2 & \frac{b}{2} - 2by & c/2 \end{bmatrix}$	Solved Numerically	$[0.44 \ 1.60 \ 0.88]$	1.00
4 atoms in corru- gated plane	$\begin{bmatrix} 0 & 0 & 0 \\ a & 0 & 0 \\ 0 & -2by & c/2 \\ a & -2by & c/2 \end{bmatrix}$	$\begin{bmatrix} a/2 & -by & c/4 \end{bmatrix}$	$[1.43 \ -0.62 \ 1.24]$	1.22
4 atoms in space	$\begin{bmatrix} 0 & 0 & 0 \\ a/2 & \frac{b}{2} - 2by & -c/2 \\ a/2 & \frac{b}{2} - 2by & -c/2 \\ 0 & -2by & -c/2 \end{bmatrix}$	$\begin{bmatrix} 0 & \frac{a^2 + b}{4b} - 2by & -\left\{c/4 + \frac{by}{c} \left(\frac{a^2}{b} + b - 4by\right)\right\} \end{bmatrix}$	$[0 \ 0.58 \ -1.84]$	1.10
5 atoms in space	$\begin{bmatrix} 0 & 0 & 0 \\ a & 0 & 0 \\ 0 & -2by & c/2 \\ a & -2by & c/2 \\ a/2 & -b/2 & 0 \end{bmatrix}$	$\begin{bmatrix} a/2 & \frac{a^2}{4b} - \frac{b}{4} & \left\{c/4 - \frac{by}{c} \left(b - \frac{a^2}{b} - 4by\right)\right\} \end{bmatrix}$	$[1.43 \ -1.12 \ 0.99]$	1.36

TABLE II

LOCATIONS OF VARIOUS INTERSTITIAL HOLES IN THE URANIUM UNIT CELL

Surrounded by
Four Atoms in Space

$$\begin{aligned} & \left[0 \quad u \quad \frac{1}{2}+v \right] \\ & \left[0 \quad u \quad -v \right] \\ & \left[0 \quad -u \quad v \right] \\ & \left[0 \quad -u \quad \frac{1}{2}-v \right] \\ & \left[\frac{1}{2} \quad \frac{1}{2}+u \quad \frac{1}{2}+v \right] \\ & \left[\frac{1}{2} \quad \frac{1}{2}+u \quad -v \right] \\ & \left[\frac{1}{2} \quad \frac{1}{2}-u \quad v \right] \\ & \left[\frac{1}{2} \quad \frac{1}{2}-u \quad \frac{1}{2}-v \right] \end{aligned}$$

where $u = \frac{a^2}{4b^2} + \frac{1}{4} - y$

$$v = \frac{by}{c^2} \left(\frac{a^2}{b} + b - 4by \right)$$

Surrounded by
Five Atoms in Space

$$\begin{aligned} & \left[0 \quad \frac{1}{2}-U \quad \frac{1}{2}-V \right] \\ & \left[0 \quad \frac{1}{2}-U \quad V \right] \\ & \left[0 \quad \frac{1}{2}+U \quad -V \right] \\ & \left[0 \quad \frac{1}{2}+U \quad \frac{1}{2}+V \right] \\ & \left[\frac{1}{2} \quad -U \quad \frac{1}{2}-V \right] \\ & \left[\frac{1}{2} \quad -U \quad V \right] \\ & \left[\frac{1}{2} \quad U \quad -V \right] \\ & \left[\frac{1}{2} \quad U \quad \frac{1}{2}+V \right] \end{aligned}$$

where $U = \frac{1}{4} - \frac{a^2}{4b^2} - y$

$$V = \frac{by}{c^2} \left(b - \frac{a^2}{b} - 4by \right)$$

TABLE III

LOCATIONS OF VARIOUS INTERSTITIAL HOLES AROUND AN ATOM
(THE ORIGIN TAKEN AT THE LATTICE POSITION OF THE ATOM)

<u>Type</u>	<u>General Coordinates</u>	<u>Evaluated Coordinates</u>
4 atoms in space	$\left[0, \frac{b}{4} + \frac{a^2}{4b} - 2by, \pm \left\{ \frac{c}{4} + \frac{by}{c} \left(\frac{a^2}{b} + b - 4by \right) \right\} \right]$ $\left[\pm \frac{a}{2}, \frac{b}{4} - \frac{a^2}{4b}, \pm \left\{ \frac{c}{4} - \frac{by}{c} \left(\frac{a^2}{b} + b - 4by \right) \right\} \right]$ $\left[0, -\frac{b}{4} - \frac{a^2}{4b}, \pm \left\{ \frac{c}{4} - \frac{by}{c} \left(\frac{a^2}{b} + b - 4by \right) \right\} \right]$	$[0, 0.58, \pm 1.84]$ $[\pm 1.43, 1.12, \pm 0.64]$ $[0, -1.81, \pm 0.64]$
5 atoms in space	$\left[0, \frac{b}{4} + \frac{a^2}{4b}, \pm \left\{ \frac{c}{4} - \frac{by}{c} \left(b - \frac{a^2}{b} - 4by \right) \right\} \right]$ $\left[\pm \frac{a}{2}, -\frac{b}{4} + \frac{a^2}{4b}, \pm \left\{ \frac{c}{4} - \frac{by}{c} \left(b - \frac{a^2}{b} - 4by \right) \right\} \right]$ $\left[\pm \frac{a}{2}, \frac{b}{4} - \frac{a^2}{4b} - 2by, \pm \left\{ \frac{c}{4} + \frac{by}{c} \left(b - \frac{a^2}{b} - 4by \right) \right\} \right]$	$[0, 1.81, \pm 0.99]$ $[\pm 1.43, -1.12, \pm 0.99]$ $[\pm 1.43, -0.11, \pm 1.48]$

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Fig. 1 - Effect of Temperature on Growth Rate - Growth Rate at 200°C = 1.

Fig. 2 - Model of uranium lattice illustrating interstitial surrounded by four atoms

Fig. 3 - Model of uranium lattice illustrating interstitial surrounded by five atoms

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