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ACTINIDE REDISTRIBUTION DUE TO PORE MIGRATION IN HYPOSTOICHIOMETRIC MIXED OXIDE FUEL PINS

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ACTINIDE REDISTRIBUTION DUE TO PORE MIGRATION IN HYPOSTOICHIOMETRIC MIXED OXIDE FUEL PINS

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ABSTRACT

Redistribution of plutonium due to migration of pores in hypostoichiometric mixed oxide fuel is analyzed. The unequal volatilities of the oxides of uranium and plutonium cause changes in the Pu/U ratio in the solid ahead of and in the wake of a moving pore. These elementary redistribution events of single pores are then combined to determine the gross redistribution in the fuel. For typical LMFBR fuel operating conditions, calculated redistribution due to pore migration is orders of magnitude smaller than that observed in irradiated pins.

1. Introduction

Actinide redistribution caused by the migration of porosity up the temperature gradient in a mixed oxide fuel pin has been the subject of several recent theoretical studies [1-6]. According to the model described in Ref. 4, the hot face of a moving disk-shaped pore becomes enriched in the less volatile of the two heavy metal components of the mixed oxide (usually plutonium) by preferential evaporation of the more volatile species across the pore. Solid state diffusion permits the concentration perturbation on the hot face to extend into the solid ahead of the moving pore. As a result, a "bow wave" of excess plutonium precedes the pore as it travels up the temperature gradient towards the central void. In order to satisfy conservation of plutonium along the path of the pore, a plutonium-deficient spike is generated at the initial position of the pore. The plutonium missing from this "start-up spike" is contained primarily in the plutonium-rich distribution preceding the migrating pore. However, the excess plutonium in the bow wave changes during migration for two reasons: (1) the pore moves into regions of increasing temperature and (2) at a fixed position, the ambient temperature gradually diminishes due to restructuring and central void formation. These two effects alter the plutonium fraction in the columnar grain formed between the initial and final pore locations. Figure 1 shows in schematic fashion the redistribution resulting from the movement of a single pore through virgin fuel (i.e., wherein the plutonium-to-heavy metal ratio ahead of the pore is the as-fabricated value q_o).

The analysis of Ref. 4 was restricted to hyperstoichiometric mixed oxide and considered only the redistribution effect of a single pore. In the present paper, the same model is applied to hypostoichiometric fuel and the cumulative effect of the migration of many pores in a cylindrical fuel pin on the time and radial distribution of the gross Pu/U+Pu ratio is considered. Section 2 describes the calculation of the pore velocity as a function of temperature, temperature gradient, ambient plutonium concentration (i.e., q_o) and local oxygen-to-metal ratio. Section 3 presents an analysis of the net effect of many pores sweeping a given position on the gross Pu/U+Pu ratio and Section 4 presents numerical results for typical irradiation conditions and operating times.

2. Actinide Redistribution Due to a Single Pore

We treat lenticular pores as disks of thickness δ and diameter d oriented perpendicular to the radial temperature gradient. For pores originating from the closed voidage in the as-fabricated fuel or those spawned during irradiation by cracks in the fuel, δ is typically of the order of 10 μm and the diameter is ~100 μm. During migration, cations (uranium and plutonium) and anions (oxygen) are transported from the hot face to the cold face. If the pore were infinite in extent transverse to the temperature gradient, diffusion of vapor species through the filling gas in the pore would be the only transport mechanism. For pores of finite diameter, however, fuel constituents can leak around the periphery of the disk by solid state diffusion [5,6]. The importance of the solid state diffusion path can be assessed by comparing the depth of penetration of the concentration distribution attached to the front face of the pore (λ, see Fig. 1) with the pore diameter (d). If $\lambda/d \ll 1$,

solid state diffusion around the edges of the disk is unimportant and only vapor transport across the gas space need be considered. At the opposite extreme of $\lambda/d >> 1$, the solid state diffusional mode of transport would be expected to dominate vapor transport.

As shown in Ref. 4, the characteristic penetration depth of the exponentially decaying concentration distribution in front of the pore is related to the pore velocity v_p and the solid state diffusion coefficient D_s by:

$$\lambda = D_s/v_p \tag{1}$$

Using the diffusivity of plutonium in mixed oxide suggested by Bober and Schumacher [7] (with an activation energy of 93 Kcal/mole) and the pore velocities calculated by the method described later in this section, λ for Pu is found to be ~1 μm at 2000°K and smaller at higher temperatures. Thus λ/d for plutonium is $\sim 10^{-2}$, which is sufficiently smaller than unity for solid state diffusion of this species around the edges of the lenticular pore to be neglected. For oxygen, on the other hand, D_s is about five orders of magnitude greater than it is for plutonium at 2000°K[8]. The penetration depth λ for oxygen is ~10 cm, which renders λ/d very much larger than unity. In this case, solid state diffusion is so rapid that any difference in O/M between the hot and cold faces of the pore which the vapor transport process attempts to establish cannot be sustained because of rapid oxygen migration in the matrix surrounding the pore. Consequently, the oxygen-to-metal ratios on both the hot and cold faces of the pore are taken to be equal to the local value in the

solid through which the pore is passing. Although oxygen concentration differences in the immediate vicinity of the pore cannot be set up, an O/M gradient along the radius of the fuel pin is established by other mechanisms (e.g., vapor transport in radial cracks or thermal diffusion in the solid).

The analysis reduces to determining: (1) The Pu/U+Pu ratio on the hot and cold faces of the pore (q_h and q_c, respectively) and (2) the pore velocity, v_p. The local values of the temperature, temperature gradient, O/M and the Pu/U+Pu ratio ahead of the pore (q_o) are assumed to be specified.

The plutonium concentration distribution in front of the pore shown in Fig. 1 decreases from q_h at $z = 0$ (the hot face of the pore) to q_o in the fuel well ahead of the pore. The quantity of excess plutonium (i.e., above that contained in the same volume of undisturbed fuel) per unit pore hot face surface area is (4):

$$A = \rho_s \Delta q_h D_s / v_p \tag{2}$$

where ρ_s is the theoretical density of the oxide and $\Delta q_h = q_h - q_o$.

Neglecting the amount of heavy metals contained in the vapor phase, a plutonium balance over the control volume moving with the pore and delineated by planes (1) and (3) in Fig. 1 gives:

$$\rho_s q_o v_p - \rho_s q_c v_p = \frac{dA}{dt}$$

or

$$\Delta q_c = -\frac{1}{\rho_s v_p} \frac{dA}{dt} \tag{3}$$

where $\Delta q_c = q_c - q_o$ and d/dt is the time derivative in the frame of reference moving with the pore. When converted to a stationary reference frame, the latter becomes:

$$\frac{d}{dt} = \frac{\partial}{\partial t} - v_p \frac{\partial}{\partial r}$$

where r is the radial location in the fuel pin. The minus sign appears because v_p is considered positive if the pore moves towards the center of the fuel rod. Combining Eqs. (2), (3) and (4) results in:

$$\Delta q_c = -\frac{1}{v_p} \frac{\partial}{\partial t}\left(\frac{\Delta q_h D_s}{v_p}\right) + \frac{\partial}{\partial r}\left(\frac{\Delta q_h D_s}{v_p}\right) \tag{5}$$

The first term on the right hand side of Eq. (5) represents loss of plutonium from the bow wave due to the change in fuel temperature at a fixed location. The second term on the right hand side represents rejection of plutonium from the hot face as the pore moves up a time-independent temperature gradient. These two loss terms are manifest as an increase in the plutonium fraction on the cold face of the pore.

A plutonium balance between planes (2) and (3) in Fig. 1 gives:

$$q_c = \frac{J^T_{Pu}}{J^T_{Pu} + J^T_U} \tag{6}$$

where J^T_{Pu} and J^T_U are the fluxes of plutonium and uranium, respectively, across the pore. These fluxes consist of components due to all vapor phase atomic and molecular forms of the heavy metals:

$$J^T_{Pu} = J_{Pu} + J_{PuO} + J_{PuO_2} \tag{7a}$$

$$J^T_U = J_U + J_{UO} + {}_{UO_2} + J_{UO_3} \tag{7b}$$

the flux of species i is given by Fick's first law integrated across the pore thickness:

$$J_i = \frac{D_{gi}}{RT} \frac{P_{ih} - P_{ic}}{\delta}, \quad i = Pu, PuO, \ldots UO_3 \tag{8}$$

where R is the gas constant and D_{gi} is the diffusion coefficient of gaseous species i in the inert gas filling the pore. The latter is assumed to be helium, and all the D_{gi} are assumed to be equal to that of UO_2 in helium. In all previous analyses of pore migration in oxide fuels, D_g has been calculated from the kinetic theory formula for hard sphere particles [9]. However, more accurate values of D_g may be obtained from the exact kinetic theory of gases, which gives the diffusivity in terms of the collision cross section σ and the collision integral Ω_D [10]:

$$D_g = \frac{1.858\text{x}10^{-3}\ T^{3/2} \left[(M_{UO_2} + M_{He})/M_{UO_2} M_{He} \right]^{1/2}}{p\sigma^2\ \Omega_D}$$

where p is the inert gas pressure in the pore and the collision integral is a function of temperature and the force constant ε of the interaction between UO_2 and He. For helium the force constants are estimated from

gas phase viscosity data [10]:

$$\sigma_{He} = 2.55\ \mathring{A}$$

$$\frac{\varepsilon_{He}}{k} = 10\ \mathring{K}$$

where k is Boltzmann's constant. For UO_2, the force constants may be estimated either from the properties at the boiling point ($T_B \simeq 3600°K$) or from the critical properties ($T_c \simeq 6000°K$, $V_c \simeq 90\ cm^3/mole$ [11]). The average values of the force constants of UO_2 estimated by these two methods are:

$$\sigma_{UO_2} = 3.78\ \mathring{A}$$

$$\frac{\varepsilon_{UO_2}}{k} = 5200°K$$

The force constants for the He-UO_2 interaction are computed from the usual combining rules (arithmetic mean for σ and geometric mean for ε). The diffusivity of UO_2 in He calculated from Eq. (9) is approximately six times larger than that obtained by the analogous hard sphere formula [9] at all temperatures of interest (2000°K to 3000°K).

In order to determine the inert gas pressure p in Eq. (9), we assume that helium at 1 atm pressure is sealed into the fuel by sintering during fuel fabrication. The pressure during reactor operation is therefore:

$$p = \frac{T}{T_{sint}} \tag{10}$$

where $T_{sint} \simeq 2000°K$ is the temperature at which the fuel was sintered during preparation of the pellets.

The partial pressures p_i in Eq. (8) are determined by the temperature and oxygen-to-metal ratio in the solid at the radial location of the pore and by the Pu/U+Pu ratio on the particular face of the pore under consideration [12]. The temperatures on the two faces of the pore are given by:

$$\begin{aligned} T_h &= T + \frac{1}{2}\,\delta\nabla T_p \\ T_c &= T - \frac{1}{2}\,\delta\nabla T_p \end{aligned} \tag{11}$$

where T is the temperature in the fuel at the position and time in question. ∇T_p is the temperature gradient through the pore, which is equal to the macroscopic temperature gradient in the solid multiplied by a factor of ~ 4 to account for the lower thermal conductivity of the gas in the pore compared with that of the surrounding matrix [9]. Both T and ∇T_p are strong functions of radial position and, because of redistribution, vary slightly with time as well.

As discussed earlier, the oxygen-to-metal ratio is the same on both sides of the pore:

$$(O/M)_h = (O/M)_c = (O/M)_o \tag{12}$$

where $(O/M)_o$ is the local oxygen-to-metal ratio at the radial position of the pore.

Finally, the pore velocity is given by:

$$v_p = \frac{J^T_{Pu} + J^T_U}{\rho_s} \tag{13}$$

The remaining unknowns, q_c and q_h, are determined by simultaneous solution of Eqs. (5-13) with p_{ih} $[T_h, q_h, (O/M)_o]$ and p_{ic} $[T_c, q_c, (O/M)_o]$ given by the thermochemistry of mixed oxide vaporization.

In order to simplify the calculation, we tentatively assume that $\Delta q_c \simeq 0$, or $q_c \simeq q_o$. This simplification requires that the time and distance derivatives on the right hand side of Eq. (5) be small, or that the quantity of plutonium accompanying the front face of the pore (A) change slowly with t and r. With this simplification, the remaining equations are all algebraic and can be readily solved for q_h. Using the value of $\Delta q_h = q_h - q_o$ so computed to determine the time and position dependence of $(\Delta q_h D_s/v_p)$, Δq_c can be calculated from Eq. (5). If $|\Delta q_c| << |\Delta q_h|$ under all conditions, the approximate method is satisfactory. In the numerical example presented in Sec. 4, $|\Delta q_c|/|\Delta q_h| < 10^{-3}$. The simplified calculational method was also utilized in Ref. 4 for the analysis of pore migration in hyperstoichiometric fuel. It may be shown that the conditions for its validity are satisfied in this case as well.

The analysis described above permits computation of the entire plutonium concentration distribution shown schematically in Fig. 1. The distribution satisfies the overall plutonium balance:

$$A_o(r_o) = A(r) + \rho_s \int_r^{r_o} \Delta q_c(r)\,dr \qquad (14)$$

where the subscript o refers to the initial location of the pore.

3. Gross Plutonium Redistribution

Section 2 described the method of computing the redistribution of plutonium arising from the migration of a single pore up a temperature gradient. In this section, we describe the method of determining the gross change in the Pu/U+Pu ratio in a region of fuel which contains many pores and through which many pores have passed in the restructuring process. In the calculation, we assume that the pore number distribution function, $N(r,t)$ = number of pores per unit volume of fuel at radial position γ and time t, is known. In addition, all pores are assumed to be of the same size, namely disks of diameter d and thickness δ. The surface area of each of the faces of the pore perpendicular to the temperature gradient is:

$$S_p = \frac{\pi}{4} d^2 \qquad (15)$$

and the volume of each pore is $S_p\delta$.

To determine the gross plutonium redistribution due to the migration of pores, consider the cylindrical annulus of thickness dr and unit height at radial location γ shown in Fig. 2. The total mass of plutonium contained in this volume element in excess of what would be there if the plutonium fraction were everywhere equal to q_o (i.e., if single pores

performed no redistribution) is composed of three contributions:

(1) The amount of plutonium left by the pores which were initially in the volume element at startup. This contribution is negative and is composed of the startup spikes from the initial porosity in dr:

$$Q_1 = - S_p A_o(r) N_o (2\pi r dr) \tag{16}$$

where N_o is the pore density in the as-fabricated fuel.

(2) The amount of plutonium contained in the bow waves attached to the front of the pore which happen to be in the volume element at time t:

$$Q_2 = S_p A(r,t) N(r,t) (2\pi r dr) \tag{17}$$

(3) The amount of plutonium left by the trails of pores which have passed completely through the volume element of Fig. 2 between startup (t=0) and time t. The number of pores which pass radial position r in a time interval dt' is equal to the difference between the number of pores between r and the outer fuel radius R at time t' and the number at time t' + dt':

$$\int_r^R [N(r',t') - N(r',t'+dt')]\, 2\pi r' dr' = - \int_r^R \frac{\partial N}{\partial t'}\, dt'\, 2\pi r' dr'$$

The plutonium enrichment in the volume element of Fig. 2 due to each of the transit pores is $(S_p dr)\rho_s \Delta q_c$ which, when multiplied by the above

expression yields the amount of plutonium left in the volume element by moving pores in time interval dt':

$$S_p dr \rho_s \Delta q_c \left[- \int_r^R \frac{\partial N}{\partial t'} \, dt' \, 2\pi r' dr' \right]$$

Integrating the above expression from t'=0 to t'=t gives the third component of the plutonium enrichment in the volume element:

$$Q_3 = - 2\pi dr \, S_p \rho_s \int_0^t \Delta q_c(r,t') dt' \int_r^R \frac{\partial N(r',t')}{\partial t'} \, r' dr' \tag{18}$$

The gross enrichment in the fuel in the cylindrical volume element of Fig. 2 is:

$$\Delta \bar{q} = \frac{Q_1 + Q_2 + Q_3}{\rho_s \, 2\pi r dr \, (1 - P)} \tag{19}$$

where the denominator represents the total mass of heavy metals in the volume element and P is the porosity at r and t. P is related to the pore density by:

$$P(r,t) = S_p \delta \, N(r,t) \tag{20}$$

Substituting Eqs. (16-18) into Eq. (19) and replacing N by P according to Eq. (20) yields:

$$\Delta\bar{q} = \frac{1}{\delta\rho_s[1 - P(r,t)]} \left\{ A(r,t)P(r,t) + A_o(r)P_o - \frac{\rho_s}{r} \int_0^t \Delta q_c(r,t')dt' \cdot \int_r^R \frac{\partial P}{\partial t'} r'dt' \right\} \tag{21}$$

Although the pore thickness appears in the denominator of Eq. (21), this parameter has little effect on actinide redistribution; the quantity of plutonium carried by each pore, A, is proportional to Δq_h, and both Δq_h and Δq_c are proportional to δ. To first order, Eq. (21) is independent of pore thickness.

4. Numerical Example

Eq. (21) shows that determination of the redistribution $\Delta\bar{q}$ due to pore migration requires knowledge of the time dependent porosity distribution during restructuring. The pore conservation equation which determines P(r,t) is (3);

$$\frac{\partial P}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}(rv_p P) \tag{22}$$

The pore velocity in this equation is a function of the temperature distribution, which is given by solution of:

$$\frac{1}{r}\frac{d}{dr}\left[rk(P,T)\frac{dT}{dr}\right] + H(r,t) = 0 \tag{23}$$

where k(P,T), the thermal conductivity of the fuel, is a function of local porosity and temperature. H(r,t) is the volumetric heat generation

rate in the fuel. It depends upon the radial distribution of plutonium, so that all three conservation equations (energy, porosity and plutonium) are coupled to each other. In general, simultaneous solution of Eqs. (21-23) is needed to describe analytically the restructuring/redistribution process. In order to simplify the calculation, we assume that the heat source H is not significantly affected by actinide redistribution, which is valid when $\Delta\bar{q}$ is small. The validity of this simplification can be verified at the end of the computation.

The only remaining dependence of Eqs. (22) and (23) upon the nature of the fuel (assuming that the thermal conductivity of $(U,Pu)O_2$ and UO_2 are approximately equal) is in the pore velocity. Using the method described in Sec. 2, we have computed v_p in $U_{0.8}Pu_{0.2}O_{2-x}$ and compared the results with those in pure UO_2 at the same temperature and temperature gradient. The latter were taken from Sens' (9) calculations. Over a wide range of values of T, ∇T and x, we find that the pore velocities in the mixed oxide are $\sim$5 times larger than those calculated by Sens for the same T and ∇T. This factor consists of two parts: First, our pore velocities are a factor of 6 larger than Sens' because the diffusion coefficient of the heavy metal oxides in helium computed from the exact kinetic theory (Eq. (9)) differs from that obtained from the hard sphere gas kinetic theory employed by Sens by this amount. Second, even for the same gas phase diffusivity, our calculations for the mixed oxide predict velocities lower by a factor of $\sim$5/6 than those of pores moving in pure UO_2. The reason for this difference is that the plutonium component of hypostoichiometric fuel is slightly less volatile than pure UO_2 at the same temperature. Despite the presence of volatile UO_3, the

pore velocity is controlled by vapor transport of the least volatile component across the pore. Even in highly hyperstoichiometric mixed oxide, the pore velocities are only 3-1/2 times larger than in pure UO_2(4).

The fact that the pore velocities in our calculation for the hypostoichiometric mixed oxide differ from those obtained by Sens for pure UO_2 by a constant factor means that Sens' numerical solution of Eqs. (22) and (23) for pure UO_2 can be applied directly to the mixed oxide case simply by scaling the time. Sens' results are reproduced in Fig. 3. We apply these distributions to the mixed oxide fuel by dividing the times listed on the figure by a factor of 5. In addition, the curve for 60 sec is assumed to represent the startup temperature distribution. While this device for avoiding numerical solution of Eqs. (22) and (23) is approximate, its accuracy is believed to be sufficient for our primary purpose, which is to ascertain whether actinide redistribution by pore migration is at all significant. Consequently, when P(r,t) or T(r,t) are required in Eq. (21), we have used the curves shown in Fig. 3 with the times reduced by a factor of 5.

Calculation of the quantity of plutonium carried ahead of the pores (Λ in Eq. (21)) requires specification of the O/M ratio. We have used the method of Adamson and Aitken[13] for computing oxygen redistribution in hypostoichiometric fuel for determining the radial distribution of O/M. Figure 4 shows the O/M profile for the temperature distributions shown in Fig. 3 and for an average O/M of 1.98.

Figure 5 shows the plutonium enrichment at the hot face of the pore for various times during the process. The plutonium enrichment at the

cold face (calculated from Δq_h and Eq. (5)) is shown in Fig. 6. Δq_c is negative in the center of the fuel and becomes positive towards the edge of the columnar grain zone. The sign changes because of changes in the relative magnitudes of the two terms on the right hand side of Eq. (5). At all radial positions and times, the magnitude of Δq_c is less than 10^{-3} Δq_h, which justifies a postiori the use of the simplified calculational method described in Sec. 2.

Insertion of all of the quantities determined as described above into Eq. (21) produces the gross plutonium redistribution profiles shown in Fig. 7. The simplified method used to compute Δq_h and Δq_c fails as the pores approach the central void, where all of the excess plutonium carried by the pores is deposited. Hence, except for the single time (t_1 = 3 min) at which the central porosity is less than unity, the plutonium distributions cannot be calculated all the way to the central void. However, the overall plutonium balance:

$$\int_{r_o}^{R} \Delta\bar{q}(r,t)\,r\,dr = 0 \tag{24}$$

where r_o = radius of the central void, must be satisfied. We have used this relation to estimate the distributions in the region close to $r = r_o$ and these estimates are shown as dashed lines in Fig. 7. The calculated plutonium concentrations differ by less than 0.05% from the original concentration (q_o = 0.2).

5. Conclusion

The extent of plutonium redistribution calculated in this study is approximately two orders of magnitude smaller than that observed experimentally [14]. Although the times involved in our study are quite short, they are sufficient to cause appreciable restructuring, as evidenced by the porosity distributions in Fig. 3. Our calculations suggest that migration in hypostoichiometric mixed oxides is not the mechanism responsible for the actinide redistribution observed in irradiated fuel pins.

Akcnowledgment

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References

[1] W. J. Lackey, F. J. Homan and A. R. Olsen, Nucl. Tech. 16, (1972) 120.

[2] R. O. Meyer, J. Nucl. Mater. 50 (1974) 11

[3] D. R. Olander, J. Nucl. Mater. 47 (1973) 251.

[4] D. R. Olander, J. Nucl. Mater. 49 (1973/74) 35.

[5] R. C. Meyer, J. Nucl. Mater., to be published.

[6] D. R. Olander, J. Nucl. Mater., to be published.

[7] M. Bober, G. Schumacher and D. Beithoff, J. Nucl. Mater., 47 (1973) 187.

[8] P. E. Blackburn, ANL-RDP-20 (1974).

[9] P. F. Sens, J. Nucl. Mater., 43 (1972) 293.

[10] R. C. Reid and T. K. Sherwood, "The Properties of Gases and Liquids," 2nd Ed., (1960) McGraw-Hill, New York, pp 268-271.

[11] D. C. Menzies, AERE Report TRG 1119(D) (1966).

[12] P. E. Blackburn, Computer program for calculating equilibrium pressures over mixed uranium-plutonium oxides, personal communication (1974).

[13] M. G. Adamson and E. A. Aitken, NEDO-12324 (1972).

[14] R. O. Meyer, E. M. Butler and D. R. O'Boyle, ANL-7929 (1972)

Figure Captions

1. Plutonium redistribution due to motion of a pore.
2. Diagram for computing the plutonium balance on a cylindrical annulus in the fuel.
3. Porosity and temperature distributions in a UO_2 fuel pin during restructuring. Fuel radius: 0.5 cm; surface temperature: 600°C; linear power: 600 W/cm; initial porosity: 10% (after Ref. 9).
4. Calculated O/M. distribution for the fuel pin of Fig. 3 (times divided by factor of 5, see text) Avg. O/M: 1.98 (after Ref. 13).
5. Plutonium enrichment on the hot faces of pores moving up the temperature distribution shown in Fig. 3 and the O/M distribution of Fig. 4 as a function of pore radial location and time.
6. Plutonium enrichment on cold faces of moving pores corresponding to the hot face enrichments shown in Fig. 5.
7. Gross plutonium redistribution in hypostoichiometric fuel due to pore migration. Avg. O/M: 1.98; thermal conditions listed in caption of Fig. 3.

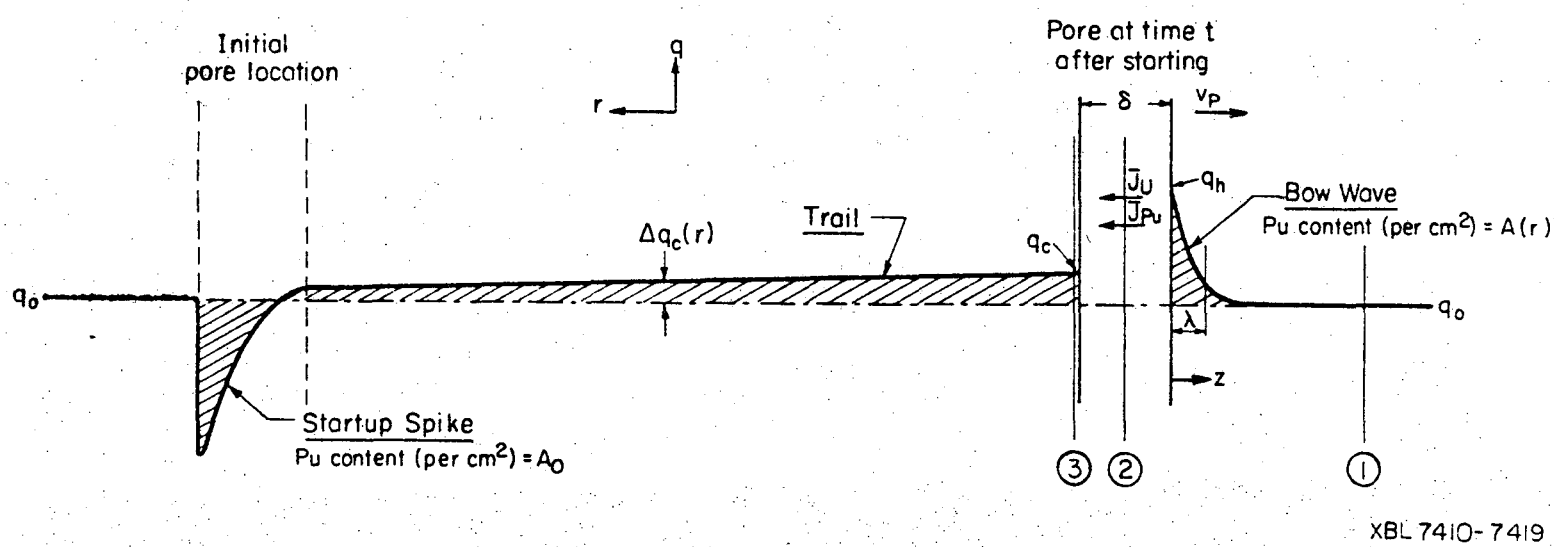
Initial
pore location
q
r
Pore at time t
after starting
δ
v_P
J_U
J_{Pu}
q_h
Bow Wave
Pu content (per cm²) = A(r)
$\Delta q_c(r)$
Trail
q_c
q_o
q_o
λ
z
Startup Spike
Pu content (per cm²) = A_0
3
2
1
XBL 7410-7419

Fig. 1

0 0 0 0 4 2 0 2 0 5 8

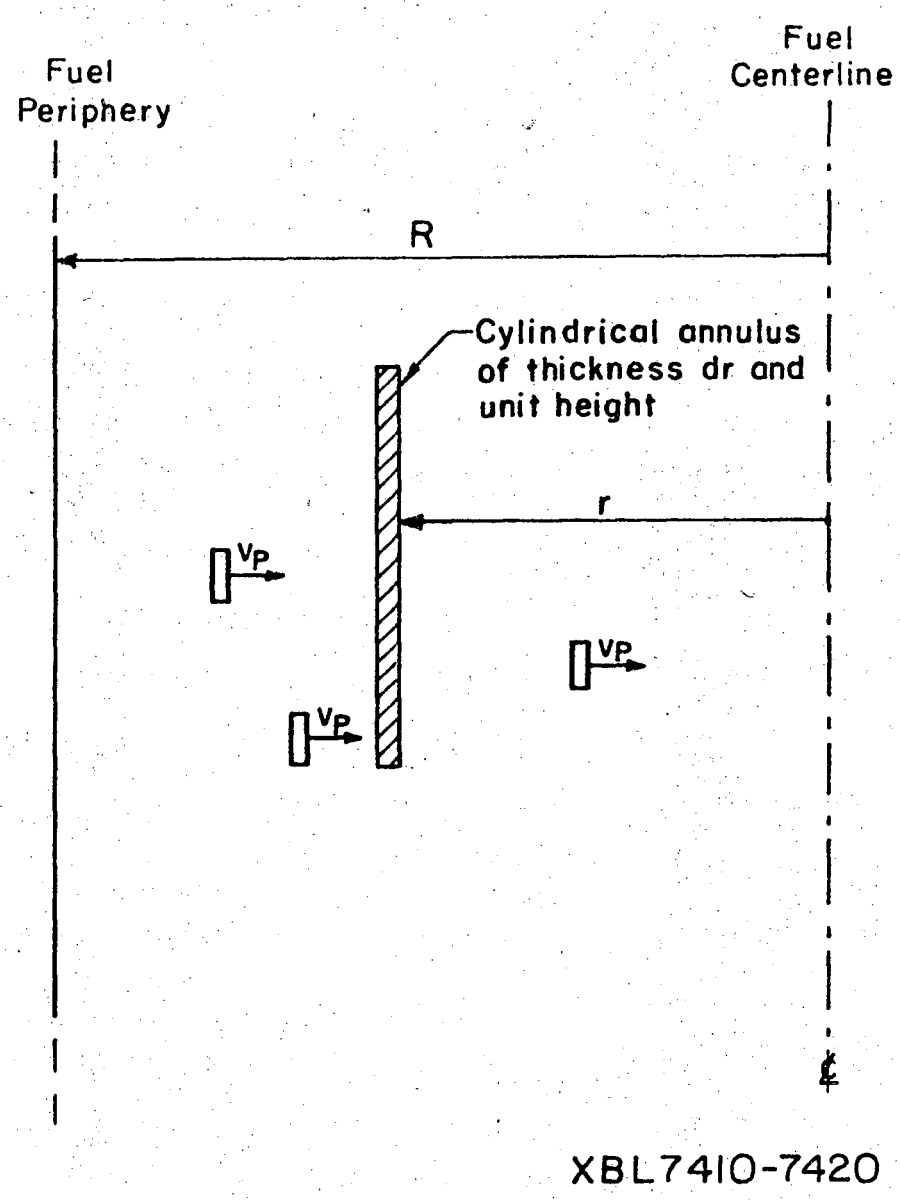

Fig. 2

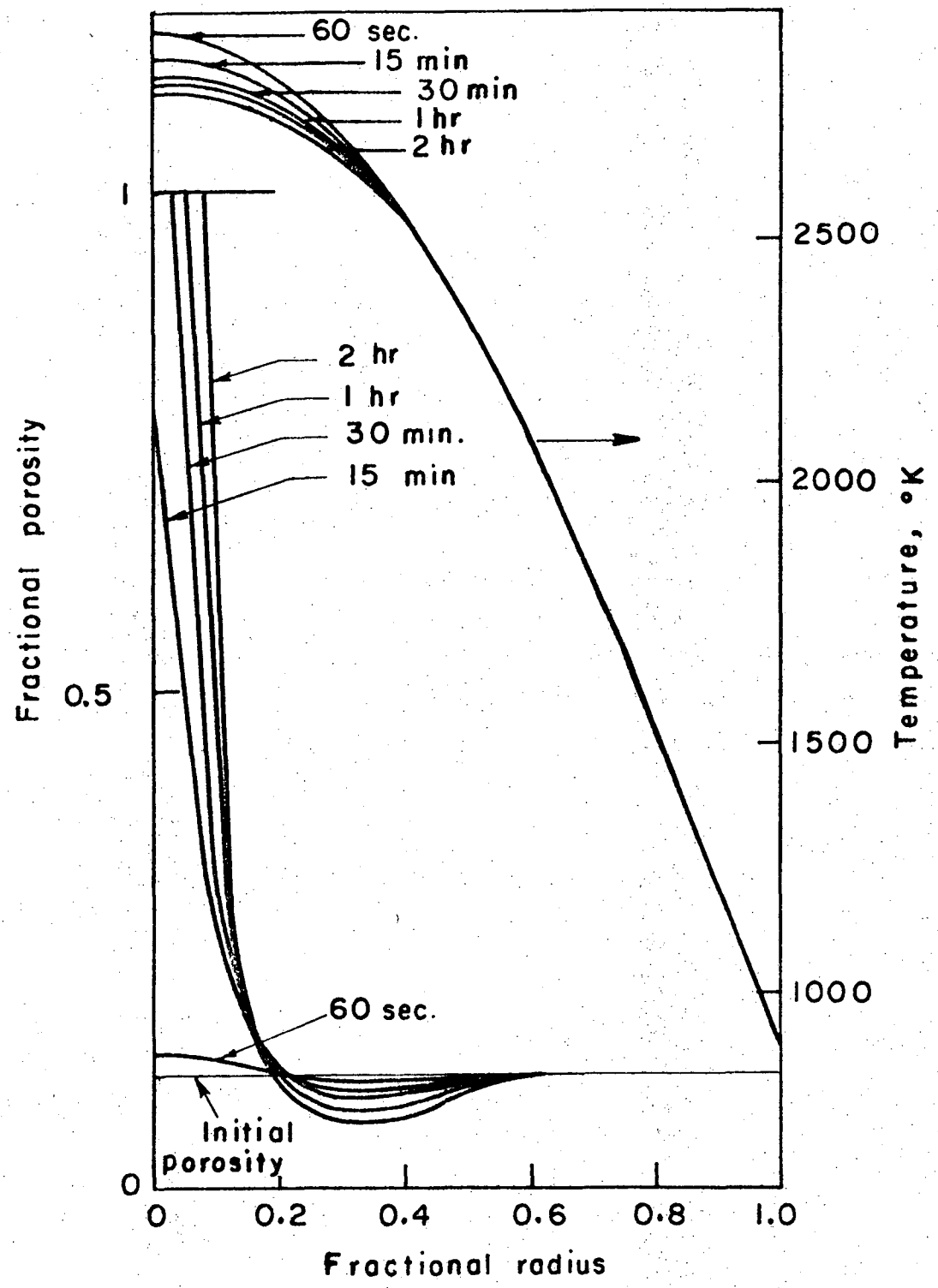

Fig. 3 XBL733-2582

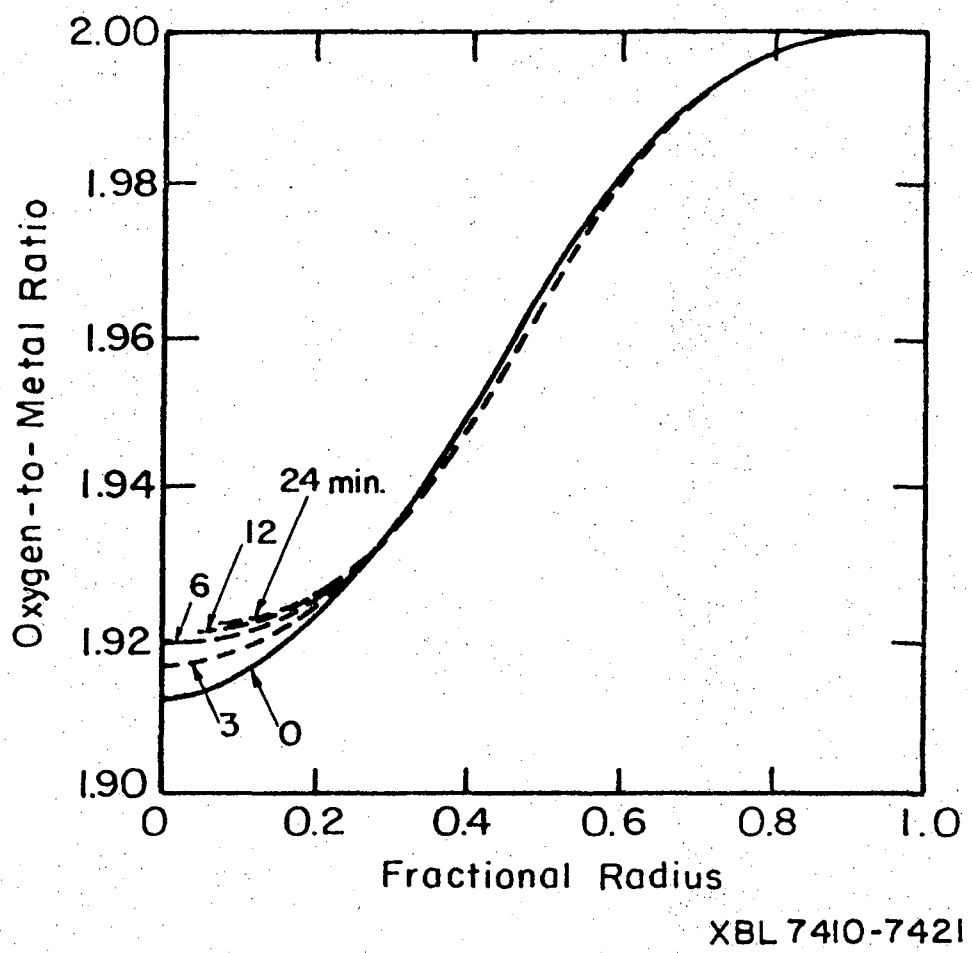
2.00
1.98
1.96
1.94
1.92
1.90
Oxygen-to-Metal Ratio
24 min.
12
6
3
0
0
0.2
0.4
0.6
0.8
1.0
Fractional Radius
XBL 7410-7421

Fig. 4

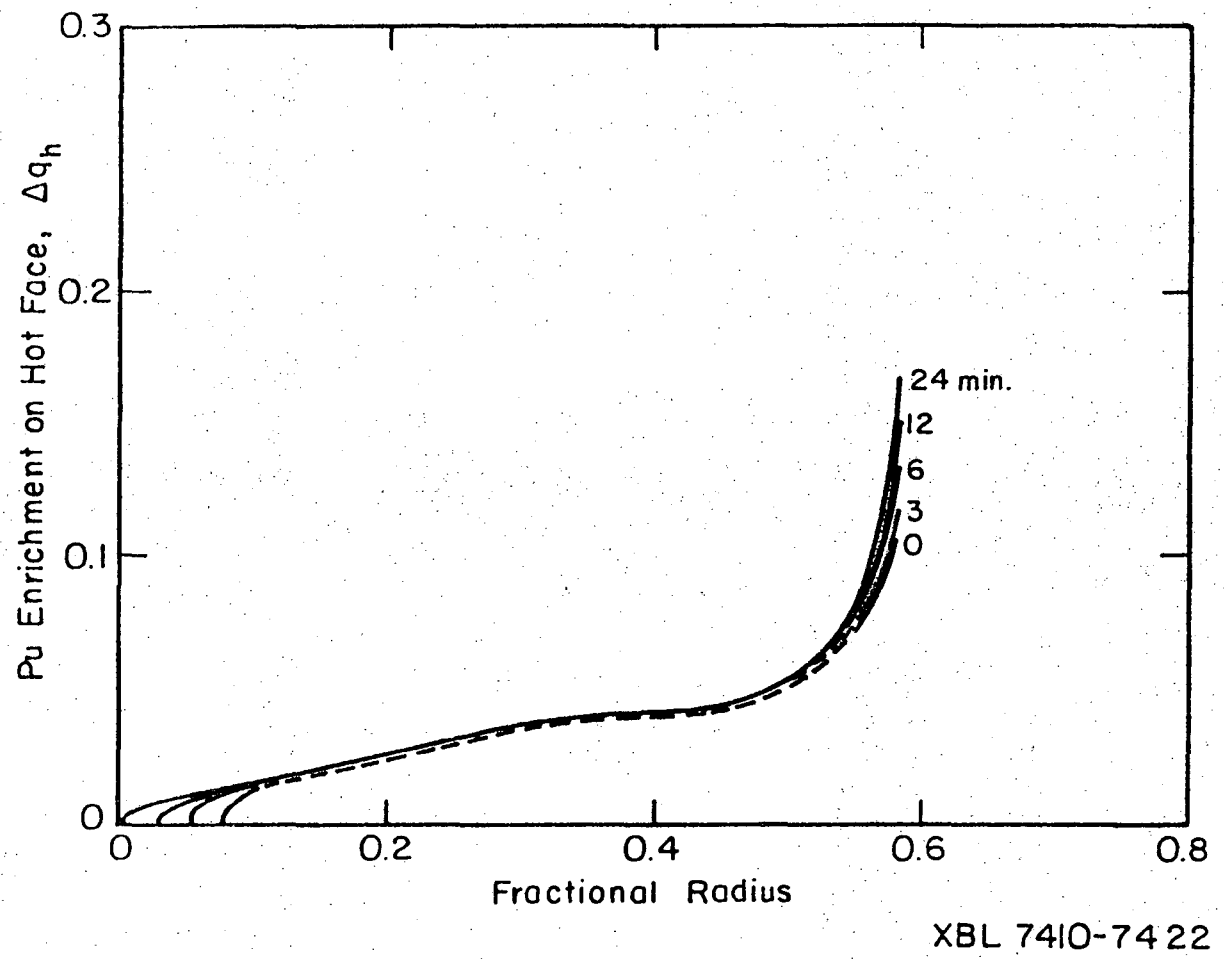
0.3
0.2
0.1
0
Pu Enrichment on Hot Face, Δq_h
24 min.
12
6
3
0
0
0.2
0.4
0.6
0.8
Fractional Radius
XBL 7410-7422

Fig. 5

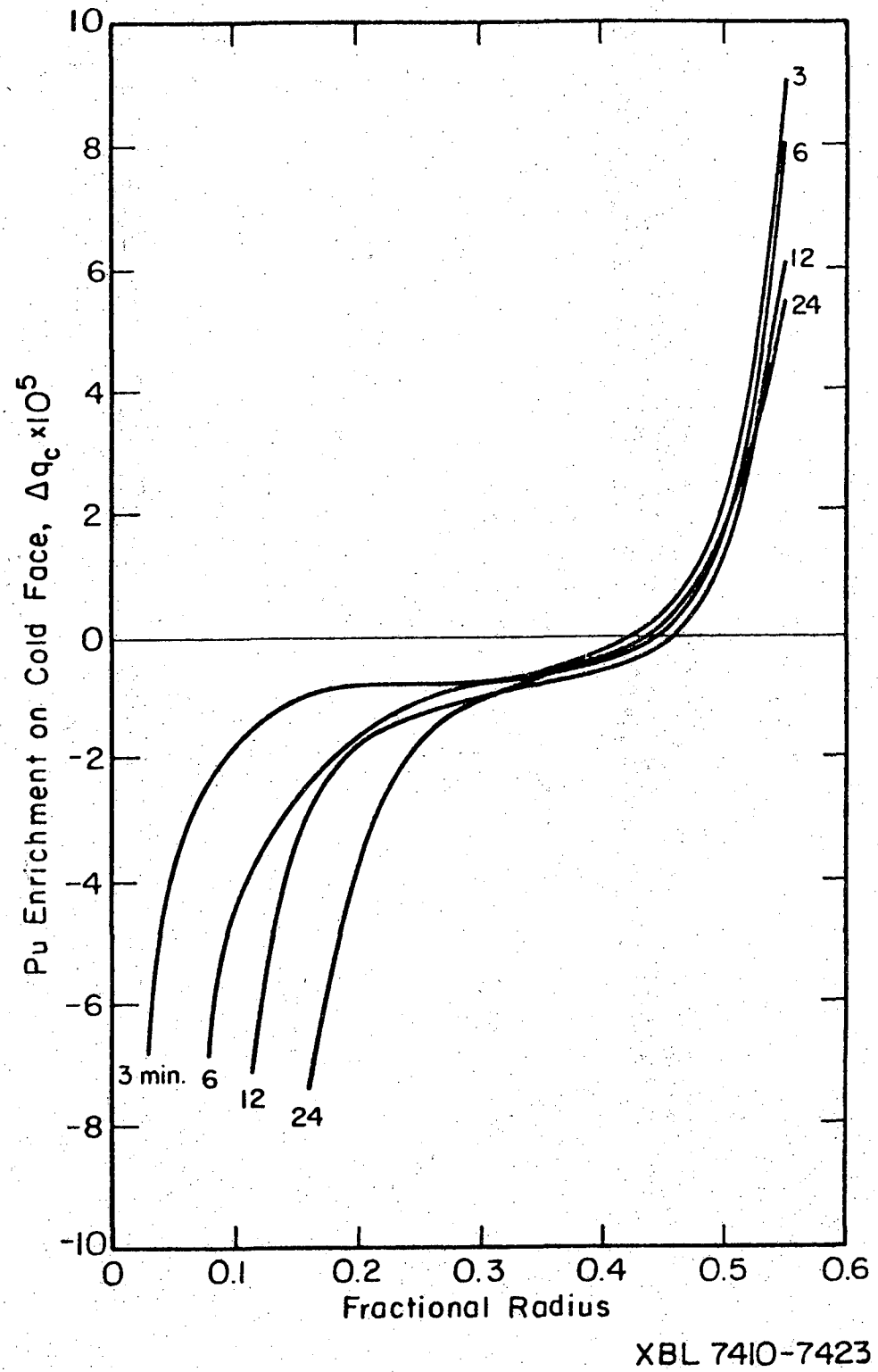
10
8
6
4
2
0
-2
-4
-6
-8
-10
Pu Enrichment on Cold Face, $\Delta q_c \times 10^5$
0
0.1
0.2
0.3
0.4
0.5
0.6
Fractional Radius
3
6
12
24
3 min.
6
12
24
XBL 7410-7423

Fig. 6

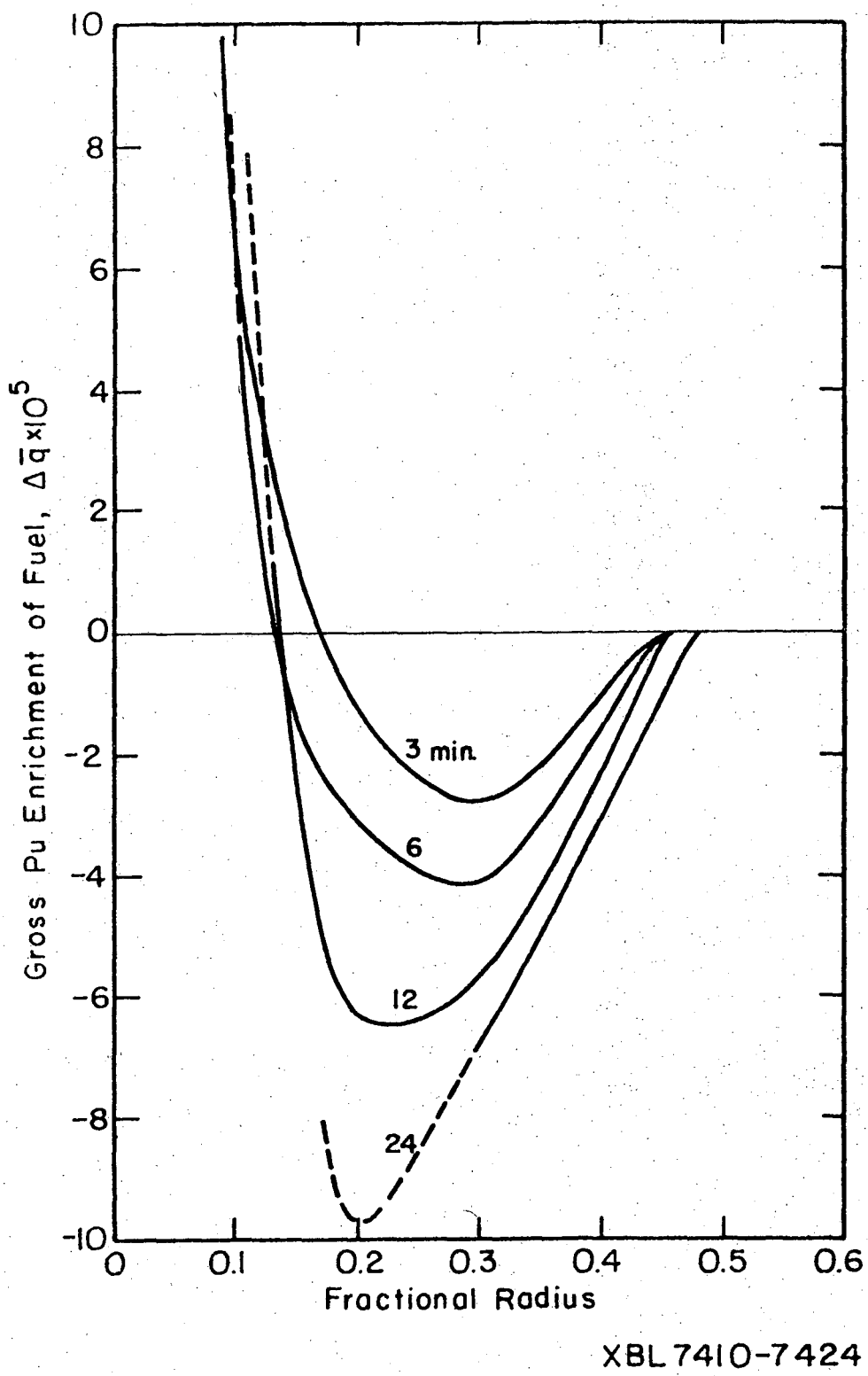
10
8
6
4
2
0
-2
-4
-6
-8
-10
Gross Pu Enrichment of Fuel, $\Delta\bar{q}\times10^5$
3 min.
6
12
24
0
0.1
0.2
0.3
0.4
0.5
0.6
Fractional Radius
XBL 7410-7424

Fig. 7

0 0 0 0 4 2 0 2 0 6 1

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