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SOME FIRE AND FISSION PRODUCT RELEASE POSSIBILITIES FOR DISCHARGED FUEL SLUGS

AUTHOR

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SOME FIRE AND FISSION PRODUCT RELEASE POSSIBILITIES FOR DISCHARGED FUEL SLUGS

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SOME FIRE AND FISSION PRODUCT RELEASE POSSIBILITIES FOR DISCHARGED FUEL SLUGS

INTRODUCTION

Increasing power levels and consideration of fuel discharge techniques which will minimize production interference due to fuel exchange with the reactor have prompted increasing attention to the hazards associated with abnormal residence of slugs in the rear face area, i.e., "held-up" slugs.

In response to Ref. 1 an attempt has been made to appraise the slug fire and fission product release possibilities associated with held-up slugs, as a function of slug power and down-time before discharge. The slug temperature history data were calculated by F. Zaloudek⁽²⁾. The fission product release estimates were calculated from fission product release data of R. K. Hilliard⁽³⁾. The present document is essentially a commentary on these two studies. The limitations of the data involved here will be discussed briefly and some suggestions given for further work to permit needed refinement of these, and related hazards analyses.

SUMMARY AND CONCLUSION

1. The fission product release potential of a single slug is more than sufficient to justify techniques for minimizing the incidence of slug hold-up in the rear face area. Such techniques are under development and should reduce the possibility of slug hold-up to the point where operational discharge is justifiable on a hazards basis.
2. Normal slugs discharged from an operating reactor (~ 0 decay time) might be expected to reach a fire-sensitive state in a time the order of 5 minutes following hold-up. Abnormal slugs, bearing pyrophoric corrosion derivatives such as UH_3 , could ignite any time after discharge, or during in-reactor residence following loss of cooling control.
3. The temperature, chemical, and physical histories of the uranium following discharge are obscured by uncertainty concerning the influence of U-Al diffusion products and various oxidation products on the response of the slug to the rear face environment.
4. The rear face fog spray system has demonstrated an ability to markedly reduce the radiological contamination resulting from slug fires in the discharge area⁽⁵⁾. Heat transfer calculations indicate that the fog spray is also a potent heat dissipation agency and, if applied in time, can substantially reduce the temperature excursion experienced by the slug.
5. Consideration of relaxing restrictions on public access to lands adjoining the Hanford site will require refinement of the fission product release analyses permitted by present knowledge. Basically two aspects of a reactor accident require clarification, namely: a) the physical-chemical history of radioactive material following loss of cooling control--for both the in-pile and ex-pile environments; b) the ability of the reactor structure proper, and the containment system leading to the high stack, to remove harmful fission products from the effluent air. A significant extension of present knowledge of these complex

matters will require, in addition to studies of fission product characteristics, further experimental and theoretical analysis of the physical and chemical response of a slug to simulated accident conditions. The rear face area of Hanford production reactors provides radiation protection and disposal features which could not be duplicated in a separate test facility without a prohibitive expenditure. Consideration should be given to rear face area tests oriented toward obtaining better knowledge of a) the fission product decay power, for various exposure histories and decay times, and b) the ability of the rear-face environment to cool held-up slugs, with and without the fog spray. Such tests, employing full-scale operational slugs, are regarded as an indispensable aid to greater realism in reactor hazards analyses.

SLUG TEMPERATURE HISTORY FOLLOWING DISCHARGE

Ref. 2 contains the details of the slug temperature calculations and they will not be repeated here. The calculations are based on a standard K III I&E slug, horizontally situated in the rear face air. The following variables were considered: slug power (52.3 to 67.3 KW, 1 year exposure); down time before discharge (0 to 2 hours). An "average" slug temperature was determined as a function of time after discharge--the calculations stopping at the inception of uranium melting. Because of time limitations, no attempt was made to take into account possible physical/chemical interaction between the cladding and the uranium, or chemical reactions between the ambient air and the uranium following melting, and loss, of the cladding. It was recognized, however, that the chemical power generated by the U-air reaction could significantly affect the temperature history of the slug.

A summary of some of the data of Ref. 2 is given in Table I. The data for 0 decay and 1 hour decay are plotted in Figure 1, to permit more convenient interpolation and extrapolation to other slug powers. For the purposes of estimating fission product release, a rough estimate was made of the time to completely melt the slug (cf App. A). This melting time was added to the ϕ_u time of Zaloudek to obtain the ϕ_{melt} time (cf Table I).

Because some oxidation of the uranium would be inevitable under the assumptions of these calculations, an attempt was made to estimate the effect of air oxidation on the slug temperature history (cf App. B). It was assumed that the U-air reaction begins at the time the cladding is melted. By a stepwise correction to the T- ϕ characteristic as given by Zaloudek, a rough estimate was obtained of the chemical heating effects. Corrected times to reach the MP of the U, and to melt the slug, are plotted in Figure 1 along with Zaloudek's original data. The possible protective effects of a U-Al diffusion layer (cf Ref. 4), or an oxide layer, make these estimates of chemical heating highly speculative. High slug powers and zero decay tend to favor the assumptions of App. B. This simple analysis of what is, in reality, a complex heat exchange problem suffices, however, only to indicate the influence that chemical reaction might have on the slug temperature history.

An attempt was made in Ref. 2 to appraise the effectiveness of the rear face fog spray system as a slug cooling agency. Zaloudek, using heat transfer data obtained with similar spray systems, estimates that the fog spray would turn back the slug temperature excursion short of the uranium MP if the spray were activated at any time prior to the ϕ_u time in the absence of the spray. Zaloudek also estimates that the fog spray would turn back the slug temperature excursion short of cladding melting

TABLE I

TIME FOR VARIOUS EVENTS IN SLUG POST-DISCHARGE HISTORY (From Ref. 2)
(Assuming no chemical heating)

Slug Power (kw)- 1 yr. 52.3				59.7			67.3		
ϕ_D	ϕ_{A1}	ϕ_U	ϕ_{Melt}	ϕ_{A1}	ϕ_U	ϕ_{Melt}	ϕ_{A1}	ϕ_U	ϕ_{Melt}
(Min)	(Min)	(Min)	(Min)	(Min)	(Min)	(Min)	(Min)	(Min)	(Min)
0	2.33	6.81	9.81	2.08	5.92	8.5	1.83	5.25	7.5
1	4.34	10.76	15.4	3.83	9.50	13.5	3.33	8.25	11.8
10	7.58	16.9	24.1	6.65	14.4	20.6	5.75	12.7	18.1
60	11.23	27.7	39.4	10.0	22.6	32.2	8.75	19.2	27.0
120	14.8	40.0	60.0	12.5	31.5	44.1	10.56	24.1	34.3

Definitions:

- ϕ_D - Time between shutdown and discharge.
- ϕ_{A1} - Time between discharge and start of cladding (A1) melting.
- ϕ_U - Time between discharge and start of uranium melting.
- ϕ_{Melt} - Time between discharge and complete melting of slug.

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FIGURE 1

Approximate Time for Various Events in Slug History Following Discharge.

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if it were activated prior to discharge of the slug to the air. These conclusions apply to the highest slug power (67.3 KW) considered in these calculations and to the zero decay case.

No attempt was made to factor chemical heating into the estimate of fog spray cooling effectiveness. The $\text{U-H}_2\text{O}$ reaction, which would be active along with the $\text{U-air (O}_2\text{)}$ reaction under fog spray conditions, generates roughly 1/2 the heat generated by the U-O_2 reaction. There is, however, evidence (cf Ref. 4) that water vapor, up to 100% relative humidity, increases the U-O_2 reaction rate. If true, then the lesser heat release of the $\text{U-H}_2\text{O}$ reaction would have little significance. Very little can be said about the physical characteristics, e.g., the protective quality, of the oxides generated under the conditions of interest here. At temperatures below roughly 400 C, hydrogen involvement in the oxidation process appears to discourage the formation of an adherent, protective, oxide layer. This action would tend to improve the cooling power of the fog spray system. In general, however, gross uncertainties concerning the oxidation kinetics of U in an air-steam atmosphere discourage any attempts to estimate the effect of chemical reaction on fog spray cooling effectiveness. This state of affairs, of course, seriously limits the significance of the chemical heating effects given in App. B because of the probability that water vapor will be a participant in any slug incident in the rear face area.

SLUG DAMAGE AND FIRE POSSIBILITIES

Some of the factors which affect the complex phenomenon of pyrophoricity were reviewed in Ref. 4. It was suggested that most of the slug fires experienced at Hanford reactors are attributable to pyrophoric corrosion derivatives, such as UH_3 , or powdered U generated by UH_3 decomposition. There is little, or no evidence that the massive uranium of the slug participated in the slug fire events to any extent. Work at Argonne, Stanford Research and elsewhere on the pyrophoricity of metals has clearly identified the dominant role of specific area (surface area/mass) in ignition and propagation of combustion in metals. Ignition of powder, foil and small cube specimens is one thing. Ignition and propagation involving a massive system, such as a fuel slug is quite a different matter. Without a large increase in the effective surface area of the U, caused by gross irradiation damage, it is not a certainty that a fire could be developed, and sustained, in a massive piece of solid, irradiated, U exposed to the rear face atmosphere. Work at ORNL has suggested that irradiation markedly increases the oxidation rate of U in air. Hanford studies, however, have not revealed any significant increase in the oxidation susceptibility of U due to irradiation per se. As suggested in Ref. 4, irradiation influence on the pyrophoricity of U remains a moot subject--a state of affairs indicating that the evidence for a powerful irradiation influence is not overwhelming.

While the ignition and burning characteristics of irradiated U are obscure, it is a reasonable assumption that the fire potential associated with molten U is substantially greater than with solid U. The primary reason for this belief being the ability of molten metal to assume a highly dangerous, finely divided, form under suitable impetus. Molten U, spilling from a UAl_3 /oxide container into air would probably ignite. It is somewhat doubtful that a fog spray could prevent ignition under these conditions.

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Quite arbitrarily, for this study, a normal slug is said to reach a fire sensitive state when the slug mean temperature (as interpreted by Zaloudek in Ref. 2) reaches the MP of U, i.e., the sensitive state is reached at the time ϕ_u (cf Table I). Abnormal slugs, bearing pyrophoric corrosion products, obviously can show fire action long before ϕ_u . The history of such an incident after exhaustion of the "sensitive" material is, however, difficult to predict. The closer the mean slug temperature is to the MP of U at the time of the "trigger" fire the greater is the probability that the main U mass could sustain the fire. Accordingly, the ϕ_u criterion for the fire sensitive state may also have some relevance for abnormal slugs. Any attempt to attach numbers to predictions of pyrophoric behavior is, however, a highly speculative business with normal, or abnormal slugs.

As noted previously, an attempt was made to estimate the effect of the U-air reaction on the slug temperature history (cf App. B). The "corrected" ϕ_u and ϕ_{melt} times, for 0 and 1 hour decay, are plotted in Figure 1. In Figure 2, the ϕ_u and ϕ_u^c data (cf Figure 1) are replotted for the 0 and 1 hour decay cases. The band between the ϕ_u and ϕ_u^c curves is arbitrarily designated as the time zone within which the slug might reach a fire sensitive state. Decreasing the slug irradiation exposure and increasing the decay time before discharge will displace the bands to the right on the time axis. It is felt that the assumptions involved in Figure 2 do not justify the work of preparing graphical or numerical aids for interpolation, or extrapolation, to other exposure and/or decay times.

FISSION PRODUCT RELEASE POSSIBILITIES

The principal hazard associated with a slug fire, or temperature excursion, lies in the possibilities for release of the fission product burden of the slug. The ultimate radiological consequences of such an incident are determined by the effectiveness of the various components of the confinement system in preventing release of the harmful radioactive products to the ex-105 Bldg. environment, and by the ability of this environment to dispose of the released material in a harmless fashion. The focus of attention in the present report is on the fission product release at the slug. As indicated above, this permits only a very limited, and necessarily pessimistic, view of the radiological hazard potential of single slug incidents.

R. K. Hilliard and D. L. Reid (Ref. 3) have estimated the % release of various biologically significant fission products on the basis of Zaloudek's (Ref. 2) temperature history data. These data were obtained from HLO studies of fission product release from metallic U. Ref. 3 should be consulted for details of the % release estimates. The % release estimates given here correspond to the time that the slug is completely melted. Melting time data were estimated from Zaloudek's data by the method of App. A. Hilliard and Reid also estimated the % release at the time the cladding starts to melt and at the time the U starts to melt. Because a slug residence time at least equal to ϕ_{melt} (cf Table I) is considered likely for the short decay cases, the release prior to ϕ_{melt} was not of primary interest here.

The absolute release data were obtained by applying the % release figure to the slug burden for the particular isotope. Slug burden data were obtained from Ref. 6. Generally the isotope burden has saturated prior to 1 year exposure. When this was not the case, the burden data were interpolated to obtain the 1 year data. The fission derivatives considered for the present study were: iodine, xenon, tellurium,

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FIGURE 2

Approximate Time to Reach Fire Sensitive State Following Discharge.

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ruthenium, strontium and cesium. The various isotopes taken into account in the release figures are listed in Table II. Only biologically significant isotopes having a half life greater than 1 hour were considered. It is felt that the uncertainties embodied in the % release figures--including the temperature history of the slug--do not justify a more comprehensive fission product and isotope accounting for this report.

The fission product release data are summarized in Table II. Three slug powers--52.3, 59.7 and 67.3 KW--were considered. Five decay times (time of discharge after reactor shut-down)--0, 1M, 10M, 60M and 120M--were considered for each slug power. Because of the interest in the 0 decay case, the fission product release data for the 0 decay case are plotted in Figure 3 as a function of slug power. Estimates of the time to melt the slug--the time corresponding to the release data--are also given in Figure 3. As noted in Table II and Figure 3, the release data are estimated on the basis of Zaloudek's data, i.e., release data are uncorrected for chemical heating effects. Although chemical heating appears to have considerable influence on the ϕ -T characteristic of the slug (cf Figure 1), there was felt to be little, or no, incentive for applying a chemical heating correction to the data of Table II. Work at HLO and elsewhere has shown that, in general, the fission product release while the uranium is in the molten state overshadows solid state release for comparable holding times in the two regimes. Hence, the temperature history of the slug after reaching the MP of U, rather than before, is of primary concern in fission product release estimates. As no attempt was made in the present study to predict the post-melting history of the U, a chemical heating correction to the data of Table II was not attempted, or regarded as important.

The kinetics of fission product release from metallic U, under conceivable accident conditions, poses a formidable physical-chemical problem. The ultimate disposition of released material depends on its physical and chemical state and, in addition, on multitudinous factors of the environment. Both the physical-chemical characterization of the fission products upon release from the slug and the fission product--environment interactions are under study at HLO and other laboratories. Advances in reactor hazard analyses will be largely determined by advances in these areas.

G. E. Zima

Process and Reactor Development Subsection
RESEARCH AND ENGINEERING SECTION

GEZ:kmm

TABLE II

N PRODUCTS AT TIME SLUG IS MELTED

	10			60			120		
	52.3	59.7	67.3	52.3	59.7	67.3	52.3	59.7	67.3
Burden (curies)									
IODINE									
70	1453	1659	1870	1409	1609	1813	1409	1609	1813
53	1673	1910	2153	1629	1860	2097	1629	1860	2097
60	3699	4222	4760	3611	4122	4647	3523	4021	4533
73	2422	2765	3117	2246	2564	2890	2026	2312	2607
56	9247	10556	11900	8895	10155	11447	8587	9802	11050
1	10.3	8.8	7.7	16.0	13.8	11.3	24.8	18.4	14.7
0	952	929	916	1423	1401	1294	2130	1804	1624
XENON									
60	3699	4222	4760	3699	4222	4760	3699	4222	4760
43	2598	2966	3343	2554	2915	3286	2554	2915	3286
03	6297	7188	8103	6253	7137	8046	6253	7137	8046
-----90%-----									
93	5667	6469	7293	5628	6423	7241	5628	6423	7244
TELLURIUM									
97	1629	1860	2097	1629	1860	2097	1629	1860	2097
78	216	246	278	211	241	278	207	236	278
08	84	96	108	84	96	108	84	96	108
61	436	498	561	423	483	544	392	447	504
17	13	15	17	13	15	17	13	15	17
08	84	96	108	84	96	108	84	96	108
69	2462	2811	3169	2444	2791	3152	2409	2750	3112
68	7.31	6.22	5.46	12.3	9.82	7.49	17.6	13.2	10.5
7	180	175	173	301	274	236	423	362	327

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	10	60	120
Burden (curles)	52.3	59.7	67.3
	52.3	59.7	67.3
	52.3	59.7	67.3

PERCENTUM.									
70	1453	1659	1870	1453	1659	1870	1453	1659	1820
67	436	498	561	410	468	527	361	412	465
36	106	121	136	106	121	136	106	121	136
73	1995	2278	2567	1969	2248	2533	1920	2192	2471
25	0.53	0.49	0.39	0.88	0.70	0.58	1.17	0.94	0.59
4	10.6	11.2	10.0	17.3	15.7	14.7	22.5	20.6	14.6
RONITIUM.									
93	1938	2212	2493	1938	2212	2493	1938	2212	2493
79	62	70	79	62	70	79	62	70	79
60	2334	2664	3030	2246	2564	2890	2114	2413	2728
30	2422	2765	3117	1938	2212	2493	1541	1759	1983
62	6756	7711	8719	6184	7058	7955	5655	6454	7275
42	.085	.074	.067	.143	.116	.092	.21	.15	.12
7	5.7	5.7	5.8	8.8	8.2	7.3	11.9	9.7	8.7
ESIUM.									
44	4.23	4.83	5.44	4.18	4.77	5.38	4.18	4.78	5.38
	52.8	60.3	68	52.8	60.3	68	52.8	60.3	68
4	57	65.1	73.4	57	65	73	57	65	73
19	5.93	5.02	4.37	9.83	7.85	5.81	14.3	10.6	8.44
6	3.4	3.3	3.7	5.6	5.1	4.2	8.1	6.9	6.2

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FIGURE 3

Estimated Single Slug Release of Various Fission Products
for 0 Decay Before Discharge.

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APPENDIX A

Estimation of Time to Melt Slug (ϕ_{melt})

For a unit length of slug, the rate of change of the "mean" slug temperature may be expressed:

$$\frac{dT}{d\phi} = \dot{T}_d = P_d / Mc_p$$

$$\text{or } P_d = \dot{T}_d Mc_p$$

where P_d is the net FP decay power absorbed by the slug/length

M is the mass/length

c_p is the specific heat of U near the MP of U

The time to melt the slug, $\Delta\phi_{\text{melt}}$, may be approximated by:

$$\Delta\phi_{\text{melt}} = M \Delta H_f / P_d = M \Delta H_f / \dot{T}_d Mc_p \quad (1)$$

$$\text{or } \Delta\phi_{\text{melt}} = \Delta H_f / \dot{T}_d c_p \quad (2)$$

$\dot{T}_d$ is taken as the slope of the T- ϕ characteristic given in Ref. 2 at the inception of U melting. ΔH_f and c_p values used are 12.6 cal/gm U and 0.0383 cal/gm U°C, respectively.

ϕ_{melt} (cf Table I) is then equal to: $\phi_u + \Delta\phi_{\text{melt}}$, where ϕ_u is taken from Ref. 2.

APPENDIX BEstimation of Effect of U-air (O_2) Reaction on Slug Temperature History

Let P_c = power absorbed by the slug/length from U- O_2 reaction

$$\text{Now } P_c = pk_o \Delta H_{ox} \alpha \exp(-E/RT) \quad (1)$$

Where p = perimeter of U core

ΔH_{ox} = heat of reaction/gm U

α = fraction of ΔH_{ox} absorbed by slug/length

and k_o , E and R have their usual significance in an Arrhenius type representation of the U- O_2 reaction temperature dependence. k_o is expressed as gms U reacting per unit area per unit time.

The net FP decay power absorbed per unit length of the slug, at any time is (cf App. A):

$$P_d = \dot{T}_d Mc_p \quad (2)$$

$$\text{therefore, } P_c/P_d = pk_o \Delta H_{ox} \alpha \exp(-E/RT) / (\dot{T}_d Mc_p) \quad (3)$$

$$\text{Now } p = 2\pi(r_i + r_o) \quad (4)$$

Where r_i and r_o are the inside and outside radii of the U core, respectively.

$$M = \text{mass of U/length} = A \ell \quad (5)$$

Where A is the sectional area of the U core. A may be expressed in terms of r_i and r_o as follows:

$$A = \pi(r_i + r_o)(r_o - r_i)$$

$$\text{or } A = p(r_o - r_i)/2 \quad (6)$$

Substituting in (5),

$$M = p \ell (r_o - r_i)/2 \quad (7)$$

Substituting (7) in (3),

$$P_c/P_d = 2k_o \Delta H_{ox} \alpha \exp(-E/RT) / (\dot{T}_d \ell c_p (r_o - r_i)) \quad (8)$$

APPENDIX B (CONT'D)

Now making the poor assumption that chemical reaction does not change the rate of heat dissipation as calculated by Zaloudek in Ref 2, i.e., heat dissipation rate is a function of T only:

$$\dot{T}_{c+d} = (P_d + P_c)/Mc_p = (1 + P_c/P_d)/(Mc_p/P_d) \quad (9)$$

where $\dot{T}_{c+d}$ is the instantaneous slope of the T- ϕ characteristic when both FP decay and chemical reaction are supplying power to the slug.

From Equation (2),

$$Mc_p/P_d = 1/\dot{T}_d \quad (2a)$$

Substituting in (9),

$$\dot{T}_{c+d} = \dot{T}_d(1 + P_c/P_d) \quad (10)$$

Substituting for P_c/P_d from (8):

$$\dot{T}_{c+d} = \dot{T}_d \left[1 + 2 k_o \Delta H_{ox} \alpha \exp(-E/RT) / (\dot{T}_d e c_d (r_o - r_i)) \right] \quad (11)$$

The second term in the bracket is thus the correction factor applied to $\dot{T}_d$ to approximate the "true" instantaneous slope, $\dot{T}_{c+d}$.

Starting at the time that the cladding is completely melted, $\dot{T}_d$ was determined from Figure 1, Ref. 2, and $\dot{T}_{c+d}$ calculated from equation (11), for 100°F (55.5°C) intervals up to the MP of U. Using the relationship,

$$\Delta\phi = 55.5^\circ\text{C}/\dot{T}_{c+d}$$

the time points determining the 100°F intervals were computed.

The 100°F intervals and the corresponding time points gave an approximation to the T- ϕ characteristic, corrected for chemical reaction. The time for the new T- ϕ characteristic to reach the MP of U is designated as ϕ_c^c (cf Figure 2 this report). Taking the slope of the new characteristic at the inception of the U melting, a corrected time for melting the slug was obtained, using the method of App. A. These data are designated as ϕ_{melt}^c and are given in Figure 2 of this report.

In applying (11), the following values as assumptions were used:

$$\alpha = 1 \text{ (100\% absorption of chemical heat)}$$

$$k_o = 3.2 \text{ gm U/cm}^2 \text{ sec. (from Ref. 4 data)}$$

$$E/R = 8.1 \times 10^3 \text{ (}^\circ\text{K)} \text{ (from Ref. 4 data)}$$

$$\Delta H_{ox} = 1.13 \text{ Kcal/gm U (from Ref. 4 data)}$$

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APPENDIX B (CONT'D)

(This heat release is for the reaction, $U + O_2 \rightarrow UO_2$, and is averaged between the MP of Al and the MP of U. The heat release for the reaction $U + 4/3 O_2 \rightarrow 1/3 U_3O_8$, averaged over the same temperature range, was calculated to be 1.19 Kcal/gm U-- which is not regarded as a significant deviation from the ΔH_{ox} value for the $U-O_2-UO_2$ reaction considering the uncertainty in the $U-O_2$ reaction kinetics.)

$$\rho = \rho(T) \text{ gms/cm}^3 \quad (\text{taken from Ref. 4})$$

$$c_p = c_p(T) \text{ Kcal/gm}^\circ\text{C} \quad (\text{taken from Ref. 4})$$

and $r_0 - r_1$ was taken as 1.12 cm