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COPY NO. - 67

DATE - October 18, 1961

Buildup of Fission Products in Reactor Fuel and Coolant with 'Unclad' Fuel Material

Wm. B. Cottrell and L. A. Mann *

Abstract

Concentrations of fission products in the fuel and in the coolant stream of a generalized reactor power plant employing unclad fuel material have been calculated using actual fission-product chains and wide ranges of assumed values of 1) escape rate from the fuel and 2) removal rate from the coolant stream. The calculations were made on an IBM 704 digital computer. Some typical results are displayed in the form of graphs to illustrate the effects of the variables. The complete tables of calculated data are available but are not included with the present report.

As data on escape rates from fuel and removal rates from coolant become available, the tables and graphs developed by this study can be used to estimate the fission-product concentration in the coolant stream, which would in turn furnish a basis for determining the degree of coolant purification required to maintain the circulating activity below a given level.

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*W. L. Albrecht of TVA participated in the formulation of this study before being subsequently reassigned.

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Introduction

This study is directed toward predicting the buildup of concentration of fission-product activity in a gas-cooled power reactor coolant stream as a function of time, fission-product escape rates and removal rate. For the purposes of this study, all removal processes including deposition, leakage, and purification have been considered together as the removal rate. A corollary study would be the determination of the distribution of deposited activity in a coolant system when sufficient data on deposition become available.

The problem of coolant activity is a general one for power reactors using either clad or coated solid fuels, since all power reactors depend on coolant flow to remove the heat of fission and decay and since the probability always exists that fission products will enter the coolant stream via imperfections in the fuel coating or cladding. The same theory and calculational methods used here could be applied to liquid-cooled reactors with appropriately chosen ranges of removal rate parameters.

The level of activity in the coolant gas will be important to the determination of the hazards of routine and accidental releases of the primary coolant. The cost and difficulty of maintaining the coolant system will depend largely on the locations and amounts of deposited activity, which this study does not attempt to analyze. In this study, the deposited activity is combined with that removed from the coolant by whatever cleaning methods are used (e.g., a filter in a bypass system) and the total removal rate thus obtained is designated by the symbol r (fraction removed per second). However, this does permit the determination of purification system requirements to obtain a cooling system concentration of activity less than any given value, if indeed a purification system is needed at all.

Fission-Product Escape from Fuel

The escape of fission products from a fuel element is taken here to be a diffusional process, first in diffusion to the surface of the particle or crystal in which the fission occurred, then out through the fuel element

matrix to its surface, where it is assumed that the atoms of fission products enter the flowing gas coolant stream through a defective or porous fuel container or coating. This analysis assumes that the diffusion of the fission products is controlled by the fuel particle and not by the processes by which it is subsequently released to the coolant stream. For an element consisting of UO_2 particles in a graphite matrix, the diffusion to the surface of the UO_2 particle is indeed controlling.[1] For other types of fuel element, diffusion through the matrix may be more significant or even controlling (for example, UO_2 particles in a stainless steel matrix coated in turn by a stainless steel sheath). Since the sizes and shapes of fuel particles and the escape routes from fuel particles to the fuel element surface vary to a degree that has so far prevented use of the actual diffusion coefficients (where known), experimental "virtual" diffusion coefficients (D') have been employed.[2,3] The theory of the escape process inside the fuel element is explained in detail by Eichenberg, et al.,[2] and is summarized by Cottrell, et al.[1] Alternatively, an isotope may form within the fuel element or the gas by decay of its parent or disappear by decay to its daughter. Decay chains of up to four generations are considered in this analysis.

This report is not intended to be specific as to what numerical values of D' and ν (escape parameters defined in the section on analysis) should be expected to be valid for isotope escape from fuel, but covers the range 10^{-14} to 10^{-7} fraction removed per second because some experiments [4,5] indicate that the value may eventually be found to be within that range at fuel element temperatures of most interest.

Fission-Product Fate in the Coolant

When a radioactive isotope enters the coolant stream it may decay, deposit on a container surface, leak out, be removed by a chemical or physical process (e.g., a filter), or remain suspended in the fluid stream. It is assumed here that no isotopes move from the fluid stream into the fuel.[6]

This report makes no attempt to specify the actual means of removal from the coolant but covers the range r = 0 to r = 1 of removal rates, where r = fraction removed per sec by all processes combined except radioactive

decay which is included separately and is considered invariant. On the other hand deposition, leakage and purification are or may be subject to control. Deposition on a container surface is controllable to some extent by design or material specification although there may be some practical limitations on this process. Normal leakage from the coolant system can be controlled by design and maintenance specifications, whereas removal by chemical or physical processing may be effected either in the main coolant stream or, more usually, in a bypass circuit. Some tests[7,8] indicate that deposition is sometimes much more dependent on chemisorption than on condensation or physical (e.g., van der Waals) adsorption. It is hoped that the range of the removal rate (r) selected for use in this study will be amenable to interpolation to experimental values. The removal rates used here represent the sums of the removal constants for leakage, deposition and purification.

Not only is it not now possible to separate deposited activity from that removed by other processes, but also the lack of either data or accepted theory prevents determination of the distribution of deposited radioactive isotopes in the coolant system. It has become evident[7,8] that deposition by condensation of fission products from the coolant caused by exceeding the saturation concentration in the coolant is frequently completely invalid as a basis for predicting removal rates or deposition locations. Study and testing of other factors (such as chemisorption, van der Waals adsorption, etc.) which influence the removal of fission products is at an early stage of development.

Range of Parameters Examined

The basic parameters considered in this study are 1) those affecting rates of diffusion from the fuel elements (D'), and 2) the assumed removal rates (r) from the coolant, and 3) operating time (t).

D' (sec^{-1}) may be defined as a constant times the amount of an isotope diffusing out per second evaluated at the surface of the fuel particle. It arises from a theoretical derivation[2] as an experimentally determinable value for a particular set of conditions. $D' = D/a^2$ (sec^{-1}), where D is the conventional diffusion coefficient (cm^2/sec) and a is a characteristic length[2] associated with the fuel (cm), such as the radius in the case of a

spherical fuel particle.

Three "cases" were included in the calculations of both the elements and the isotopes, as follows.

Case I. In the Case I calculations, the value of D' ($=D'_a$) used is the same for every element, over the range of values $D' = 10^{-12}$ to $D' = 10^{-7}$ sec^{-1}.

Case II. In the Case II calculations the value of D' ($= D'_b$) used for elements of atomic numbers 35-37 and 51-55 inclusive is 10^{-1} times the value of D' ($= D'_a$) for the other elements in Table I over the range $D' = 10^{-13}$ to $D' = 10^{-7}$ sec^{-1}. The reason is explained below.

Case III. In the Case III calculations the value of D' ($= D'_c$) used for elements of atomic numbers 35-37 and 51-55 inclusive is 10^{-2} times the value of D' ($= D'_a$) for the other elements in Table I. The reason is explained below. The total range of D' covered for these elements is thus 10^{-14} to 10^{-7} sec^{-1}.

Reason for Cases II and III. Cottrell, et al.,[1] reported that experimental data show that the D' values for some elements are significantly different from others at the same temperatures (pp 76-81, ORNL-2935). The evidence is not clear as to the quantitative relations, but it appears that factors of difference of at least up to 100 are involved. Therefore, this analysis assumed the factors of difference listed above for the Case II and Case III calculations. It should be emphasized that the selections are illustrative, not quantitative, and that experiments may show the factors of difference to be off by orders of magnitude. As experimental values accumulate, values within the ranges selected may be used for interpolation, while values outside the selected ranges may be used as input data for new calculations.

The parameter r (sec^{-1}) is the fractional rate of disappearance from the coolant stream, that is, it is the fraction of the total inventory of the isotope in the coolant which disappears from the coolant per second. This parameter includes loss by leakage, deposition, and purification. Values of r selected for this study range all the way from zero to one, and were chosen with the intent of permitting interpolation between the values selected. The same values of r were used in each of the three cases studied.

The parameter of time (t) is given in seconds after reactor startup. It is assumed that startup is instantaneous and that operation is steady state to the time in question. The range of values of t used was from 10^5 seconds (approximately 28 hours) to 10^8 seconds (approximately 3.2 years), to cover the range from reasonably short operating times to times long enough to be at or near equilibrium concentrations of most fission products.

Mathematical Analysis

Using the basic parameters defined above and the basic radioactive decay parameters, the generalized system was analyzed mathematically by J. Replogle of Central Data Processing, ORGDP. Calculations were performed on an IBM 704 digital computer using a program written by J. L. Lucius of Central Data Processing. The analysis follows.

Only the parameters used throughout the analysis are defined in the Nomenclature, page 17. Other constants and parameters are defined as needed. The analysis is for a four-membered decay chain and results in equations defining the existing amount of each isotope as a function of time in both the fuel and the gas coolant. The calculated amounts were printed out as the number of atoms of isotope per fission per second in the fuel elements leaking to the coolant.

I. Summary of the Differential Equations for a Four-Membered Chain

A. Fission product in the fuel.

$$\frac{dN_a^{\,f}}{dt} = \delta\, y_a - N_a^{\,f}\,(\lambda_a + \nu_a)$$

$$\frac{dN_b^{\,f}}{dt} = y_b + N_a^{\,f}\,\lambda_a - N_b^{\,f}\,(\lambda_b + \nu_b)$$

$$\frac{dN_c^{\,f}}{dt} = y_c + N_b^{\,f}\lambda_b - N_c^{\,f}\,(\lambda_c + \nu_c)$$

$$\frac{dN_d^{\,f}}{dt} = \frac{\lambda_c\, N_c^{\,f}}{1 + F} - \left(\frac{F'}{1 + F}\right) N_d^{\,f}$$

where,

$F = F(t)$ is the fraction of an isotope in the gas, of the total in both the gas and the fuel at the time under consideration[9]

The solution to this system is;

$$N_a^f = \frac{\delta y_a}{A_1} (1 - e^{-A_1 t})$$

$$N_b^f = A_4 + (A_5 - A_4) e^{-A_2 t} - A_5 e^{-A_1 t}$$

$$N_c^f = A_9 + A_{12} e^{-A_6 t} + A_{10} e^{-A_2 t} - A_{11} e^{-A_1 t}$$

$$N_d^f = \frac{\lambda_c}{1 + F(t)} \left[A_9 t + \frac{A_{12}}{A_6} (1 - e^{-A_6 t}) + \frac{A_{10}}{A_2} (1 - e^{-A_2 t}) - \frac{A_{11}}{A_1} (1 - e^{-A_1 t}) \right]$$

where

$$A_1 = \lambda_a + \nu_a$$

$$A_2 = \lambda_b + \nu_b$$

$$A_3 = \delta \lambda_a y_a / A_1$$

$$A_4 = (y_b + A_3)/A_2$$

$$A_5 = A_3/(A_2 - A_1)$$

$$A_6 = \lambda_c + \nu_c$$

$$A_7 = A_4 \lambda_b$$

$$A_8 = A_5 \lambda_b$$

$$A_9 = (A_7 + y_c)/A_6$$

$$A_{10} = (A_8 - A_7)/(A_6 - A_2)$$

$$A_{11} = A_8/(A_6 - A_1)$$

$$A_{12} = A_{11} - A_9 - A_{10}$$

B. Fission Products in the gas (coolant).

$$\frac{d\,N_a^{\,g}}{dt} = N_a^{\,f}\,\nu_a - N_a^{\,g}\,(\lambda_a + r)$$

$$\frac{d\,N_b^{\,g}}{dt} = N_b^{\,f}\,\nu_b + N_a^{\,g}\,\lambda_a - N_b^{\,g}\,(\lambda_b + r)$$

$$\frac{d\,N_c^{\,g}}{dt} = N_c^{\,f}\,\nu_c + N_b^{\,g}\,\lambda_b - N_c^{\,g}\,(\lambda_c + r)$$

$$\frac{d\,N_{d2}^{\,g}}{dt} = \lambda_c\,N_c^{\,g} - r\,(F\,N_d^{\,f} + N_{d2}^{\,g})$$

where

$N_d^{\,g} = N_{d1}^{\,g} + N_{d2}^{\,g}$, atoms fission^{-1} sec^{-1}

$N_{d1}^{\,g}$ = amount in the gas which had diffused out from the fuel

$N_{d2}^{\,g}$ = amount in the gas which appeared from decay of its parent which was already in the gas

The solution to this system is:

$$N_a^{\,g} = A_{15} + A_{17}\,e^{-A_{13}t} - A_{16}\,e^{-A_1 t}$$

$$N_b^{\,g} = A_{25}\,e^{-A_{18}t} + A_{21}\,e^{-A_2 t} - A_{22}\,e^{-A_1 t} + A_{23}\,e^{-A_{13}t} + A_{24}$$

$$N_c^{\,g} = A_{36}\,e^{-A_{26}t} + A_{30}\,e^{-A_2 t} - A_{31}\,e^{-A_1 t} + A_{32}\,e^{-A_6 t} + A_{33}\,e^{-A_{18}t} + A_{34}\,e^{-A_{13}t} + A_{35}$$

$$N_d^{\,g} = F\,N_d^{\,f} + N_{d2}^{\,g}$$

where

$$N_{d2}^{g} = M(t) - r\, e^{-rt} \int_{o}^{t} e^{r\tau}\, N_d^{f}(\tau)\, F(\tau)\, d$$

and

$$M(t) = \lambda_c \Big[\frac{A_{35}}{r} + \frac{A_{36}\, e^{-A_{26}t}}{r - A_{26}} + \frac{A_{30}\, e^{-A_2 t}}{r - A_2} - \frac{A_{31}\, e^{-A_1 t}}{r - A_1} + \frac{A_{32}\, e^{-A_6 t}}{r - A_6} + \frac{A_{33}\, e^{-A_{18}t}}{r - A_{18}} + \frac{A_{34}\, e^{-A_{13}t}}{r - A_{13}} - e^{-rt} \Big(\frac{A_{35}}{r} + \frac{A_{36}}{r - A_{26}} + \frac{A_{30}}{r - A_2} - \frac{A_{31}}{r - A_1} + \frac{A_{32}}{r - A_6} + \frac{A_{33}}{r - A_{18}} + \frac{A_{34}}{r - A_{13}} \Big) \Big]$$

and

r = fractional rate of disappearance from suspension in gas (assumed constant), sec^{-1}

$A_{13} = \lambda_a + r$

$A_{14} = (\delta y_a\, \nu_a)/A_1$

$A_{15} = A_{14}/A_{13}$

$A_{16} = A_{14}/(A_{13} - A_1)$

$A_{17} = A_{16} - A_{15}$

$A_{18} = \lambda_b + r$

$A_{19} = \nu_b\, (A_5 - A_4)$

$A_{20} = \nu_b\, A_5 + \lambda_a\, A_{16}$

$A_{21} = A_{19}/(A_{18} - A_2)$

$A_{22} = A_{20}/(A_{18} - A_1)$

$A_{23} = (\lambda_a\, A_{17})/(A_{18} - A_{13})$

$A_{24} = (\nu_b\, A_4 + \lambda_a\, A_{15})/A_{18}$

$A_{25} = A_{22} - A_{21} - A_{24} - A_{23}$

$$A_{26} = \lambda_c + r$$

$$A_{27} = \nu_c A_{10} + \lambda_b A_{21}$$

$$A_{28} = \nu_c A_{11} + \lambda_b A_{22}$$

$$A_{29} = \nu_c A_9 + \lambda_b A_{24}$$

$$A_{30} = A_{27}/(A_{26} - A_2)$$

$$A_{31} = A_{28} (A_{26} - A_1)$$

$$A_{32} = (\nu_c A_{12})/(A_{26} - A_6)$$

$$A_{33} = (\lambda_b A_{25})/(A_{26} - A_{18})$$

$$A_{34} = (\lambda_b A_{23})/(A_{26} - A_{13})$$

$$A_{35} = A_{29}/A_{26}$$

$$A_{36} = A_{31} - A_{30} - A_{32} - A_{33} - A_{34} - A_{35}$$

II. Derivation of the Solutions to the Differential Equations

The first three equations of a four-membered chain are solved by LaPlace transforms. The last equation for the fuel can be written as:

$$\frac{dx}{dt} + A(t)\, x = B(t)$$

Whose solution is:

$$x = e^{-\int A dt} \int \left(e^{\int A dt}\right) B dt + C\, e^{-\int A dt}$$

where

C = constant to be valuated from the condition x (0) = 0

The differential equation for $N_{d2}{}^g$ is solved by transposing the term $r\, N_{d2}{}^g$ to the left side of the equation and applying the operator $\int_0^t e^{r\tau}\, d\tau$ to both sides of the equation. However, this leaves the integral $\int_0^t e^{r\tau}\, N_d{}^f(\tau)\, F(\tau)\, d\tau$ to be evaluated numerically, and considerable care must be taken to avoid round-off errors in the

machine calculations. The form of the solution that was found to be tractable in all but a few cases is as follows:

$$N_d^g = \frac{F(t)}{1 + F(t)} K(t) + M(t) + r e^{-rt} \int_0^t \left[\frac{F(t)}{1 + F(t)} - \frac{F(\tau)}{1 + F(\tau)} \right] e^{r\tau} P(\tau) \, d\tau$$

where

$$P(t) = [1 + F(t)] \quad N_d^f$$

$$K(t) = e^{-rt} \int_0^t e^{r\tau} P'(\tau) \, d\tau$$

and M (t) has been defined previously. The latter functions are easily evaluated as they are linear in t and exponentials of t.

The integral was evaluated by the generalized mean value theorem by estimating $\bar{\tau}_j$ in each of 2^k equal subintervals. The integral is then:

$$\sum_{j=1}^{2^k} \left[\frac{F(t)}{1 + F(t)} - \frac{F(\bar{\tau}j)}{1 + F(\bar{\tau})} \right] \int_{t_j=1}^{t_j} e^{r\tau} P(\tau) \, d\tau$$

with

$$t_0 = 0$$

and

$$t_{(2^k)} = t$$

III. Solution of a Chain of Three Members or Less.

Each member of the chain is solved according to its characteristic description in the chain except for isotopes with $\lambda < 10^{-11}$. In this case the equation is solved as if the isotope were stable as represented by the equations for N_d^f and N_d^g in Part I, with N_c^f and N_c^g being replaced by the next higher element in the chain.

Nomenclature for Mathematical Analysis

F	=	$N^g/(N^g + N^b)$
N	=	number of atoms in fuel (or gas) at time t after startup
r	=	fractional rate of disappearance from suspension in gas (assumed constant), sec^{-1}
y	=	fission yield, atoms of species appearing per fission, $fiss^{-1}$
δ	=	fraction of y which is isomer being calculated
λ	=	fractional decay rate, sec^{-1}
ν	=	fractional rate of disappearance from the fuel matrix (assumed constant), $\nu = 3\sqrt{\lambda D'}$ (see ref. 2, Eg. V-10)

Superscripts

f	=	fuel
g	=	gas

Subscripts

a,b,c,d	=	member of decay chain $a \rightarrow b \rightarrow c \rightarrow d$

Results

As stated above, the calculations in this study apply only to those fuel elements which offer no resistance to the escape of fission products from the fuel material surface to the coolant gas. The tabulated results have the units "total number of atoms per fission per second" for concentrations in both uncontained fuel elements and in the coolant gas at the listed values of D', r, and time of continuous steady-state operation since reactor startup.

Results were calculated for the elements listed in Table 1 and for the isotopes and isomers listed in Appendix A. Nuclides of the elements in Table 1 with atomic mass numbers 85 through 106 and 127 through 151 were included in the calculations. This includes all isotopes of the light (groups A) elements form rubidium through molybdenum, and of the heavier (group B) elements form iodine through promethium, plus the more significant isotopes of the other fission-product elements.

The actual concentrations may be obtained from the relations:

$$C_i^{\,f} = \frac{P\,N_i^{\,f}}{v^f}$$

and

$$C_i^{\,g} = \frac{P\,N_i^{\,g}}{v^g}$$

with the nomenclature:

C_i = concentration of nuclide i, atoms cm^{-3}

P = fission power generated in the leaking fuel elements, fissions sec^{-1}

N_i = total atoms of nuclide i in the medium specified, atoms $fission^{-1}$ sec^{-1}

v = volume of the medium specified, cm^3

Table 1. Fission-Product Elements Included in this Analysis

Group A (Lighter Elements)		Group B (Heavier Elements)	
Element	Atomic No.	Element	Atomic No.
Br	35	Sb	51
Kr	36	Te	52
Rb	37	I	53
Sr	38	Xe	54
Y	39	Cs	55
Zr	40	Ba	56
Nb	41	La	57
Mo	42	Ce	58
Tc	43	Pr	59
Ru	44	Nd	60
Rh	45	Pm	61
Pd	46	Sm	62
		Eu	63

In as much as it was not practical to include all the data for all isotopes for all the cases, 12 isotopes were selected from Appendix A for illustrative treatment. They were chosen roughly on the basis of combinations of fission yield, decay constant, beta or gamma energy, and biological significance.[10] They are not intended to be inclusive, but they were chosen only as significant examples to illustrate the effects of the parameters time, D', and r, on the hazards and therefore on the design specifications of fuel and coolant systems. The 12 isotopes selected were Kr^{87}, Sr^{89}, Sr^{90}, Y^{90}, I^{131}, I^{134}, I^{135}, Cs^{138}, Ba^{140}, La^{140}, Ce^{144}, and Pr^{144}.

Some of the effects of the parameters of time, D', and r, on the concentrations of the 12 isotopes chosen are shown in Figs. 1 through 36 which are presented for convenience at the end of the report. Included are graphs of N^g (total atoms in the gas/fiss/sec) for Case I for all 12 isotopes and of N^g for Cases II and III for the long-lived (28-yr) Sr^{90} and the relatively short-lived La^{140} (whose N^g depends essentially on the decay of 12.8 day Ba^{140}). For the time scale used, concentrations of the relative short-lived isotopes which are not daughters of long-lived precursors appear as essentially straight horizontal lines. This would naturally be expected, because of the more rapid approach to equilibrium concentration as the half-life becomes shorter.

In addition to the results shown in Figures 1-36, selected isotopes have been analyzed on one of two bases, 1) those which constitute the more serious, potential inhalation exposures, and 2) those which constitute the more serious direct radiation potential. The selected isotopes in each group are tabulated in Table 2 and 4 respectively, together with nuclear or biological constants which describe the relative potential of each. Table 3 then lists the concentration of isotope given in Table 2 for a number of values of D' and r, together, in all instances, with the index of the inhalation hazard potential. Likewise, Table 5 lists the concentration of the isotopes given in Table 4 for a number of conditions of D' and r, together in all instances, with an index of the direct radiation hazard potential.

Table 2. Selected Constants of Significant Isotopes for Potential Internal Exposure

Isotope	λ (sec^{-1})	D[b] (mrem/μc)	λD (mrem/sec-μc)	$TID^{(c)}/N^{g}$ (= $8.38 \times 10^{10} \times D$)[a] [(rem/100 Mwt) (atoms)/(fiss/sec)]
Sr^{89}	1.48×10^{-7}	416	6.16×10^{-5}	5.16×10^{6}
Sr^{90}-Y^{90}	7.85×10^{-10}	44,500	3.49×10^{-5}	2.92×10^{6}
I^{131}	9.96×10^{-7}	1570	1.56×10^{-3}	1.3×10^{8}
I^{134}	2.20×10^{-4}	25	5.50×10^{-3}	4.61×10^{8}
I^{135}	2.89×10^{-5}	124	3.58×10^{-3}	3.00×10^{8}
Ba^{140} - La^{140}	6.27×10^{-7}	90	5.64×10^{-5}	4.73×10^{6}
Ce^{144} - Pr^{144}	2.76×10^{-8}	1210	3.34×10^{-5}	2.80×10^{6}

[a]Taken at 10^{8} sec after startup with steady operation at 100 Mwt.

[b]This D is a factor to convert μc to mrem TID. It is not a diffusion parameter. The values of D have been corrected from ref. 10 by using data in the ICRP Committee II Report (1959).

[c]TID for inhalation of all the coolant (to be multiplied by the fraction of fuel elements leaking and by the fraction of leaked atoms inhaled).

Table 3. Gas Concentrations and Relative Internal Hazard Potential of Selected Isotopes for various values of D' and r

Isotope	N^g (atoms / Fiss/sec)	TID (rems / 100 Mwt)	N^g	TID	N^g	TID	N^g	TID
	$D' = D'_a = D'_b = 10^{-7}$; $t = 10^8$ sec							
	$r = 0$		$r = 10^{-6}$		$r = 10^{-4}$		$r = 10^{-2}$	
Sr^{89}	2.5×10^5	1.3×10^{12}	3.1×10^4	1.6×10^{11}	3.4×10^2	1.8×10^9	3.3×10^0	1.7×10^7
Sr^{90}-Y^{90}	4.0×10^7	1.2×10^{14}	5.3×10^4	1.5×10^{11}	5.3×10^2	1.5×10^9	5.3×10^0	1.5×10^7
I^{131}	1.5×10^4	2.0×10^{12}	7.5×10^3	9.8×10^{11}	1.2×10^2	1.6×10^{10}	1.3×10^0	1.7×10^8
I^{134}	4.0×10^1	1.8×10^{10}	3.7×10^1	1.7×10^{10}	2.2×10^1	1.0×10^{10}	4.3×10^{-1}	2.0×10^8
I^{135}	3.0×10^2	9.0×10^{10}	3.0×10^2	9.0×10^{10}	6.9×10^1	2.1×10^{10}	8.8×10^{-1}	2.6×10^8
Ba^{140} - La^{140}	6.0×10^4	2.8×10^{11}	2.1×10^4	9.9×10^{10}	3.4×10^2	1.6×10^9	3.4×10^0	1.6×10^7
Ce^{144} - Pr^{144}	1.0×10^6	2.8×10^{12}	5.1×10^4	1.4×10^2	5.2×10^2	1.5×10^9	5.2×10^0	1.5×10^7
	$D' = D_a' = D_b' = 10^{-12}$; $t = 10^8$ sec							
Sr^{89}	2.6×10^3	1.3×10^{10}	3.3×10^2	1.7×10^9	3.8×10^0	2.0×10^7	3.7×10^{-2}	1.9×10^{-5}
Sr^{90}-Y^{90}	2.4×10^4	7.0×10^{10}	4.7×10^2	1.4×10^9	4.8×10^0	1.4×10^7	4.8×10^{-2}	1.4×10^5
I^{131}	9.3×10^1	1.2×10^{10}	4.6×10^1	6.0×10^9	9.0×10^{-1}	1.2×10^8	8.8×10^{-3}	1.2×10^6
I^{134}	1.3×10^{-1}	6.0×10^7	1.3×10^{-1}	6.0×10^7	7.8×10^{-2}	3.6×10^7	1.5×10^{-3}	6.9×10^5
I^{135}	1.1×10^0	3.3×10^8	1.1×10^0	3.3×10^8	2.6×10^{-1}	7.8×10^7	3.3×10^{-3}	9.9×10^5
Ba^{140} - La^{140}	3.8×10^2	1.8×10^9	1.5×10^2	7.1×10^8	2.4×10^0	1.1×10^6	2.4×10^{-2}	1.1×10^5
Ce^{144} - Pr^{144}	3.0×10^4	8.4×10^{10}	9.9×10^2	2.8×10^9	1.0×10^1	2.8×10^7	1.0×10^{-1}	2.8×10^5

Table 3. (continued)

Isotope	$D'_a = 10^{-7}$; $D'_b = 10^{-9}$; $t = 10^8$ sec							
	$r = 0$		$r = 10^{-6}$		$r = 10^{-4}$		$r = 10^{-2}$	
	N^g	TID	N^g	TID	N^g	TID	N^g	TID
Sr^{89}	7.7×10^4	4.0×10^{11}	9.9×10^3	5.1×10^{10}	1.1×10^2	5.7×10^8	9.2×10^{-1}	4.7×10^6
Sr^{90}-Y^{90}	7.5×10^5	2.2×10^{12}	1.4×10^4	4.1×10^{10}	1.4×10^2	4.1×10^8	1.3×10^0	3.8×10^6
Ba^{140}-La^{140}	1.2×10^4	5.7×10^{10}	4.4×10^3	2.1×10^{10}	7.2×10^1	3.4×10^8	7.0×10^{-1}	3.3×10^6
Ce^{144}-Pr^{144}	6.8×10^5	1.9×10^{12}	2.1×10^4	5.9×10^{10}	2.2×10^2	6.2×10^8	2.2×10^0	6.2×10^6
	$D'_a = 10^{-12}$; $D'_b = 10^{-14}$; $t = 10^8$ sec							
Sr^{89}	3.0×10^2	1.5×10^9	3.9×10^1	2.0×10^8	4.4×10^{-1}	2.3×10^6	3.8×10^{-3}	2.0×10^4
Sr^{90}-Y^{90}	2.6×10^3	7.6×10^9	5.0×10^1	1.5×10^8	5.0×10^{-1}	1.5×10^6	4.8×10^{-3}	1.4×10^4
Ba^{140}-La^{140}	4.1×10^1	1.9×10^8	1.6×10^1	7.6×10^7	2.5×10^{-1}	1.2×10^6	2.5×10^{-3}	1.2×10^4
Ce^{144}-Pr^{144}	3.0×10^3	8.4×10^9	1.0×10^2	2.8×10^8	1.0×10^0	2.8×10^6	1.0×10^{-2}	2.8×10^4

Table 4. Selected Constants of Isotopes Selected for Direct Radiation Exposure Potential

Isotope	λ (sec^{-1})	Total[b] E_γ (Mev/dis)	Activity[c]/100 Mwt N^g (= 3.1×10^{18} $\lambda E\gamma$)[a] [Mev/(sec-100 Mwt)]/[atoms/(fiss/sec)]
Kr^{87}	1.48×10^{-4}	0.56	2.6×10^{14}
I^{131}	9.96×10^{-7}	0.39	1.2×10^{12}
I^{134}	2.20×10^{-4}	1.27	8.7×10^{14}
I^{135}	2.89×10^{-5}	1.54	1.4×10^{14}
Cs^{138}	3.62×10^{-4}	2.01	2.3×10^{15}
Ba^{140}	6.27×10^{-7}	0.237	4.6×10^{11}
La^{140}	4.79×10^{-6}	2.11	3.1×10^{13}
Ce^{144}	2.76×10^{-8}	0.43	3.7×10^{10}
Pr^{144}	6.60×10^{-4}	0.08	1.6×10^{14}

[a]Calculated for 10^8 sec after startup; steady-operation at 100 Mwt.

[b]For greater refinement of accuracy, use E_γ separately for each energy group and multiply N^g by fraction in the energy group, then sum the 3.1×10^{18} N^g $\Sigma E_i \lambda_i$ fraction, since the hazard is high for harder gammas for the same Mev/sec.

[c]Total activity is all the coolant. The concentration and geometry will determine the hazards. Also see note b.

Table 5. Gas Concentrations and Relative Direct Radiation Hazard Potential of Selected Isotopes for Various Values of D' and r

$D' = D_a' = D_b' = 10^{-7}$; $t = 10^8$ sec

Case I	r = 0		r = 10^{-6}		r = 10^{-4}		r = 10^{-2}	
	N^g atoms/(fiss/sec)	Activity (Mev/sec)/100 Mwt	N^g	Activity	N^g	Activity	N^g	Activity
Kr^{87}	1.4×10^1	3.6×10^{15}	1.4×10^1	3.6×10^{15}	8.5×10^0	2.2×10^{15}	2.0×10^{-1}	5.2×10^{13}
I^{131}	1.5×10^4	1.8×10^{16}	7.5×10^3	9.0×10^{15}	1.2×10^2	1.4×10^{14}	1.3×10^0	1.6×10^{12}
I^{134}	4.0×10^1	3.5×10^{16}	3.7×10^1	3.2×10^{16}	2.2×10^1	1.9×10^{12}	4.3×10^{-1}	3.7×10^{14}
I^{135}	3.0×10^2	4.2×10^{16}	3.0×10^2	4.2×10^{16}	6.9×10^1	9.7×10^{15}	8.8×10^{-1}	1.2×10^{14}
Cs^{138}	1.3×10^1	3.0×10^{16}	1.3×10^1	3.0×10^{16}	9.6×10^0	2.2×10^{16}	2.7×10^{-1}	6.2×10^{12}
Ba^{140}	6.0×10^4	2.8×10^{16}	2.1×10^4	9.7×10^{15}	3.4×10^2	1.6×10^{14}	3.4×10^0	1.6×10^{12}
La^{140}	9.0×10^3	2.8×10^{17}	3.8×10^3	1.2×10^{17}	8.4×10^1	8.4×10^{15}	8.6×10^{-1}	2.7×10^{13}
Ce^{144}	1.0×10^2	3.7×10^{16}	5.1×10^4	1.9×10^{15}	5.2×10^2	1.9×10^{13}	5.2×10^0	1.9×10^{11}
Pr^{144}	7.3×10^1	1.2×10^{16}	2.6×10^0	4.2×10^{14}	4.4×10^{-1}	7.0×10^{13}	3.4×10^{-2}	4.8×10^{12}
	$D' = D_a = D_b' = 10^{-12}$; $t = 10^8$ sec							
Kr^{87}	4.9×10^{-2}	1.3×10^{13}	4.9×10^{-2}	4.9×10^{13}	2.9×10^{-2}	7.5×10^{12}	6.8×10^{-4}	1.8×10^{11}
I^{131}	9.3×10^1	1.1×10^{13}	4.6×10^1	5.5×10^{13}	9.0×10^{-1}	1.1×10^{12}	8.8×10^{-3}	1.1×10^{10}
I^{134}	1.3×10^{-1}	1.1×10^{14}	1.3×10^{-1}	1.1×10^{14}	7.8×10^{-2}	6.8×10^{13}	1.5×10^{-3}	1.3×10^{12}
I^{135}	1.1×10^0	1.5×10^{14}	1.1×10^0	1.5×10^{15}	2.6×10^{-1}	3.6×10^{13}	3.3×10^{-3}	4.6×10^{11}
Cs^{138}	4.4×10^{-2}	1.0×10^{14}	4.3×10^{-2}	9.9×10^{13}	3.2×10^{-2}	7.4×10^{13}	9.2×10^{-4}	2.1×10^{12}
Ba^{140}	3.8×10^2	1.7×10^{14}	1.5×10^2	6.9×10^{13}	2.4×10^0	1.1×10^{12}	2.4×10^{-2}	1.1×10^{10}
La^{140}	6.8×10^1	2.1×10^{15}	3.1×10^1	9.6×10^{14}	8.3×10^{-1}	2.6×10^{13}	8.9×10^{-3}	2.8×10^{11}
Ce^{144}	3.0×10^4	1.1×10^{15}	9.9×10^2	3.7×10^{13}	1.0×10^1	3.7×10^{11}	1.0×10^{-1}	3.7×10^9
Pr^{144}	1.3×10^0	2.1×10^{14}	5.1×10^{-2}	8.2×10^{12}	9.0×10^{-3}	1.4×10^{12}	1.2×10^{-4}	9.9×10^{10}

Table 5. (continued)

$D'_a = 10^{-7}$; $D'_b = 10^{-9}$; $t = 10^{8}$ sec

Case III	r = 0		r = 10^{-6}		r = 10^{-4}		r = 10^{-2}	
	N^g atoms/(fiss/sec)	Activity (Mev/sec)/100 Mwt	N^g	Activity	N^g	Activity	N^g	Activity
Ba^{140}	1.2×10^{4}	5.5×10^{15}	4.4×10^{3}	2.0×10^{15}	7.2×10^{1}	3.3×10^{13}	7.0×10^{-1}	3.2×10^{11}
La^{140}	2.0×10^{3}	6.2×10^{16}	8.8×10^{2}	2.7×10^{16}	2.3×10^{1}	7.1×10^{14}	2.3×10^{-1}	7.1×10^{12}
Ce^{144}	6.8×10^{5}	2.5×10^{16}	2.1×10^{4}	7.8×10^{14}	2.2×10^{2}	8.1×10^{12}	2.2×10^{0}	8.1×10^{10}
Pr^{144}	2.9×10^{1}	4.6×10^{15}	1.1×10^{0}	1.8×10^{14}	1.9×10^{-1}	3.0×10^{13}	1.3×10^{-2}	2.1×10^{12}

$D'_a = 10^{-12}$; $D'_b = 10^{-14}$; $t = 10^{8}$ sec

Ba^{140}	4.1×10^{1}	1.9×10^{13}	1.6×10^{1}	7.4×10^{12}	2.5×10^{-1}	1.2×10^{11}	2.5×10^{-3}	1.2×10^{9}
La^{140}	7.1×10^{0}	2.2×10^{14}	3.2×10^{0}	9.9×10^{14}	8.4×10^{-2}	2.6×10^{12}	8.6×10^{-4}	2.7×10^{10}
Ce^{144}	3.0×10^{3}	1.1×10^{14}	1.0×10^{2}	3.7×10^{12}	1.0×10^{0}	3.7×10^{10}	1.0×10^{-2}	3.7×10^{8}
Pr^{144}	1.3×10^{-1}	2.1×10^{13}	5.2×10^{-3}	8.3×10^{11}	9.1×10^{-4}	1.5×10^{11}	6.3×10^{-5}	1.0×10^{10}

Discussion

The concentration of radioactivity in the coolant may be important to the operation and maintenance of the system because of the hazards of direct radiation from a nonleaking cooling system, or of both direct radiation and inhalation of radioactivity from a leaking coolant system. The results of this study can be analyzed to show the relative effects on concentrations in the coolant of 1) varying the resistance to diffusion from the fuel as compared with 2) varying the rate of purification of the coolant. These effects can each be controlled over wide ranges by design specifications of the fuel elements and the coolant system. The leakage rate from the coolant system can also be controlled by design, operations, and maintenance specifications and administration. Thus, a balance can be made to obtain an optimum combination of fuel and coolant system designs, with the accuracy of the optimization bounded only by the accuracy of the parameter data.

As a basis for an illustrative partial analysis of the results, an operating time of 10^8 sec (3.2 yr) was chosen to include near-equilibrium concentrations of the shorter half-life isotopes and significant concentrations of the longer half-life materials. A power of 100 Mwt was chosen as representative of the power range of many power reactors and because the results are easily convertible to other powers by a simple factor. Tables 3 and 5 present potential activities in the coolant of 12 representative nuclides selected for comparisons.

Inhalation Exposures

For each isotope the potential inhalation dose was calculated for four values of r at each of two values of D_a' for both cases I and III. The results are presented in Table 2. Mathematically the calculation is:

$$TID = PN^g \lambda FD$$

with these definitions:

TID = total integrated dose for inhalation of all the coolant, rem.

P = power generated by all the fuel elements combined

N^g = total number atoms in the coolant gas, atoms/(fiss/sec)

λ = fraction decaying per second, sec^{-1}

F = factor to convert $PN^g\lambda$ to $\mu c \times 10^{-3}$, $\mu c/(dis/sec)$. Value of $F = 1/3.7 \times 10^7$

D = Burnett's dose parameter,[10] $mrem/\mu c$

The values of N^g were obtained from the digital computer results. Values of D* were obtained by recalculating Burnett's 1957 values[10] of $D_i = 73{,}800\ F_{a,i}\ E_i\ (RBE)\ NT/m_i$ with the new values of the components reported in 1959 by ICRP Committee II.

The equation for the potential dose at a reactor power of 100 Mwt with the above definitions is;

$$TID = 8.38 \times 10^{10} \sum_i N_i^{\ g}\ {}_iD_i = rem/(100\ Mwt)\ at\ 10^8\ sec$$

To obtain the TID to a person, the TID tabulated is multiplied by the ratio of the reactor power to 100 Mwt times the fraction of all the coolant gas which the person breathes (total coolant gas breathed per total coolant gas).

Direct Radiation

For each isotope the total gamma activity was calculated for four values of r at each of two values of D_a' for Cases I and III. The results are presented in Table 5. Mathematically the calculation is:

$$A = PN^g\lambda\ EF$$

with these definitions:

A = gamma activity in all the coolant gas, (Mev/sec)/100 Mwt

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*A dose factor, not a diffusion parameter.

P = fission power in leaking fuel elements, Mwt

N^g = total number atoms in gas per fission per second, atoms/fission-sec

λ = decay constant, dis atom^{-1} sec^{-1}

E = mean total gamma energy per disintegration, Mev dis^{-1}

F = factor to convert to (Mev/sec) per 100 Mwt, $F = 3.1 \times 10^{18}$, (fiss/sec)/100 Mwt

The values of N^g were obtained from the digital computer results.

The equation for total gamma activity in all the gas with all fuel elements leaking is:

$$A = 3.1 \times 10^{18} \sum_i N_i^g \lambda_i E_i = \text{Mev sec}^{-1}/(100 \text{ Mwt}) \text{ at } 10^8 \text{ sec}$$

The result would be multiplied by the ratio of the reactor power to 100 Mw. Since direct radiation dose calculations depend critically on the geometry of the coolant system and on the shielding effects of components and shields, no general statement on making these calculations is feasible here.

Effect of Removal Rate on Concentration in Gas

It may be seen from Tables 3 and 5 that N^g (and therefore the concentration) of most of the isotopes is reduced by about a factor of 10 for each increase in r by a factor of 10 at $D' = 10^{-7}$ to $D' = 10^{-14}$ in the range of values of r from 10^{-4} to 10^{-2}. At smaller values of r, this relation does not hold as well. Since this analysis does not separate the effects of leakage, purification, and deposition in the removal of isotopes from the coolant, no conclusions can be drawn as to their relative effects. Large values of purification removal of many of the isotopes could be obtained by passing all or a large fraction of the coolant through filters, absorbers, or chemical reactants. The feasibility of each such operation, and the fraction of the flow to be continuously treated, would be subjects for study and optimization in each case. For example, pressure drop power losses and maintenance and replacement costs would have to be considered.

Effect of D' on Concentration in Gas

The quantitative effect of a given change in D' on concentration is less predictable than for the same change in r. However, for most of the isotopes in the range $r = 10^{-6}$ to 10^{-2} the factor of decrease in N^g in going from $D' = 10^{-12}$ to $D' = 10^{-14}$ is about 10, or about a factor of 3 per decade of D'. For the same range of r values in going from $D' = 10^{-7}$ to $D' = 10^{-9}$, the factor of decrease varies more but is in the approximate range of 1.7 to 2 per decade of D'.

It would be expected that the value of D' can be lowered considerably by coating the fuel particles, by obtaining uniform fuel particles of optimum volume to surface area ratios, by controlling significant impurities in the fuel particles, or by increasing the density of the particles. Some progress has been made along each of these lines for UO_2 particle fuels,[1] and there is no apparent reason why improvement in other fuels would not be feasible.

Some Considerations in D' vs N^g Relations

Obviously, lowering the value of D' for an isotope will lower its concentration in the gas. However, the parent of an isotope with a low D' may have a comparatively high D'. In such a case the concentration of the daughter in the gas, will be less dependent on the concentration of the daughter in the fuel, since a larger fraction of the daughter in the gas will have appeared from decay of the already-escaped parent. The larger the ratio of D' parent to D' daughter, the more pronounced this effect will be. Likewise, of course, the larger the ratio of λ daughter to λ parent, the more pronounced this same effect will be. An example of combinations of λ and D' values for parent and daughter isotopes in this report shows the tendency just described, especially at low values of time operated.

From the computer output, the value of N^g (atoms fission^{-1} sec^{-1}) for Rb^{90} is essentially constant at 2.01×10^{-1} after 10^5 sec operating time with $D' = 10^{-7}$ sec^{-1}. The N^g of its daughter Sr^{90} increases markedly with time through at least 10^8 sec operating time with $D' = 10^{-7}$, as would be expected (Figs. 5 - 12) because of its long (28-yr) half-life. The N^g of

Sr^{90} consists of two parts, 1) that resulting from decay of Rb^{90} which had diffused into the gas before decaying and 2) that which was formed in the fuel and subsequently diffused out into the gas. The part of the N^g of Sr^{90} resulting from decay of Rb^{90} in the gas may be calculated as follows:

$$\frac{d\, N_{S,g}^{\;d}}{dt} = N_R^{\;g} \lambda_R - N_{S,d}^{\;g} \lambda_S$$

where

$N_{S,d}^{\;g}$ = atoms Sr^{90} in gas from decay of Rb^{90} in gas/fiss/sec

$N_R^{\;g}$ = atoms Rb^{90} in gas (constant)/fiss/sec

t = time after reactor startup, sec

λ_S, λ_R = decay constants of Sr^{90} and Rb^{90} respectively, sec^{-1}

The solution to this equation is:

$$N_{S,d}^{\;g} = N_R^{\;g} \frac{\lambda_R}{\lambda_S} (1 - e^{-\lambda_S t}) = 1.08 \times 10^6 (1 - e^{-7.85 \times 10^{-10} t})$$

Substituting values of $t = 10^5$, 10^6, 10^7, and 10^8 sec gives values of $N_{S,d}^{\;g}$ as follows:

t (sec)	10^5	10^6	10^7	10^8
$N_{S,d}^{\;g}$ ($fiss^{-1}$ sec^{-1})	8.48×10^1	8.48×10^2	8.48×10^3	8.13×10^4

The difference between these values and the total values of N^g for Sr^{90} in the gas taken from the tables represents the amount of Sr^{90} (per fission per second) which had diffused directly out of the fuel and had not yet decayed, as shown in Table 6.

Table 6. Amounts of Sr^{90} in Gas from Rb^{90} Decay and Sr^{90} Diffusion

Source Term & Condition	$t = 10^5$ sec	$t = 10^6$ sec	$t = 10^7$ sec	$t = 10^8$ sec
Case				
1) $N_{S,d}{}^g$ (calculated above; $D'_a = 10^{-7}$)	8.48×10^1	8.48×10^2	8.48×10^3	8.13×10^4
2) $N_S{}^g$ (calculated by 704, $D'_a = D'_b = 10^{-7}$)	9.30×10^1	1.62×10^3	7.90×10^4	3.69×10^6
3) $N_S{}^g$ (calculated by 704, $D'_a = 10^{-7}$, $D'_b = 10^{-9}$)	8.65×10^1	9.27×10^2	1.61×10^4	7.55×10^5
4) $N_S{}^g - N_{S,d}{}^g$ [Case 2 - Case 1] $D'_a = D'_b = 10^{-7}$	8.2×10^0	7.72×10^2	7.05×10^4	3.61×10^6
5) $N_S{}^g - N_{S,d}{}^g$ [Case 3 - Case 1] $D'_a = 10^{-7}$, $D'_b = 10^{-9}$	1.7×10^0	7.9×10^1	7.62×10^3	6.74×10^5

It can be seen that, at any time, decreasing D' for Sr^{90} relative to D' for Rb^{90}, Case 3 vs Case 2 above, significantly reduces the amount of Sr^{90} which had diffused directly into the gas and had not decayed. Table 6, Case 4 had the same D' for Rb^{90} and Sr^{90}, while Case 5 had D' for $Sr^{90} = 10^{-2}$ time D' for Rb^{90}, and at 10^8 sec after startup, the amount of Sr in Case 4 is more than five times that of Case 5.

It is also of interest to compare the amount of Sr that had diffused as Sr from the fuel to the total amount of Sr in the gas for the two cases $D'_a = D'_b = 10^{-7}$ and $D'_a = 10^{-7}$, $D'_b = 10^{-9}$. At short times (10^5) the ratio is 0.0872 $(\frac{8.2}{93})$ for

$D_a' - D_b' = 10^{-7}$ and 0.0194 $(\frac{1.7}{86.5})$ for the second case: at longer times (10^8) the ratios are 0.978 $(\frac{3.61}{3.69})$ and 0.893 $(\frac{6.74}{7.55})$ respectively. It is important to note that D_b' may be a much smaller fraction of D_a' than was assumed for Case II or Case III (10^{-1} and 10^{-2} respectively), in which case the diffusion and decay of the parent may essentially determine the N^g of the daughter. It is therefore important to determine the values of D' of parent and daughter isotopes relative to each other. Of course, if the absolute D' values can be determined, the relative values follow, and purification and leakage rates can be specified accordingly.

Conclusions and Recommendations

Every fuel considered for use in a reactor should be studied as to both the absolute and relative magnitudes of the diffusion escape rates of the fission-product elements from the fuel, as well as to the removal rates from the coolant gas. While it is evident that increasing the removal rates from the gas can effect large reductions in concentrations, it is also apparent that reductions in diffusion rates from the fuel can produce significant reductions of concentrations in the gas. The several ways known for reducing the rates of removal from the fuel to the coolant include 1) jacketing the fuel elements as with metal jackets, 2) coating the fuel element with a diffusion-resistant material (such as pryolytic graphite on graphite-based elements), 3) controlling the size and shape of the fuel particles, and 4) incorporating the fuel particles in a diffusion-resistant matrix such as a metal. An appropriate balance between control of the diffusion from the fuel and removal from the gas is finally a cost engineering problem and may be expected to be different for each type of fuel element, coolant system, and different combinations of the two.

Data are being developed on the D' values[1] obtainable by fuel treatment and manufacturing methods, and on r values attainable by physical and chemical purification of the gas coolant. By tabulating and analyzing the pertinent data from the calculation herein for the ranges of D' and r of interest, an approximate optimum combination of design specifications can then be reached on a cost basis to arrive at required maximum specifications of isotope concentrations in the coolant gas. In addition, maximum leakage rates from the

coolant system can be specified to fit the maximum concentrations allowed in the coolant in arriving at costs of protection against damage from inhaling fission products leaking from the coolant system.

The magnitudes of the effects on hazard potentials of controlling the rate of escape from the fuel and of purification of the coolant show the importance of determining fission-product diffusion rates in fuels and coolant purification rates, both as functions of costs, so that optimum designs can be approached.

Information is needed on deposition rates of fission products from the coolant, both as to amounts deposited, rates of deposition, and location of concentrated deposits. Such information would be valuable in determining the shielding requirements at all parts of the system, the requirements for maintenance, and the minimum purification requirements. It appears that the complexity of the problem will probably prevent accurate theoretical analysis, and that, therefore, most of the information needed must be determined experimentally.

This study assumes values of the parameters D' and r and presents the resulting fission product concentrations in the coolant. It does not analyze any real system except that the range of values of D' and r are expected to include those of actual real systems. Furthermore, the results show some relative relations between the effects of changes in the two parameters, and presents absolute values of concentrations for use as accurate data become available on the removal rates.

Appendix A.

Nuclear Properties of Fission Product Isotopes

Chain Atomic Mass and Branch	Chain Member	Nuclide	λ (sec^{-1})	y (Atoms $Fiss^{-1}$)	δ Fraction of y
85 - 0	1	Br_m	3.85×10^{-3}	0.015	0.8
	2	Kr^m	4.41×10^{-5}	0	
	3	Rb	0	0	
85 - 1	1	Br_m	3.85×10^{-3}	0.015	0.2
	2	Kr^m	4.41×10^{-5}	0	
	3	Kr	2.14×10^{-9}	0	
	4	Rb	0	0	
87	1	Br	1.25×10^{-2}	0.027	0.98
	2	Kr	1.48×10^{-4}	0	
	3	Rb	0	0	
88	1	Br	4.47×10^{-2}	0.029	1.0
	2	Kr	6.95×10^{-5}	0.008	
	3	Rb	6.49×10^{-4}	0	
	4	Sr	0	0	
89	1	Kr	3.63×10^{-3}	0.046	1.0
	2	Rb	7.50×10^{-4}	0.002	
	3	Sr	1.48×10^{-7}	0	
	4	Y	0	0	
90	1	Rb	4.22×10^{-3}	0.059	1.0
	2	Sr	7.85×10^{-10}	0	
	3	Y	2.98×10^{-6}	0	
	4	Zr	0	0	
91 - 0	1	Sr_m	1.99×10^{-5}	0.059	0.4
	2	Y^m	2.26×10^{-4}	0	
	3	Y	1.38×10^{-7}	0	
	4	Zr	0	0	
91 - 1	1	Sr	1.99×10^{-5}	0.059	0.6
	2	Y	1.38×10^{-7}	0	
	3	Zr	0	0	
91 - 2	1	Rb	8.25×10^{-4}	0.057	1.0

Appendix A. (con't)

92	1	Rb	8.66×10^{-3}	0.055	1.0
	2	Sr	7.13×10^{-5}	0.006	
	3	Y	5.35×10^{-5}	0	
	4	Zr	0	0	
93	1	Sr	1.65×10^{-3}	0.064	1.0
	2	Y	1.93×10^{-5}	0.001	
	3	Zr	0	0	
94	1	Sr	5.78×10^{-3}	0.058	1.0
	2	Y	7.00×10^{-4}	0.007	
	3	Zr	0	0	
95	1	Y	1.10×10^{-3}	0.064	1.0
	2	Zr	1.27×10^{-7}	0	
	3	Nb	2.29×10^{-7}	0	
	4	Mo	0	0	
96	1	Zr	0	0.064	1.0
97	1	Zr_m	1.13×10^{-5}	0.062	1.0
	2	Nb^m	1.16×10^{-2}	0	
	3	Nb	1.60×10^{-4}	0	
	4	Mo	0	0	
98	1	Mo	0	0.059	1.0
99 - 0	1	Mo_m	2.88×10^{-6}	0.061	0.1
	2	Tc^m	3.19×10^{-5}	0	
	3	Tc	0	0	
99 - 1	1	Mo	2.88×10^{-6}	0.061	0.9
	2	Tc	0	0	
100	1	Mo	0	0.065	1.0
101	1	Mo	7.91×10^{-4}	0.050	1.0
	2	Tc	8.25×10^{-4}	0	
	3	Ru	0	0	
102	1	Mo	9.63×10^{-4}	0.042	1.0
	2	Tc	2.77×10^{-2}	0	
	3	Ru	0	0	
103 - 0	1	Ru_m	1.96×10^{-7}	0.029	0.95
	2	Rh^m	2.14×10^{-4}	0	
	3	Rh	0	0	
103 - 1	1	Ru	1.96×10^{-7}	0.029	0.05
	2	Rh	0	0	

Appendix A. (con't)

104	1	Ru	0	0.018	1.0
105	1	Ru_m	4.28×10^{-5}	0.009	1.0
	2	Rh	1.54×10^{-2}		
	3	Rh	5.27×10^{-6}		
	4	Pd	0		
106	1	Ru	2.20×10^{-8}	0.0038	1.0
	2	Rh	2.31×10^{-2}	0	
	3	Pd	0	0	
127 - 0	1	Sb_m	2.07×10^{-6}	0.0025	0.16
	2	Te	8.82×10^{-8}	0	
	3	Te	2.07×10^{-5}	0	
	4	I	0	0	
127 - 1	1	Sn	1.28×10^{-4}	0.0024	1.0
127 - 2	1	Sb	2.07×10^{-6}	0.0025	0.84
	2	Te	2.07×10^{-5}	0	
	3	I	0	0	
128	1	Sb	1.75×10^{-4}	.005	1.0
	2	Te	0	0	
129 - 0	1	Sb_m	4.32×10^{-5}	0.010	0.24
	2	Te	2.43×10^{-7}	0	
	3	Te	1.60×10^{-4}	0	
	4	I	0	0	
129 - 1	1	Sb	4.32×10^{-5}	0.010	0.76
	2	Te	1.60×10^{-4}	0	
	3	I	0	0	
130	1	Sb	9.63×10^{-4}	0.020	1.0
	2	Te	0	0	
131 - 0	1	Te^m	6.42×10^{-5}	0.029	0.15
	2	Te	4.66×10^{-4}	0	
	3	I	9.96×10^{-7}	0	
	4	Xe	0	0	
131 - 1	1	Sb	5.50×10^{-4}	0.027	1.0
131 - 2	1	Te	4.66×10^{-4}	0.029	0.85
	2	I	9.96×10^{-7}	0	
	3	Xe	0	0	

Appendix A. (con't)

132	1	Sb	6.08×10^{-3}	0.034	1.0
	2	Te	2.50×10^{-6}	0.010	
	3	I	8.02×10^{-5}	0	
	4	Xe	0	0	
133 - 0	1	Te	5.78×10^{-3}	0.060	1.0
133 - 1	1	I	9.25×10^{-6}	0.065	0.976
	2	Xe	1.52×10^{-6}	0	
	3	Cs	0	0	
133 - 2	1	I	9.25×10^{-6}	0.065	0.024
	2	Xe^m	3.49×10^{-6}	0	
	3	Xe	1.52×10^{-6}	0	
	4	Cs	0	0	
134	1	Sb	1.54×10^{-2}	0.030	1.0
	2	Te	2.63×10^{-4}	0.037	
	3	I	2.20×10^{-4}	0.009	
	4	Xe	0	0	
135 - 0	1	I	2.89×10^{-5}	0.059	0.30
	2	Xe^m	7.40×10^{-4}	0	
	3	Xe	2.11×10^{-5}	0.003	
	4	Cs	0	0	
135 - 1	1	Te	5.78×10^{-3}	0.042	1.0
135 - 2	1	I	2.89×10^{-5}	0.059	0.70
	2	Xe	2.11×10^{-5}	0.003	
	3	Cs	0	0	
136	1	I	8.06×10^{-3}	0.031	1.0
	2	Xe	0	0.001	
137 - 0	1	Xe	2.96×10^{-3}	0.059	0.92
	2	Cs	8.27×10^{-10}	0	
	3	Ba^m	4.44×10^{-3}	0	
	4	Ba	0	0	
137 - 1	1	Xe	2.96×10^{-3}	0.059	0.08
	2	Cs	8.27×10^{-10}	0	
	3	Ba	0	0	
138	1	I	1.17×10^{-1}	0.034	1.0
	2	Xe	6.79×10^{-4}	0.021	
	3	Cs	3.62×10^{-4}	0.003	
	4	Ba	0	0	

Appendix A. (con't)

139	1	Xe	1.69×10^{-2}	0.047	1.0
	2	Cs	1.22×10^{-3}	0.012	
	3	Ba	1.36×10^{-4}	0.001	
	4	La	0	0	
140	1	Cs	1.05×10^{-2}	0.060	1.0
	2	Ba	6.27×10^{-7}	0.003	
	3	La	4.79×10^{-6}	0	
	4	Ce	0	0	
141	1	Ba	6.42×10^{-4}	0.059	1.0
	2	La	5.20×10^{-5}	0.001	
	3	Ce	2.51×10^{-7}	0	
	4	Pr	0	0	
142	1	Cs	1.16×10^{-2}	0.034	1.0
	2	Ba	1.93×10^{-3}	0.022	
	3	La	1.56×10^{-4}	0.003	
	4	Ce	0	0	
143	1	La	6.08×10^{-4}	0.062	1.0
	2	Ce	6.01×10^{-6}	0	
	3	Pr	5.85×10^{-7}	0	
	4	Nd	0	0	
144	1	Ce	2.76×10^{-8}	0.061	1.0
	2	Pr	6.60×10^{-4}	0	
	3	Nd	0	0	
145	1	Ce	3.85×10^{-3}	0.042	1.0
	2	Pr	3.21×10^{-5}	.0	
	3	Nd	0	0	
146	1	Ce	8.31×10^{-4}	0.032	1.0
	2	Pr	4.73×10^{-4}	0.001	
	3	Nd	0	0	
147	1	Nd	7.10×10^{-7}	0.026	1.0
	2	Pm	8.46×10^{-9}	0	
	3	Sm	0	0	
148	1	Nd	0	0.018	1.0
149	1	Nd	9.63×10^{-5}	0.013	1.0
	2	Pm	3.56×10^{-6}	0	
	3	Sm	0	0	
150	1	Nd	0	0.0074	1.0
151	1	Nd	7.7×10^{-4}	0.0048	1.0
	2	Pm	7.0×10^{-6}	0.0002	
	3	Sm	2.37×10^{-10}	0	
	4	Eu	0	0	

Appendix B

Arrangement and Use of the IBM 704 Print-Out Tables

Input Data

The input data to the IBM 704 computer are shown in Appendix A of this report. These input data used in the calculations were abstracted from Blomeke's tables[12] of nuclide parameters. Appendix A is a tabulation of 1) chain mass number, 2) nuclide code number (first, second, third, or fourth member of the chain), 3) decay constant, λ, of nuclide, 4) fission yield, y, of nuclide, and 5) fraction, δ, of fission yield involved in the decay chain branch indicated in column 1.

It is necessary in many cases to consult the input data to interpret the IBM 704 print-out of the calculated results. For example, the branches of decay chains are calculated as separate chains, and the total concentrations of the nuclides are obtained by adding the results of the calculations of the branches. For example, in the mass-85 chain, the two branches are added for 3.00 min barium, 4.36-hr krypton, and stable rubidium, but not for 10.27-yr krypton. (To clarify, see the diagram of the decay chain of Br^{85}.)[12] Thus, on the first page of the Case I isotope table, the total N^g for Br^{85} is $0.469377 \times 10^{-1} + 0.117344 \times 10^{-1} = 0.586 \times 10^{-1}$ atoms fission^{-1} sec^{-1} at $T = 10^5$ sec, $D' = 10^{-7}$, and $r = 0$. The total N^g for Rb^{85} is $0.244453 \times 10^3 + 0.521322 \times 10^{-2} = 0.244 \times 10^3$ atoms fission^{-1} sec^{-1}.

Computer Print-Out Table Headings

The complete IBM 704 print-outs of results are available although not included in this report. They organized into the following tables: individual isotopes, Cases I, II, and III respectively. Each of these tables is divided into subtables arranged in order by parameter values. The values of the parameters used are printed at the beginning of each subtable. Each subtable has the print-out or results arranged in order by mass number. A brief discussion of each table follows.

Individual Isotopes, Case I. At the top of the first page of each subtable values of D', r, and T (diffusion parameter, removal rate from gas, and time after startup) are listed in sec^{-1}, sec^{-1}, and sec respectively. On each page the extreme right-hand column lists for each row the chain mass number with an additional integer to indicate the branch of the decay chain

for which the results are printed in that row. When the same nuclide (isomer or isotope) appears in two or more separate rows, the total concentration is obtained by adding the results, as explained above.

In the Case I isotope table, the machine-printed values of D', r, and T are correct and straightforward.

Cases II and II Isotope Tables. The subtables for Cases II and III isotopes are arranged in the same order and with the same meanings as the subtables for Case I isotopes, with the following exceptions:

1) To save computer time, only the mass chains 88 through 97 and 137 through 146 were calculated for these two cases.

2) For the subtables headed $T = 0.1 \times 10^6$ (sec), the D in the heading is D'_a. For the subtables headed $T = 0.1 \times 10^7$, 0.1×10^8, and 0.1×10^9, the D in the heading is D'_b ($D'_b = 0.1\ D'_a$ for Case II; $D'_b = 0.01\ D'_a$ for Case III).

Nuclide Sums, All Cases

The tables for sums of nuclides for Case I are straightforward.

To obtain Case II nuclide sums for strontium, yttrium, zirconium, niobium, barium, lanthanum, cesium, and promethium, the Case II tables of nuclide sums are used direct with $D' = D'_b = 0.1\ D'_a$. Results from sums of other nuclides are obtained from the Case I nuclide sums tables as follows. For barium, krypton, rubidium, tin, antimony, tellurium, iodine, xenon, and cesium, the results are obtained from the Case I sums tables with $D' = D'_a$. For molybdenum, technetium, ruthenium, rhodium, neodymium, promethium, samarium, and europium, the results are obtained from the Case I nuclide sums tables with $D' = 0.1\ D'_a$. The D value listed at the beginning of each subtable is D'_b.

For the Case III nuclide sums of strontium, yttrium, zirconium, niobium, barium, lanthanum, cerium, and praseodymium, the Case III nuclide sums tables are used direct, with $D' = D'_b$. For barium, krypton, rubidium, tin, antimony, tellurium, iodine, xenon, and cesium, the results are obtained from the Case I sums tables with $D' = D'_a$. For molybedenum, technetium, ruthenium, rhodium, neodymium, promethium, samarium, and europium, the results are obtained from the Case I sums tables with $D' = 0.01\ D'_a$. The D value listed at the beginning of each subtable is D'_b.

For illustration, Table 7 lists some examples.

Table 7. Illustrations of Use of Sums Tables for Cases II and III

Case	Element	D'_a	D'_b	Find Concentration under Sums Table for Case	D
II	Kr	10^{-8}	10^{-9}	I	10^{-8}
III	Sm	10^{-9}	10^{-11}	I	10^{-11}
II	Sr	10^{-7}	10^{-8}	II	10^{-8}
III	Ba	10^{-8}	10^{-10}	III	10^{-10}
III	Rb	10^{-8}	10^{-10}	I	10^{-8}
III	Nd	10^{-12}	10^{-14}	III	10^{-14}
II	Pd	10^{-12}	10^{-13}	II	10^{-13}

APPENDIX C

Arrangement and Use of Proposed Tables of Computer Output

It is proposed that the original print-out tables from the IBM 704 computer be copied in a more convenient arrangement and form for use for inclusion as an addendum to this report at some later date. Several complications in the original form, explained in Appendix B, would be avoided in the rearranged tables. Table 8 is a sample page of the proposed new format.

There would be six tables of results in this addendum. The six tables would present the calculated N^g (number of atoms per fission per second) of individual isotopes and of individual elements in the gas coolant as a function of time after reactor startup for Cases I, II, and III. The arrangement would be such that each page presents the N^g values for one isotope, isomer, or element in the coolant for one case number, for all six values each of D' and r, calculated at each of four values of time after startup. In each table the element would be taken in order by mass number. In cases of two isomers of the same isotope, the page of results for the upper branch of the chain would precede the page for the lower branch.

The number of atoms in a leaking fuel element can be obtained by calculating the total number per fission per second in a nonleaking element and subtracting from it the number in the gas and multiplying the result by the number of fissions per second in the element.

Table 8.

Concentrations of Individual Isotopes in Gas (Case I)

			Total Atoms in Gas, $Fiss^{-1}$ sec^{-1}			
			At Time, seconds after startup			
Isotope	r (sec^{-1})	D_a' (sec^{-1})	10^5	10^6	10^7	10^8
Kr^{-85}	0	10^{-7}	2.55×10^{1}	4.51×10^{2}	8.93×10^{3}	2.14×10^{5}
		10^{-8}	8.58×10^{0}	1.56×10^{2}	3.27×10^{3}	1.28×10^{5}
		10^{-9}	2.78×10^{0}	5.09×10^{1}	1.09×10^{3}	5.32×10^{4}
		10^{-10}	8.91×10^{-1}	1.62×10^{1}	3.49×10^{2}	1.85×10^{4}
		10^{-11}	2.44×10^{-1}	5.11×10^{0}	1.11×10^{2}	6.05×10^{3}
		10^{-12}	7.91×10^{-2}	1.60×10^{0}	3.52×10^{1}	1.93×10^{3}
	10^{-6}	10^{-7}	2.42×10^{1}	2.89×10^{2}	1.24×10^{3}	2.84×10^{3}
		10^{-8}	8.17×10^{0}	1.00×10^{2}	4.71×10^{2}	2.10×10^{3}
		10^{-9}	2.64×10^{0}	3.27×10^{1}	1.58×10^{2}	2.10×10^{3}
		10^{-10}	8.40×10^{-1}	1.04×10^{1}	5.11×10^{1}	3.60×10^{2}
		10^{-11}	2.66×10^{-1}	3.31×10^{0}	1.62×10^{1}	1.19×10^{2}
		10^{-12}	8.43×10^{-2}	1.05×10^{0}	5.15×10^{0}	3.83×10^{1}
	10^{-5}	10^{-7}	1.57×10^{1}	4.36×10^{1}	1.24×10^{2}	2.78×10^{2}
		10^{-8}	5.32×10^{0}	1.51×10^{1}	4.79×10^{1}	2.09×10^{2}
		10^{-9}	1.72×10^{0}	4.94×10^{0}	1.62×10^{1}	9.89×10^{1}
		10^{-10}	5.48×10^{-1}	1.58×10^{0}	5.24×10^{0}	3.61×10^{1}
		10^{-11}	1.74×10^{-1}	5.01×10^{-1}	1.67×10^{0}	1.20×10^{1}
		10^{-12}	5.49×10^{-2}	1.59×10^{-1}	5.29×10^{-1}	3.84×10^{0}
	10^{-4}	10^{-7}	1.32×10^{0}	2.34×10^{0}	1.03×10^{1}	2.57×10^{1}
		10^{-8}	4.54×10^{-1}	8.19×10^{-1}	4.09×10^{0}	2.02×10^{1}
		10^{-9}	1.48×10^{-1}	2.68×10^{-1}	1.39×10^{0}	9.66×10^{0}
		10^{-10}	4.71×10^{-2}	8.56×10^{-2}	4.51×10^{-1}	3.54×10^{0}
		10^{-11}	1.49×10^{-2}	2.72×10^{-2}	1.44×10^{-1}	1.17×10^{0}
		10^{-12}	4.73×10^{-3}	8.60×10^{-3}	4.56×10^{-2}	3.77×10^{-1}
	10^{-2}	10^{-7}	1.07×10^{-3}	1.10×10^{-2}	9.10×10^{-2}	2.44×10^{-1}
		10^{-8}	3.68×10^{-4}	3.91×10^{-3}	3.66×10^{-2}	1.98×10^{-1}
		10^{-9}	1.19×10^{-4}	1.28×10^{-3}	1.25×10^{-2}	9.52×10^{-2}
		10^{-10}	3.81×10^{-5}	4.10×10^{-4}	4.07×10^{-3}	3.49×10^{-2}
		10^{-11}	1.21×10^{-5}	1.30×10^{-4}	1.30×10^{-3}	1.16×10^{-2}
		10^{-12}	3.82×10^{-6}	4.12×10^{-5}	4.11×10^{-4}	3.72×10^{-3}
	1	10^{-7}	9.08×10^{-6}	1.09×10^{-4}	9.08×10^{-4}	2.44×10^{-3}
		10^{-8}	3.11×10^{-6}	3.85×10^{-5}	3.66×10^{-4}	Missing
		10^{-9}	1.01×10^{-6}	1.26×10^{-5}	1.25×10^{-4}	9.52×10^{-4}
		10^{-10}	3.21×10^{-7}	4.04×10^{-6}	4.06×10^{-5}	3.49×10^{-4}
		10^{-11}	1.02×10^{-7}	1.28×10^{-6}	1.29×10^{-5}	1.16×10^{-4}
		10^{-12}	3.22×10^{-8}	4.06×10^{-7}	4.11×10^{-6}	3.72×10^{-5}

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2) J. D. Eichenberg, et al., Effects of Irradiation on Bulk UO_2, WAPD-183, Chapter III, Section F, October 1957.

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5) G. W. Parker, et al., Fission Product Release from UO_2 by High Temperature Diffusion and Melting in Helium and Air, ORNL CF-60-12-14, December 1960.

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7) P. I. Conn, et al., Isotope Deposition Hazards in Gas Cycle Reactors, presented at the Annual Meeting of American Nuclear Society, Gatlinburg, Tennessee, June 17, 1959.

8) Cottrell, op. cit., Table XVII.

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10) T. J. Burnett, Reactors, Hazard vs Power Level, Nuclear Science and Engineering, Vol 2, 382-93 (1957).

11) Report of Committee II of the International Commission on Radiological Protection on Permissible Doses for Internal Radiation, 1959, revision.

12) J. O. Blomeke and M. F. Todd, Part I, Vol. I, Uranium Fission Product Production as a Function of Thermal Neutron Flux, Irradiation Time, and Decay Time, 1. Atomic Concentrations and Gross Totals, ORNL-2127, p. 13-39, August 19, 1957.

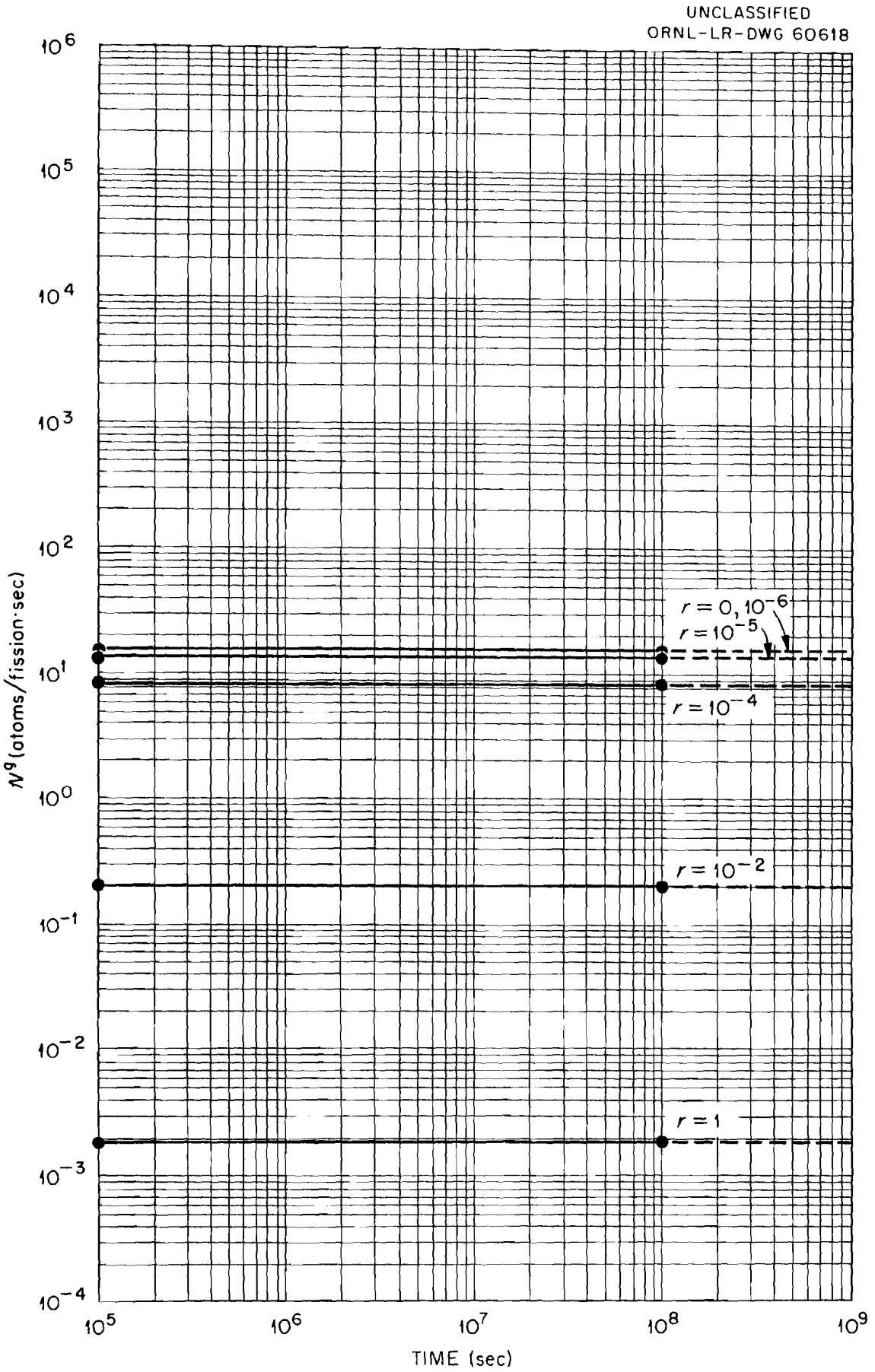

Fig. 1. Buildup of Fission Products in Coolant; Kr^{87}, Case I, $D' = 10^{-7}$, $r = 0$ to 1 (sec^{-1}).

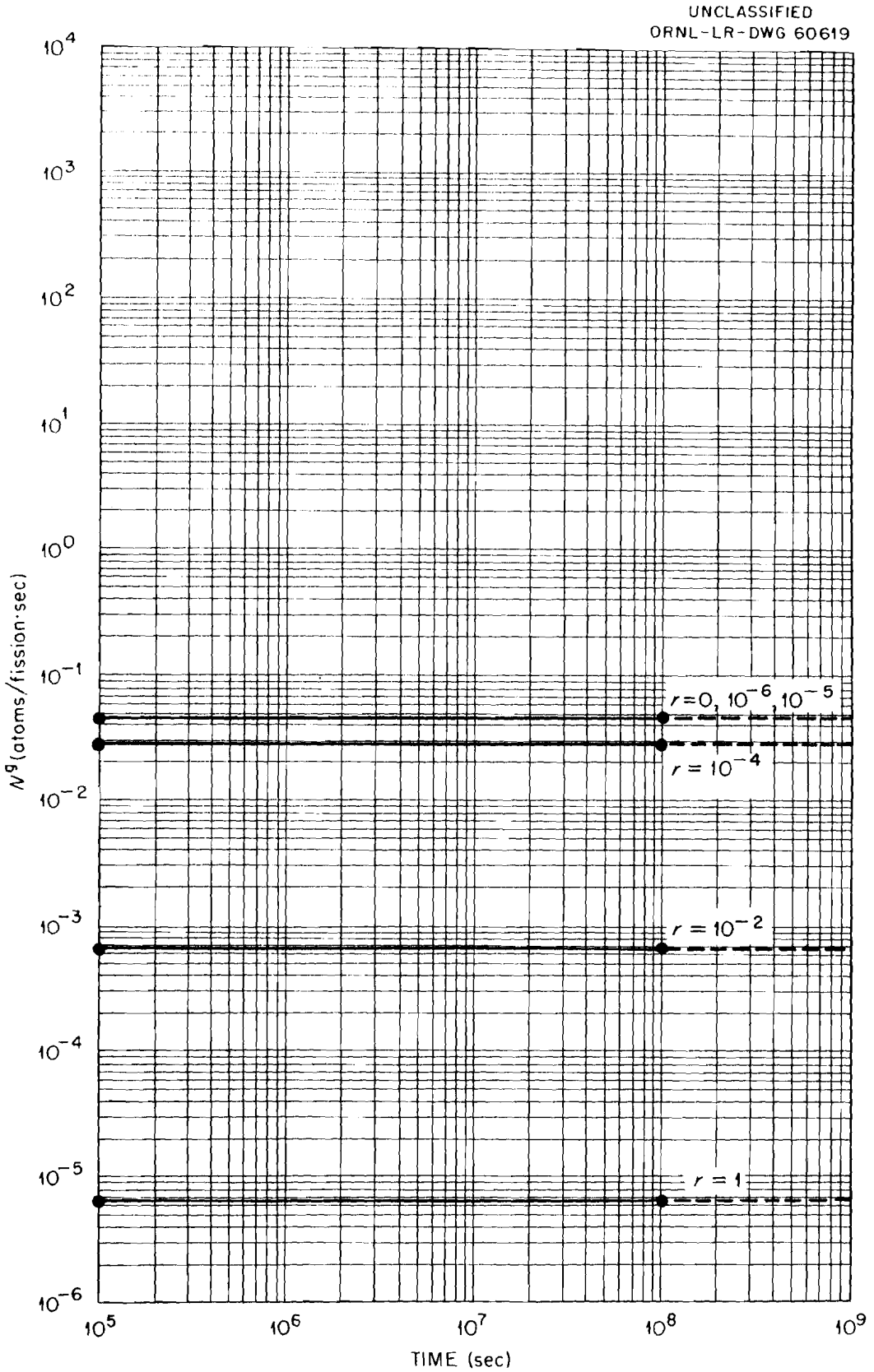

Fig. 2. Buildup of Fission Products in Coolant; Kr^{87}, Case I, $D' = 10^{-12}$, $r = 0$ to 1 (sec^{-1}).

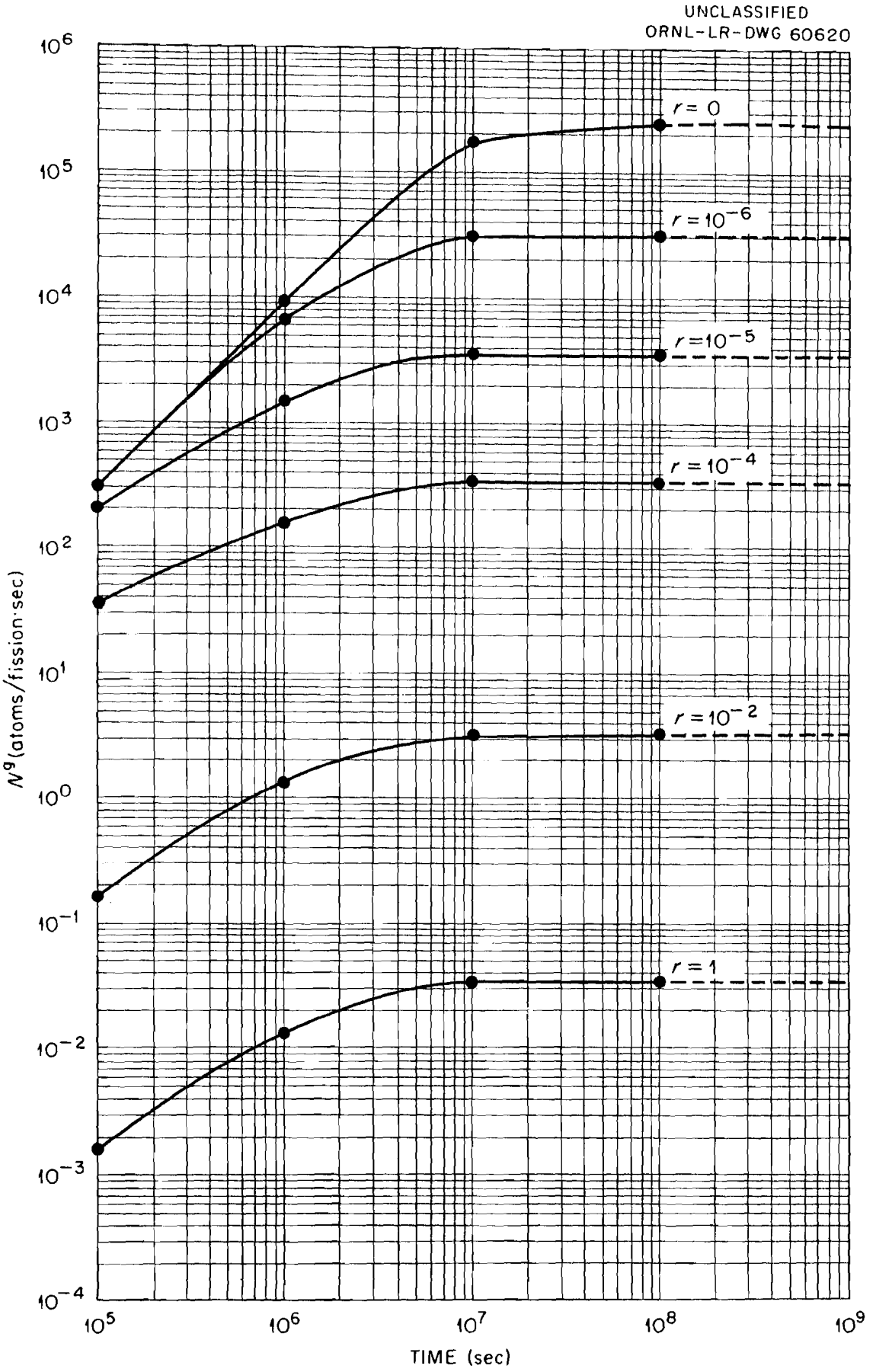

Fig. 3. Buildup of Fission Products in Coolant; Sr^{89}, Case I, $D' = 10^{-7}$, $r = 0$ to 1 (sec^{-1}).

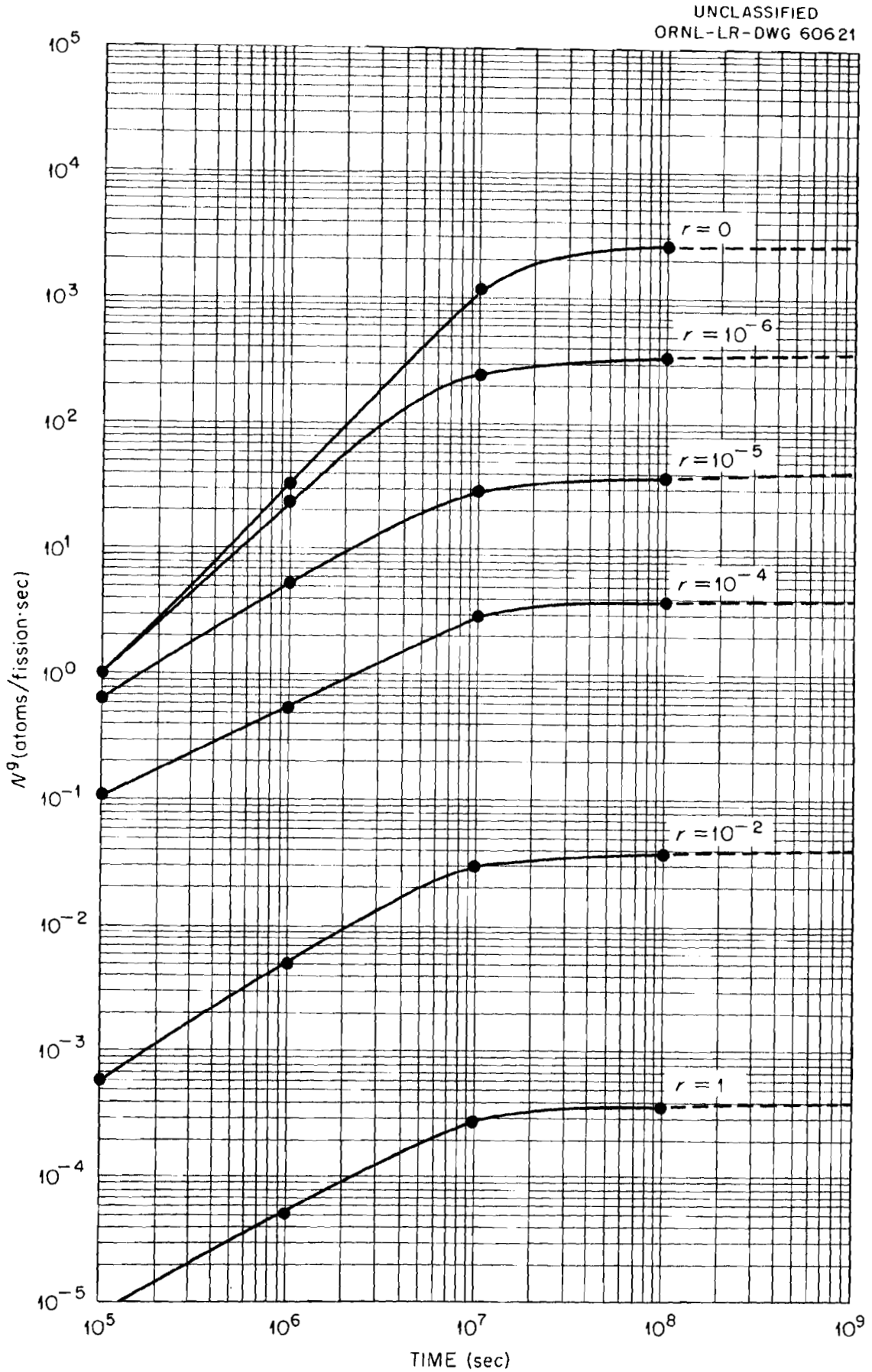

Fig. 4. Buildup of Fission Products in Coolant; Sr^{89}, Case I, $D' = 10^{-12}$, $r = 0$ to 1 (sec^{-1}).

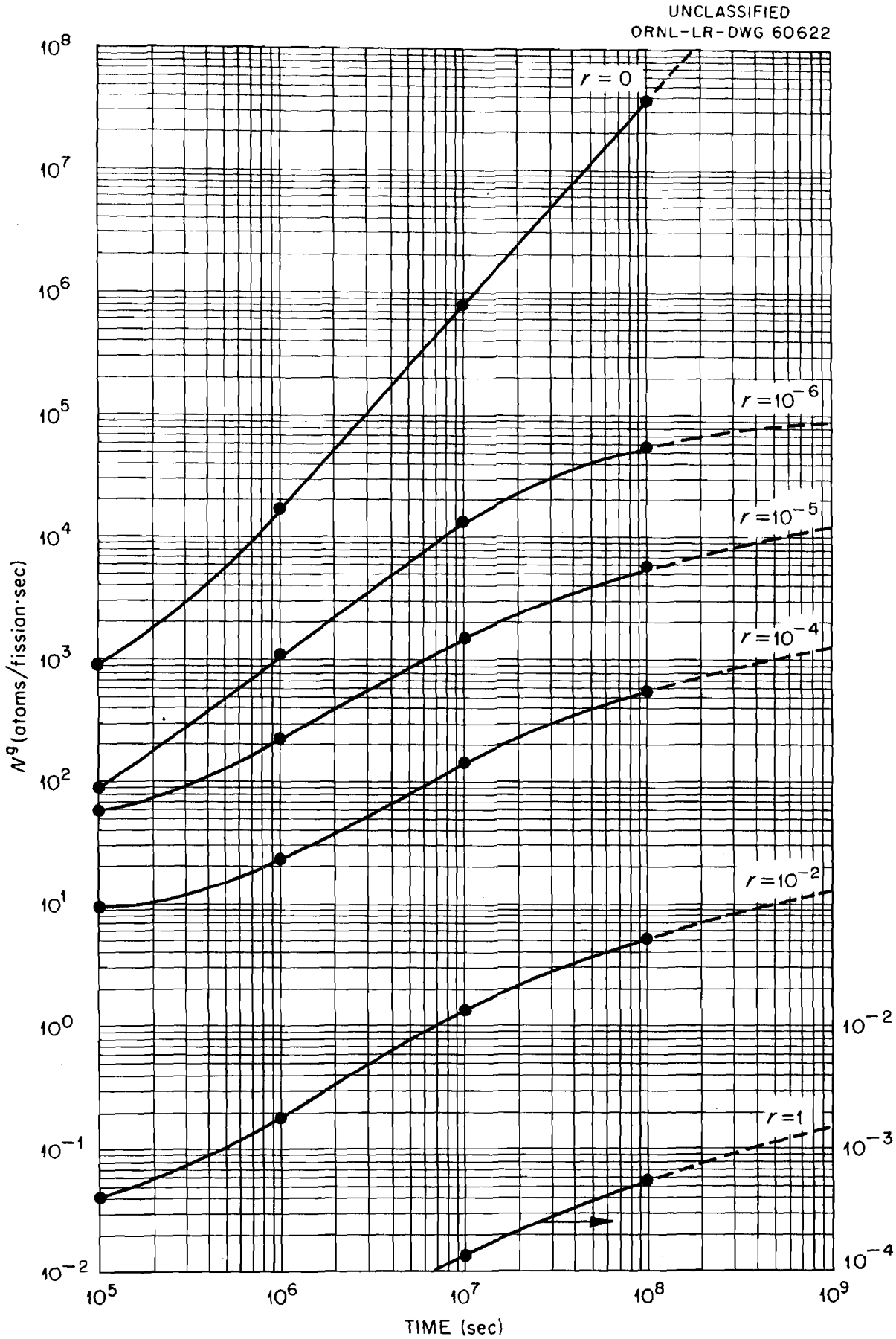

Fig. 5. Buildup of Fission Products in Coolant; Sr^{90}, Case I, $D' = 10^{-7}$, $r = 0$ to 1 (sec^{-1}).

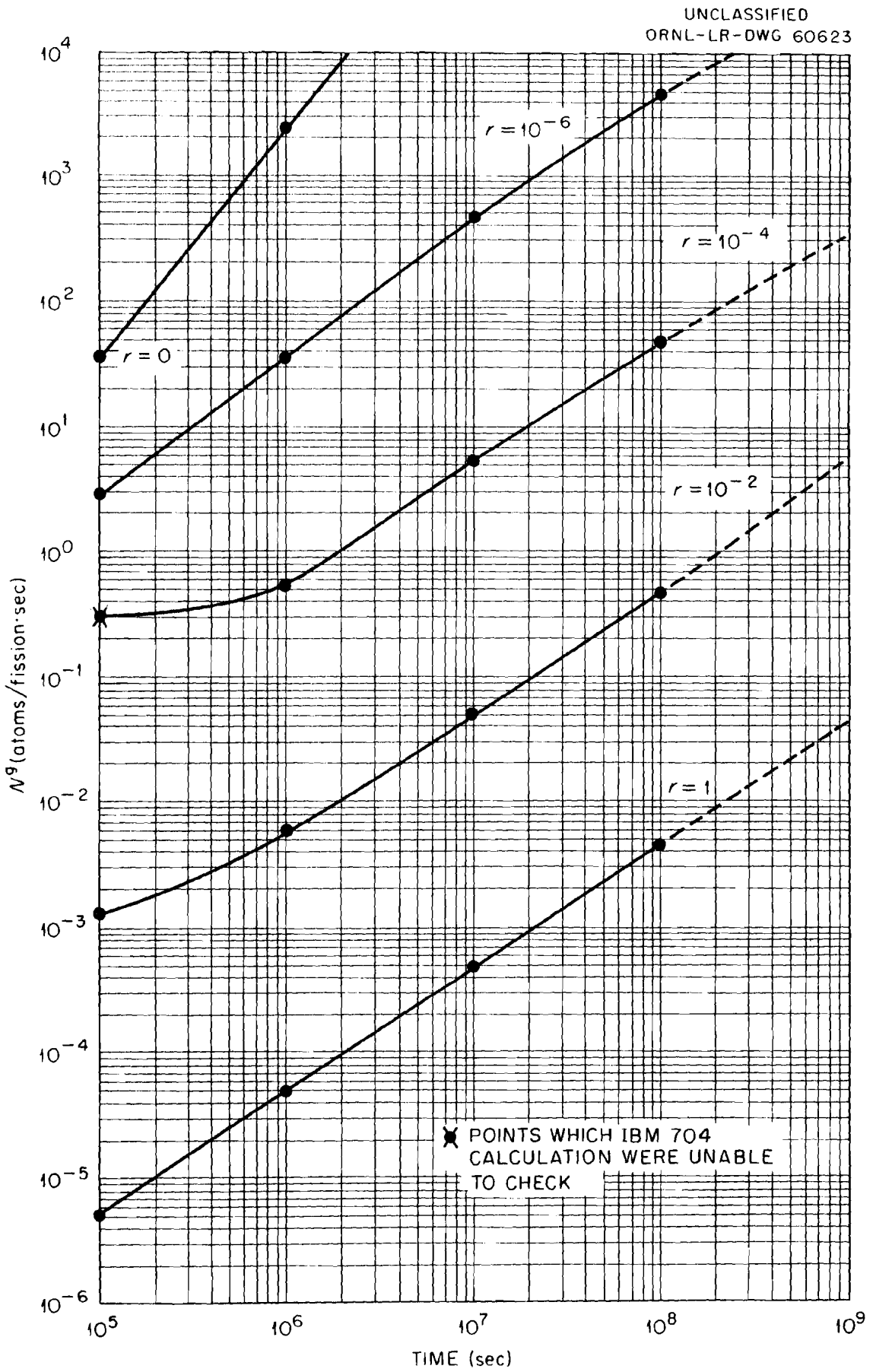

Fig. 6. Buildup of Fission Products in Coolant; Sr^{90}, Case I, $D' = 10^{-10}$, $r = 0$ to 1. (sec^{-1}).

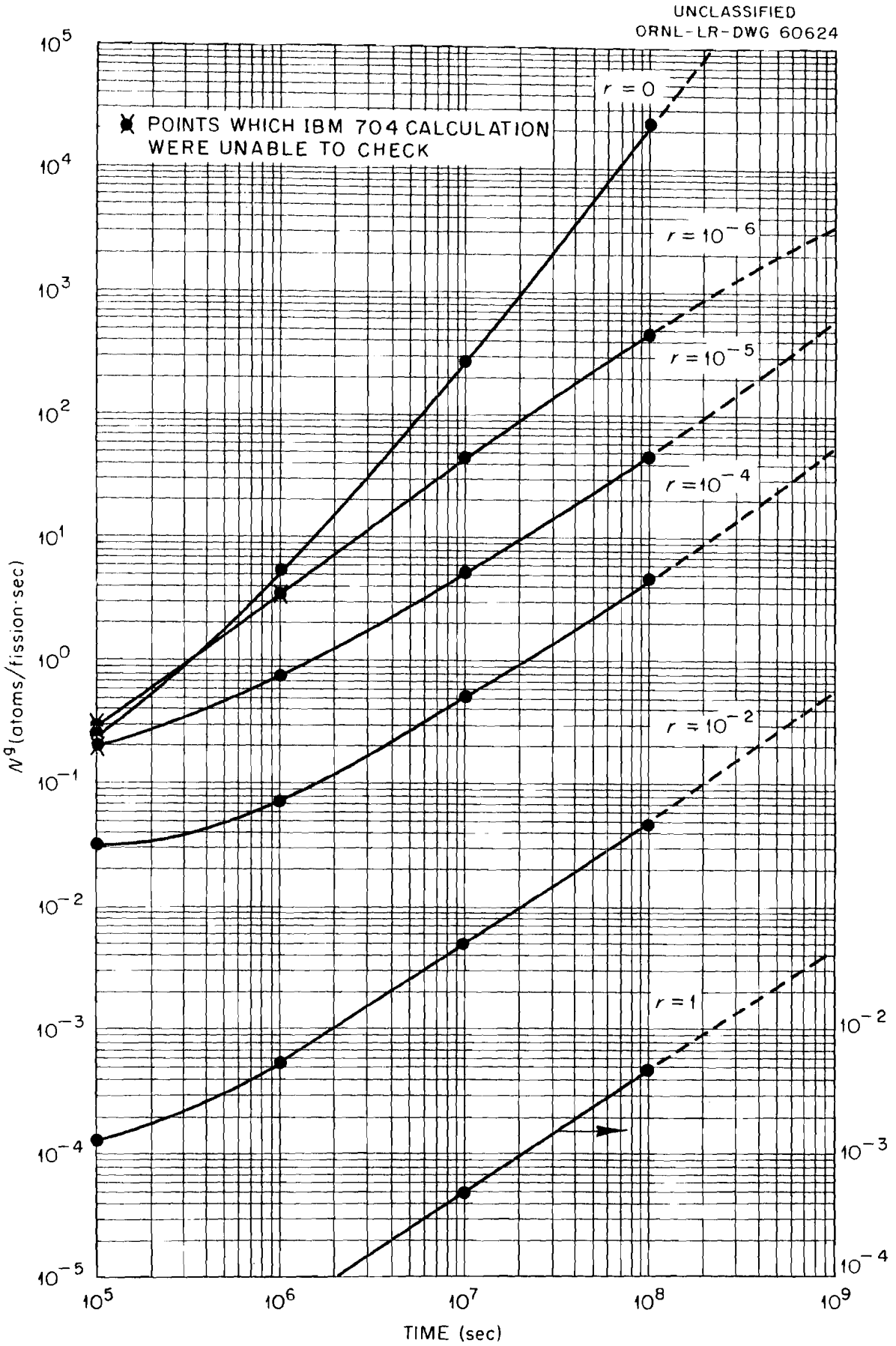

Fig. 7. Buildup of Fission Products in Coolant; Sr^{90}, Case I, $D' = 10^{-12}$, $r = 0$ to 1 (sec^{-1}).

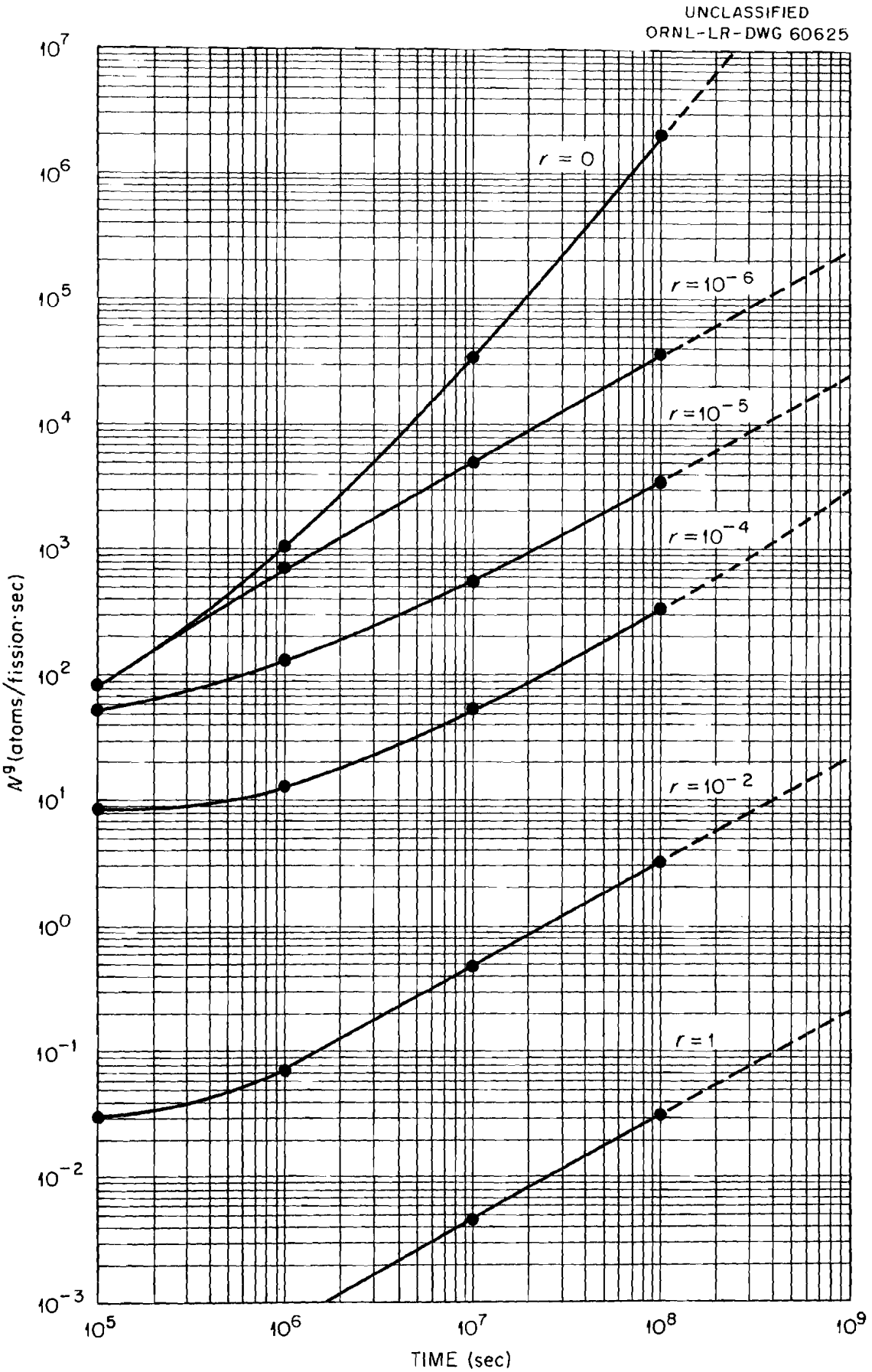

Fig. 8. Buildup of Fission Products in Coolant; Sr^{90}, Case II, $D_a' = 10^{-7}$, $r = 0$ to 1 (sec^{-1}).

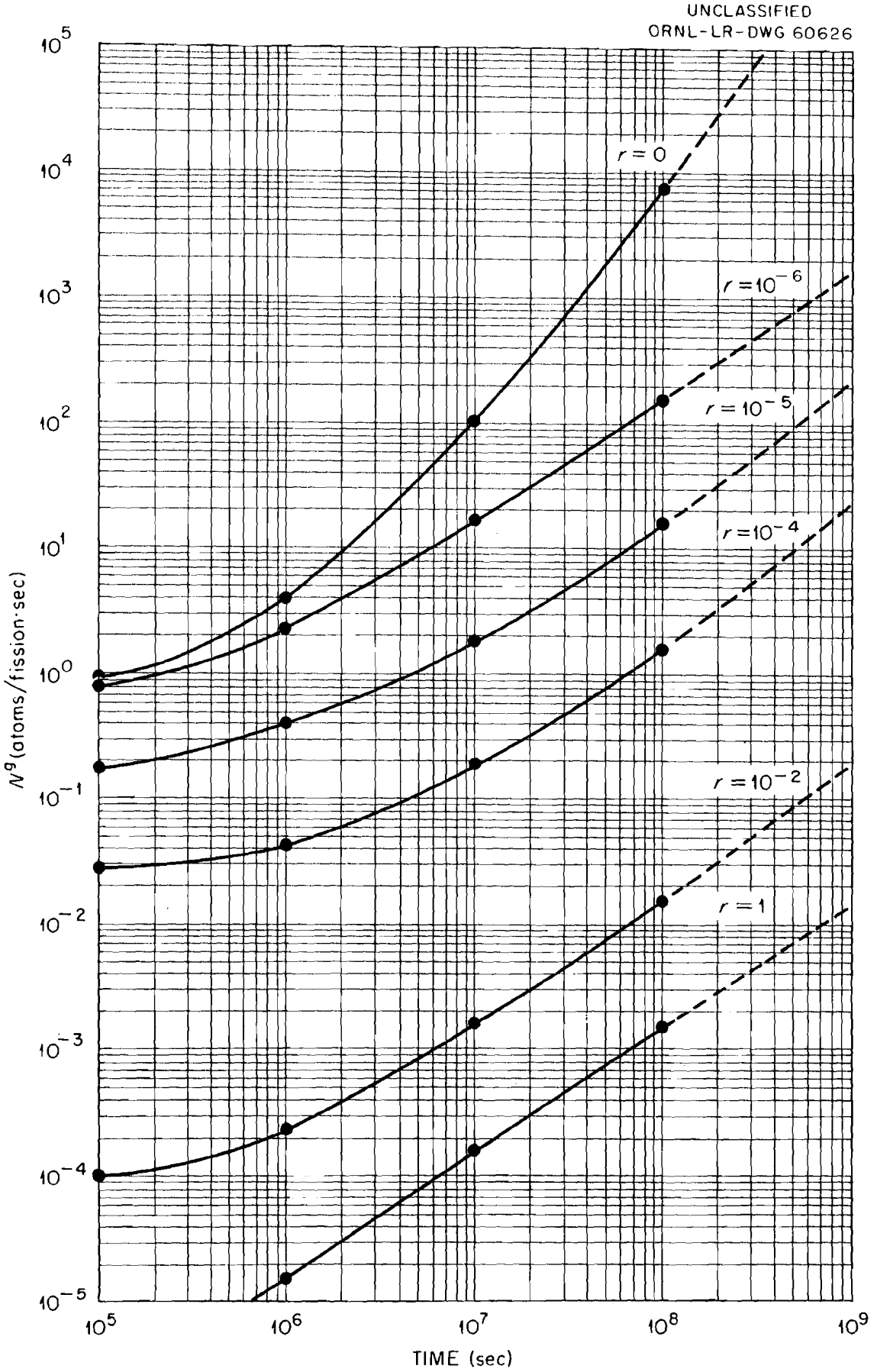

Fig. 9. Buildup of Fission Products in Coolant; Sr^{90}, Case II, $D_a' = 10^{-12}$, $r = 0$ to 1 (sec^{-1}).

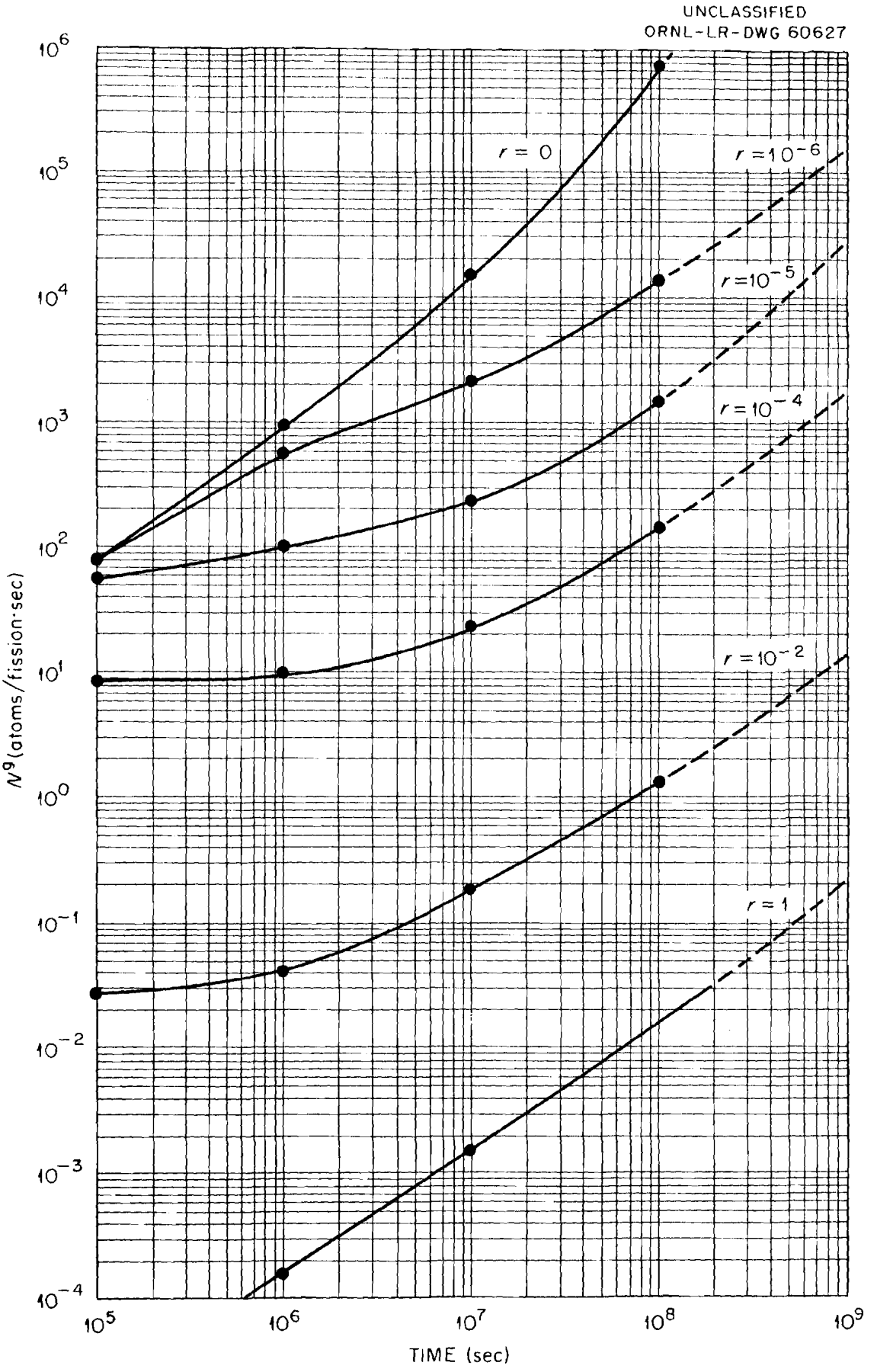

Fig. 10. Buildup of Fission Products in Coolant; Sr^{90}, Case III, $D_a' = 10^{-7}$, $r = 0$ to 1 (sec^{-1}).

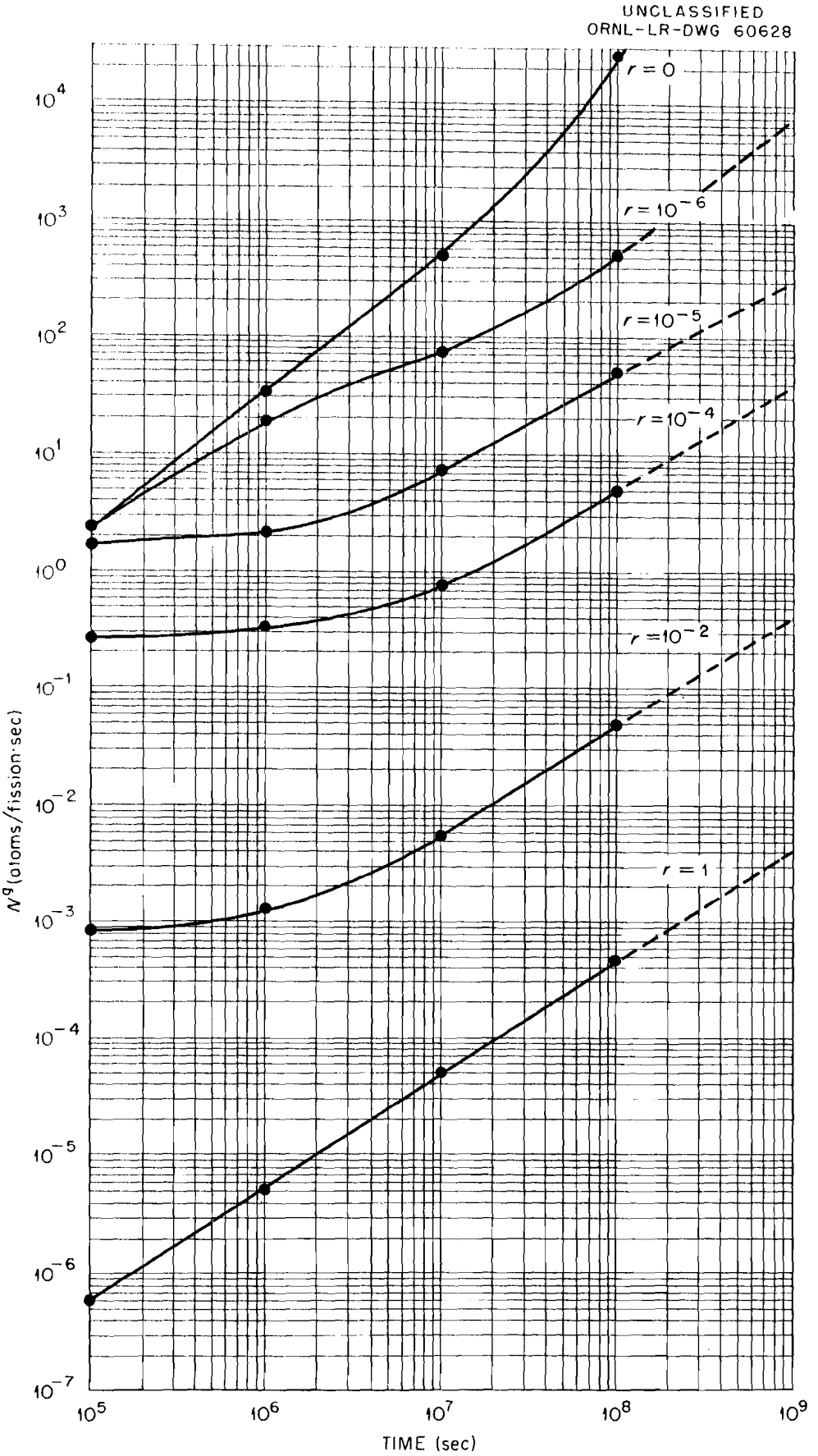

Fig. 11. Buildup of Fission Products in Coolant; Sr^{90}, Case III, $D_0' = 10^{-10}$, $r = 0$ to 1 (sec^{-1}).

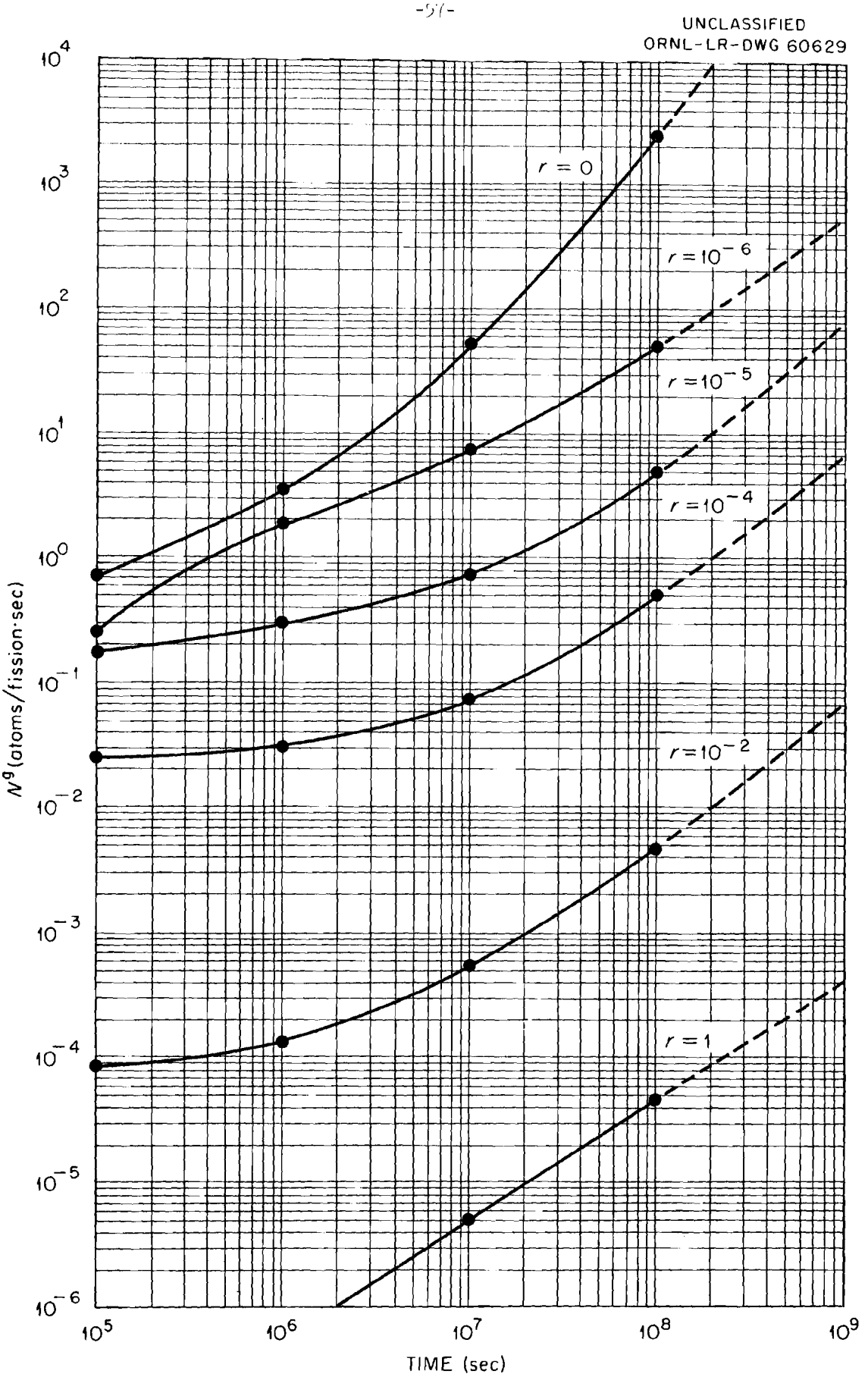

Fig. 12. Buildup of Fission Products in Coolant; Sr^{90}, Case III, $D_a' = 10^{-12}$, $r = 0$ to 1 (sec^{-1}).

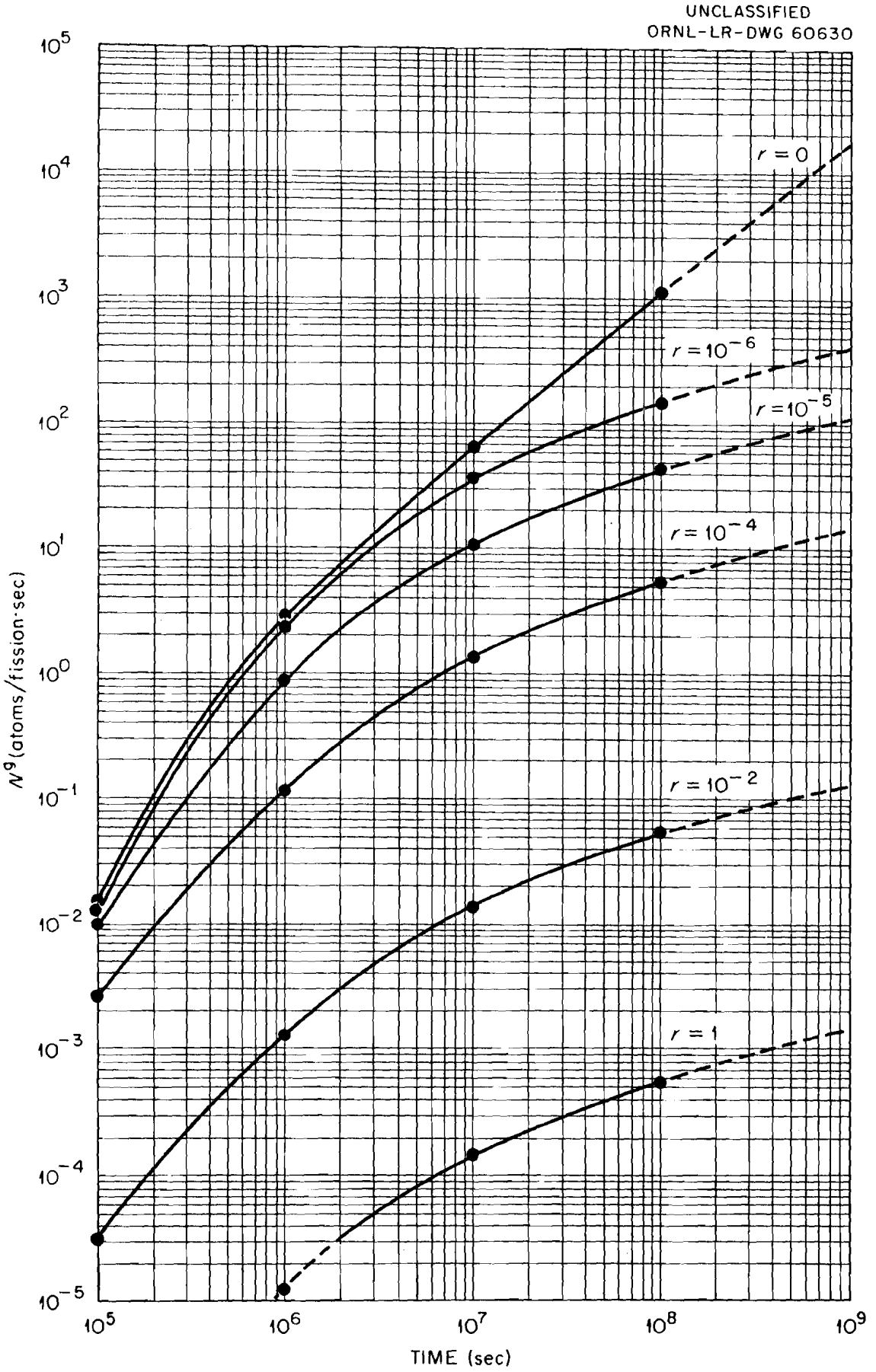

Fig. 13. Buildup of Fission Products in Coolant; Y^{90}, Case I, $D' = 10^{-7}$, $r = 0$ to 1 (sec^{-1}).

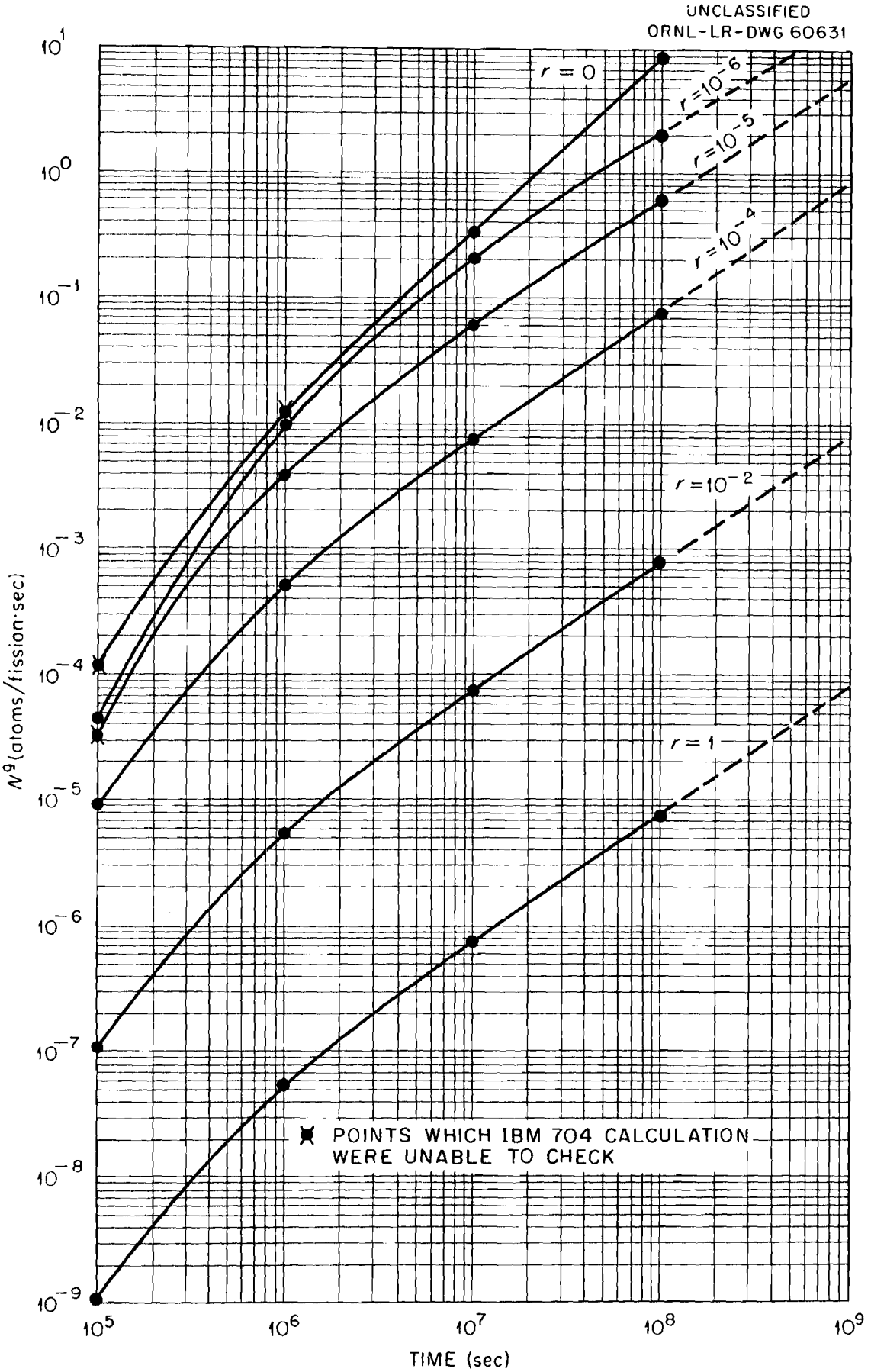

Fig. 14. Buildup of Fission Products in Coolant; Y^{90}, Case I, $D' = 10^{-12}$, $r = 0$ to 1 (sec^{-1}).

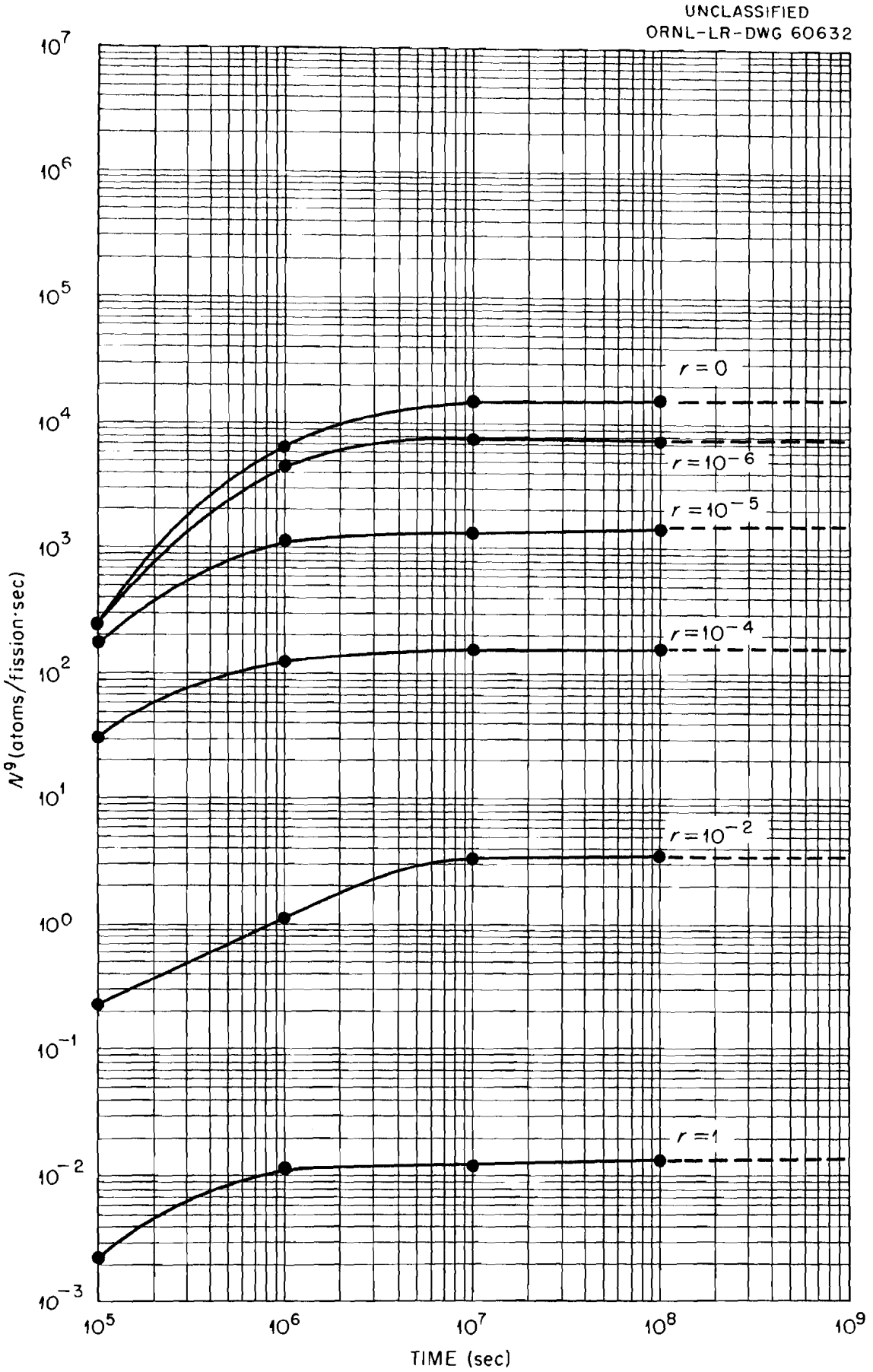

Fig. 15. Buildup of Fission Products in Coolant; I^{131}, Case I, $D' = 10^{-7}$, $r = 0$ to 1 (sec^{-1}).

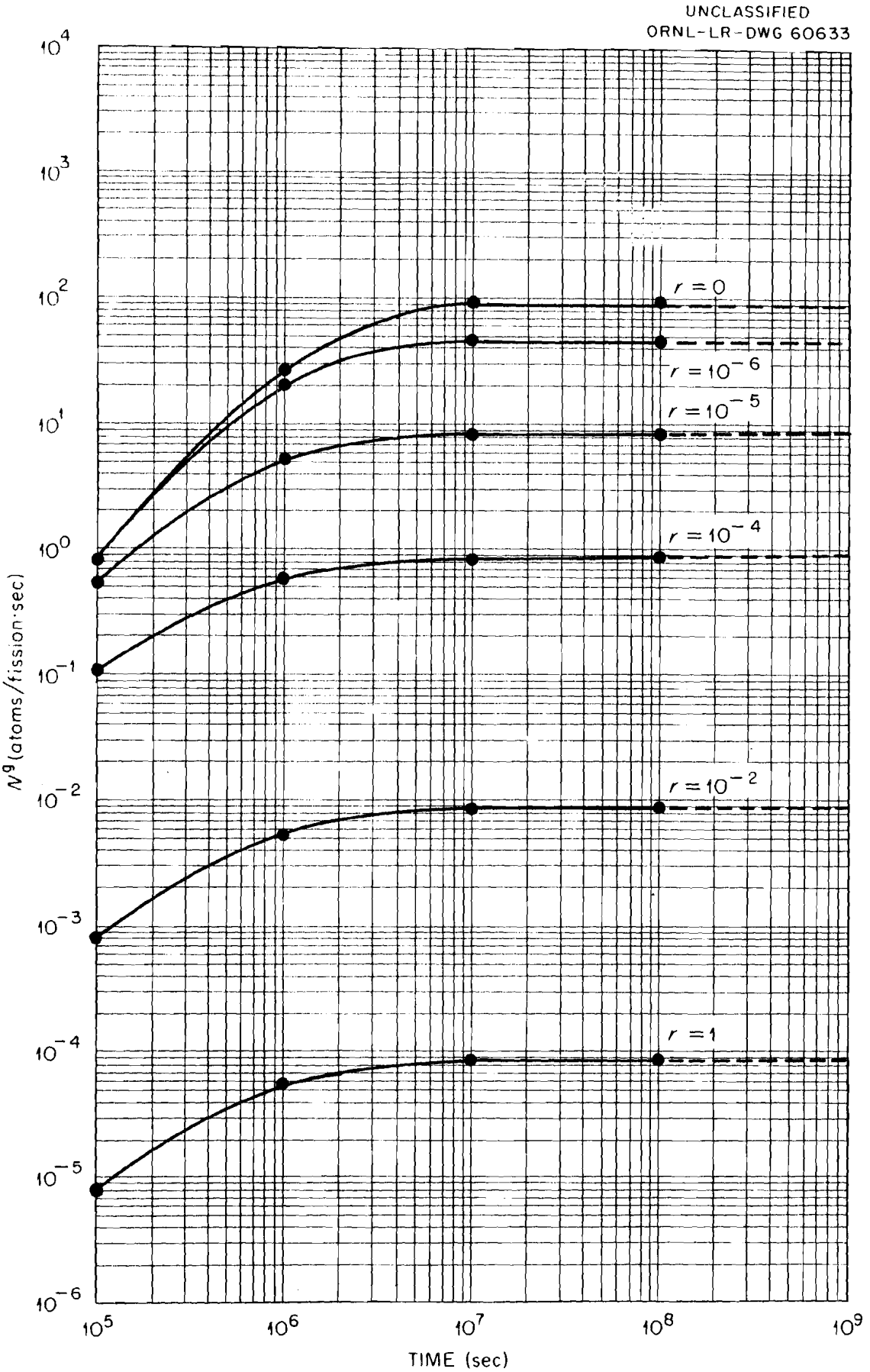

Fig. 16. Buildup of Fission Products in Coolant; I^{131}, Case I, $D' = 10^{-12}$, $r = 0$ to 1 (sec^{-1}).

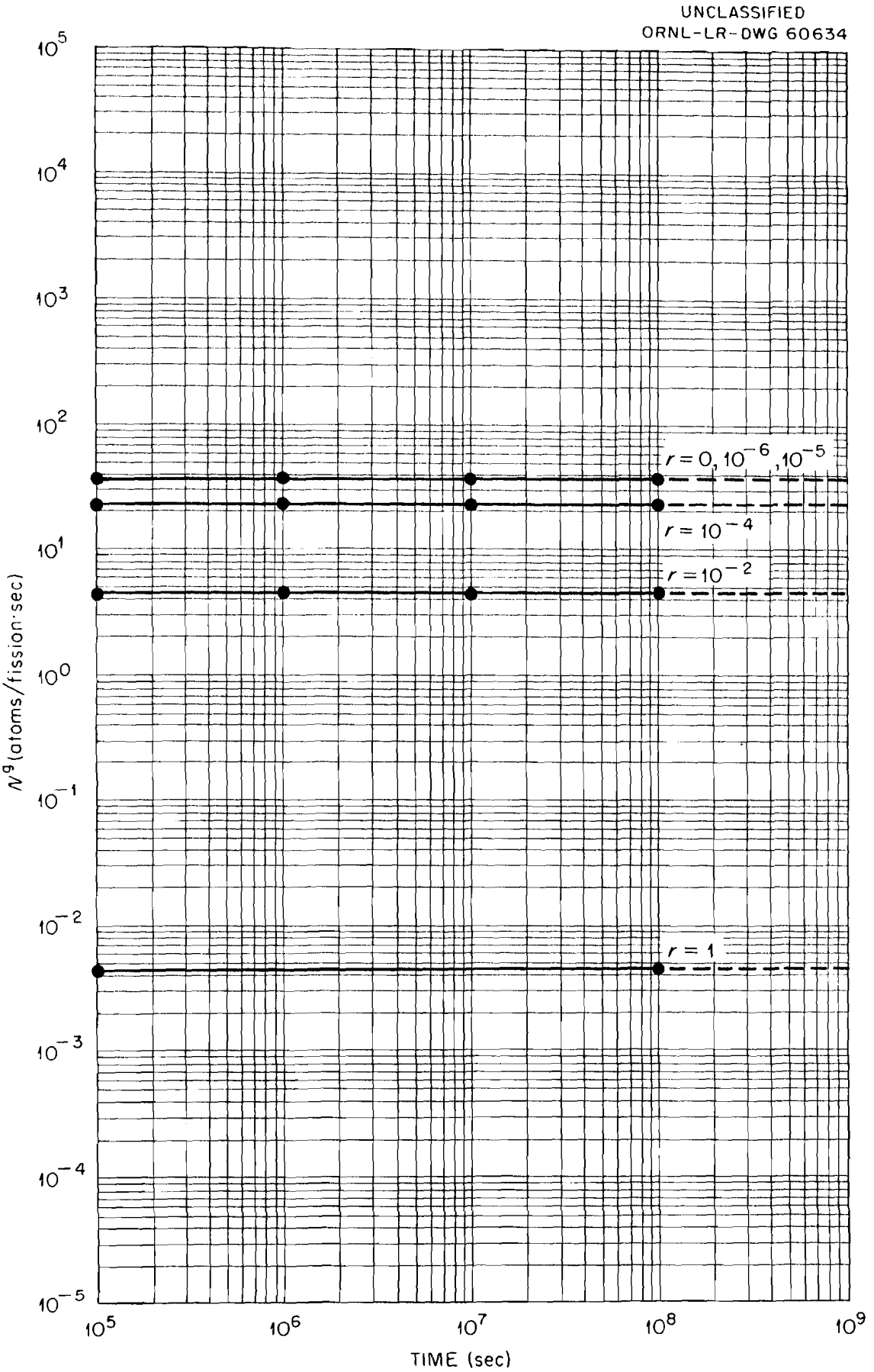

Fig. 17. Buildup of Fission Products in Coolant; I^{134}, Case I, $D' = 10^{-7}$, $r = 0$ to 1 (sec^{-1}).

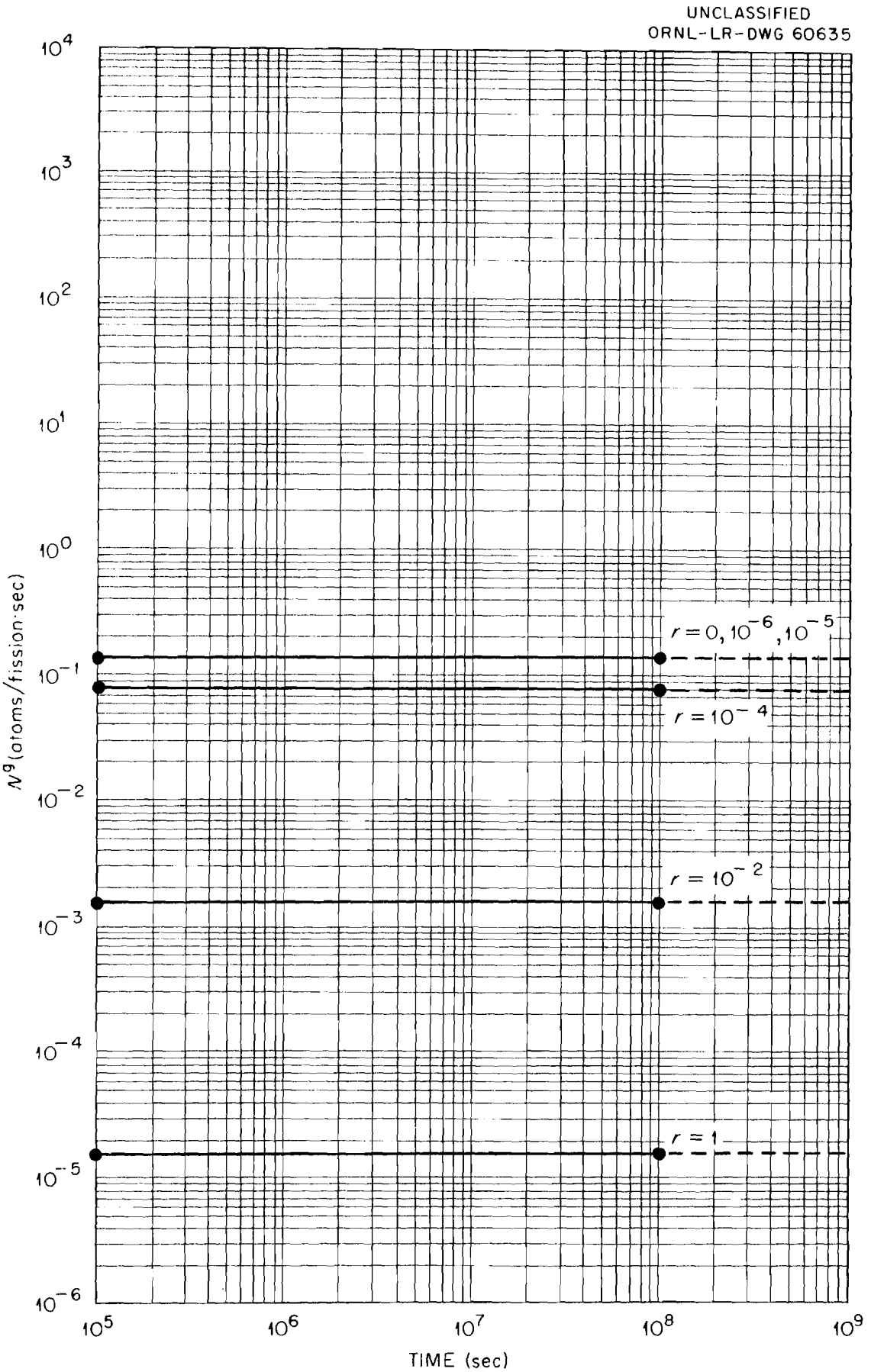

Fig. 18. Buildup of Fission Products in Coolant; I^{134}, Case I, $D' = 10^{-12}$, $r = 0$ to 1 (sec^{-1}).

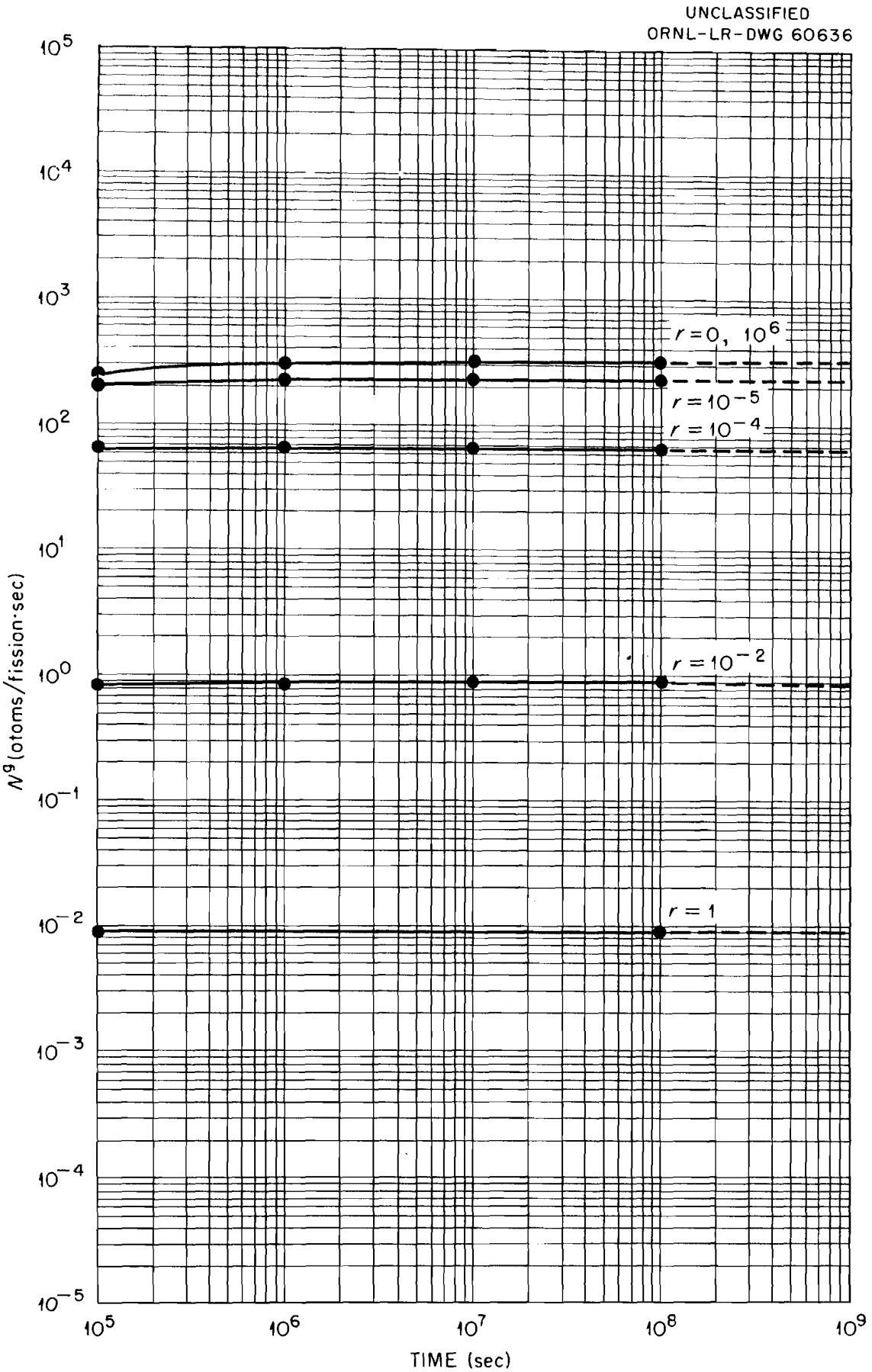

Fig. 19. Buildup of Fission Products in Coolant; I^{135}, Case I, $D' = 10^{-7}$, $r = 0$ to 1 (sec^{-1}).

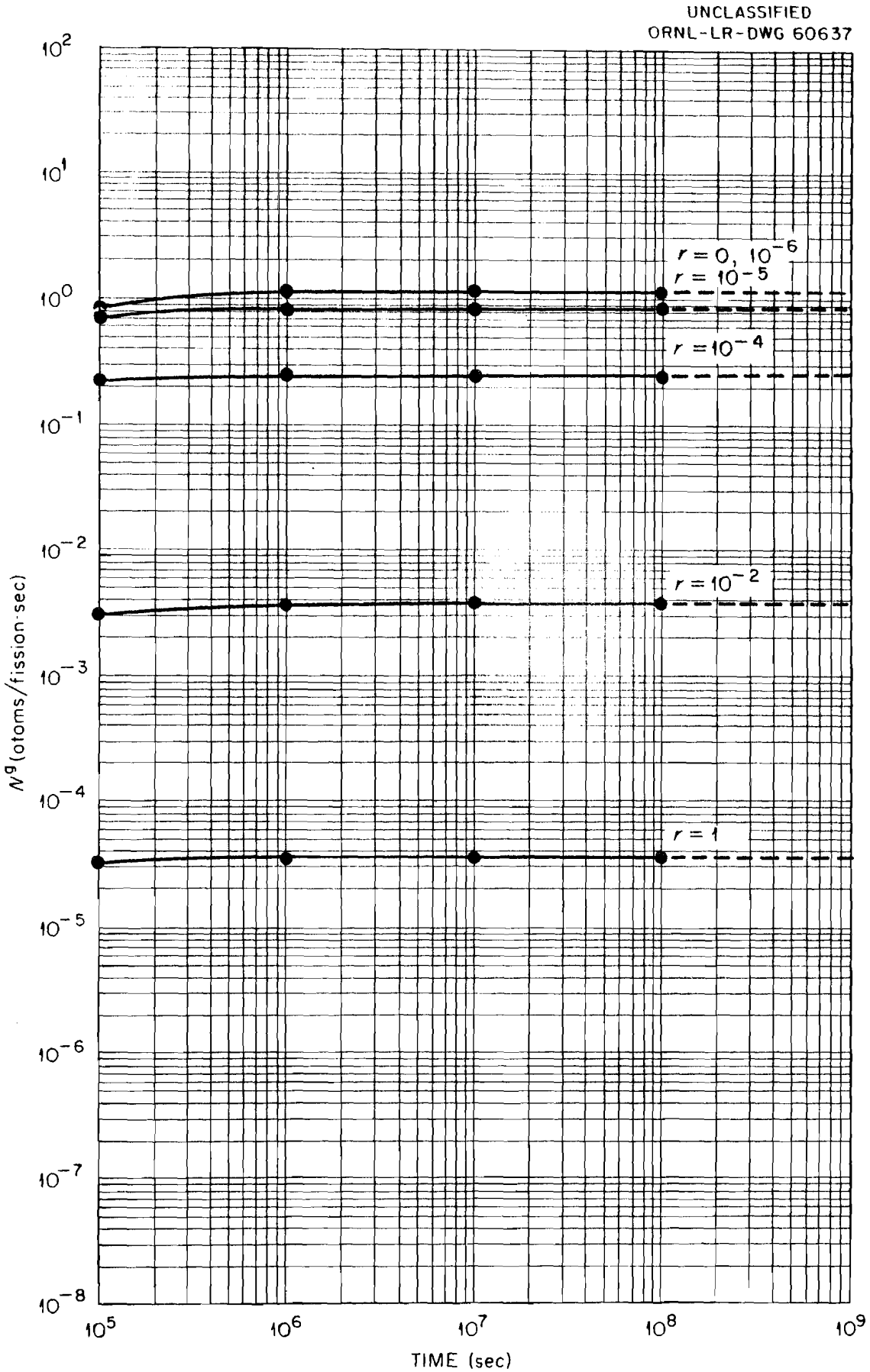

Fig. 20. Buildup of Fission Products in Coolant; I^{135}, Case I, $D' = 10^{-12}$, $r = 0$ to 1 (sec^{-1}).

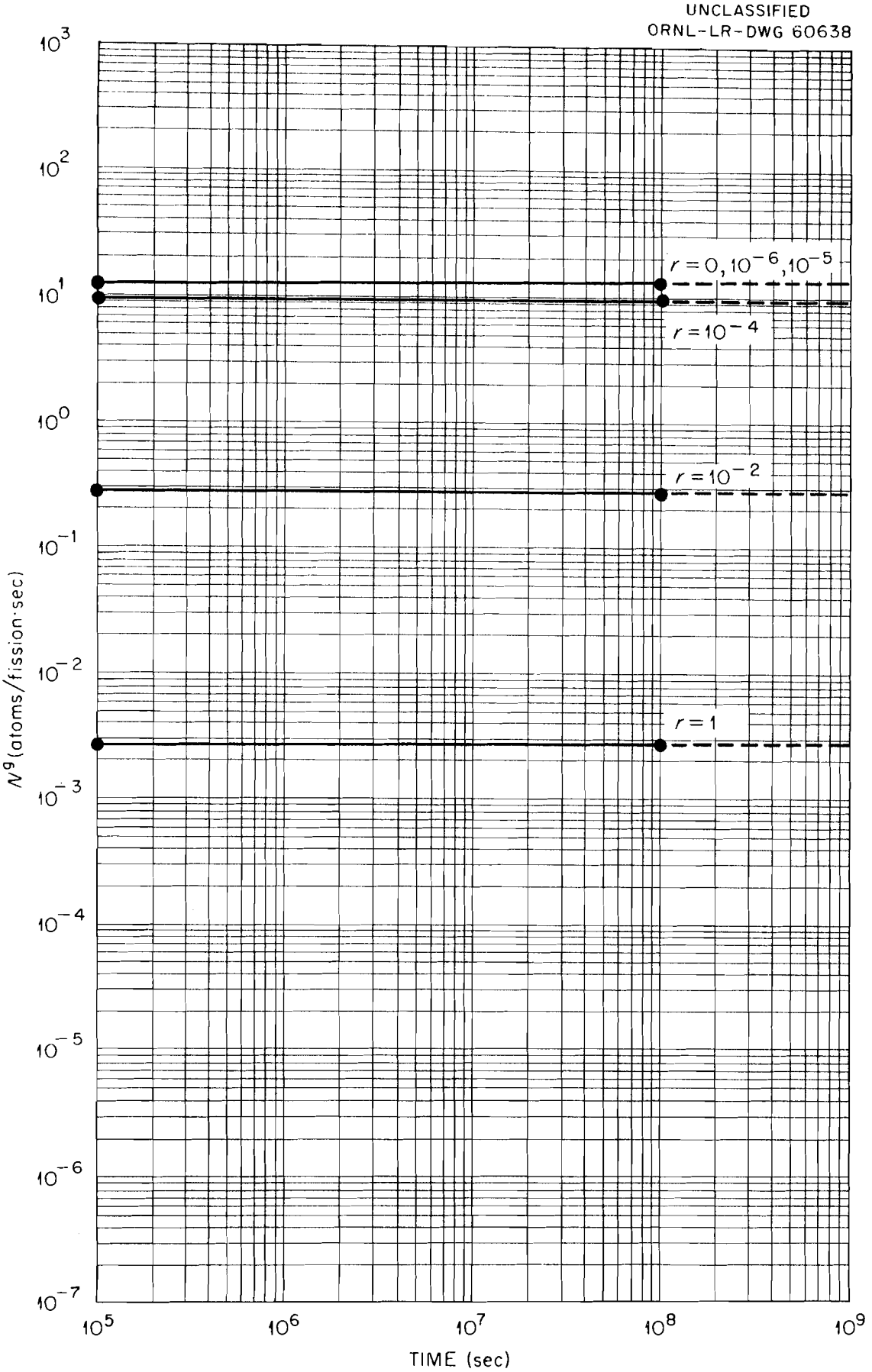

Fig. 21. Buildup of Fission Products in Coolant; Cs^{138}, Case I, $D' = 10^{-7}$, $r = 0$ to 1 (sec^{-1}).

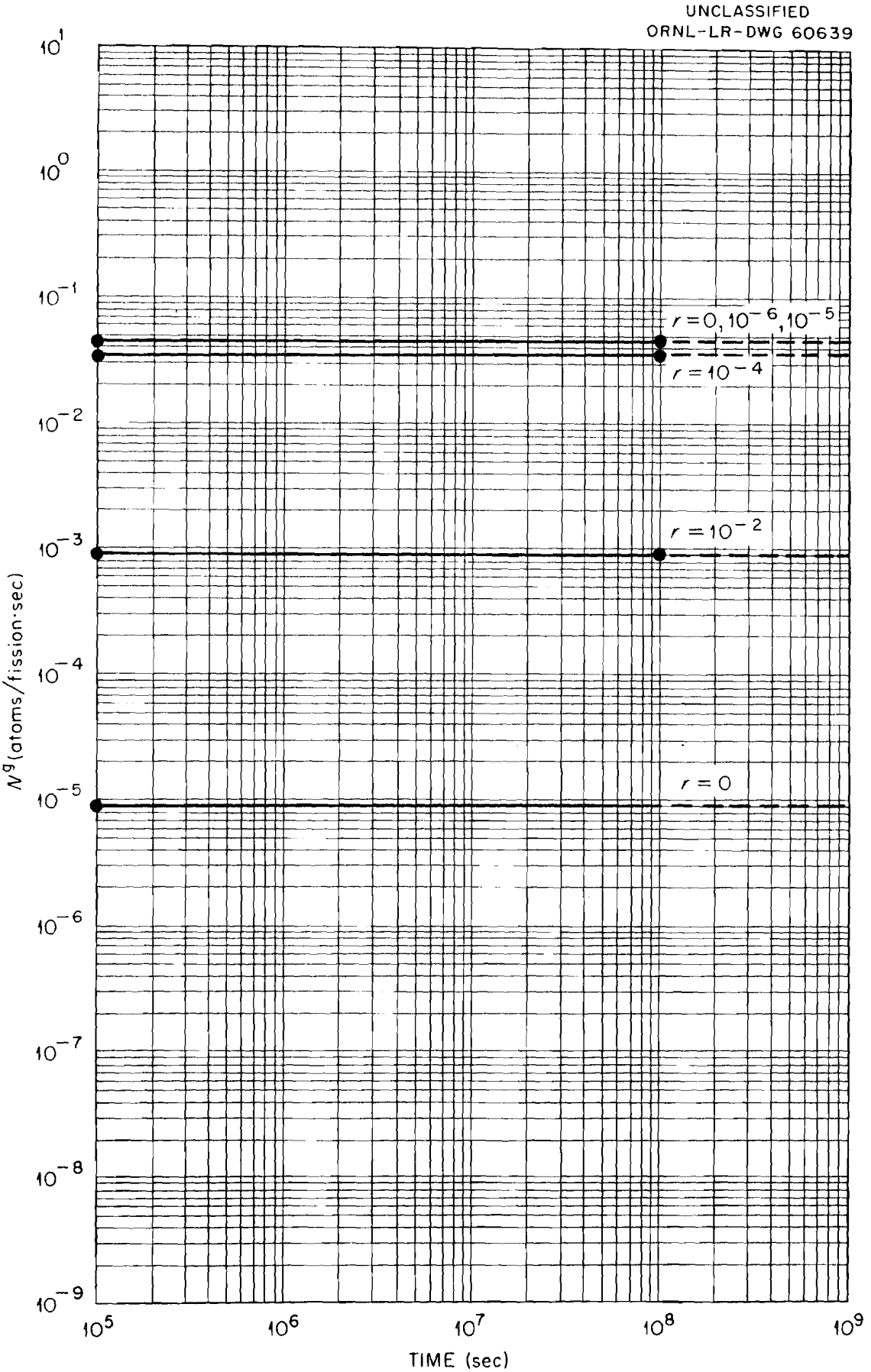

Fig. 22. Buildup of Fission Products in Coolant; Cs^{138}, Case I, $D' = 10^{-12}$ $r = 0$ to 1 (sec^{-1}).

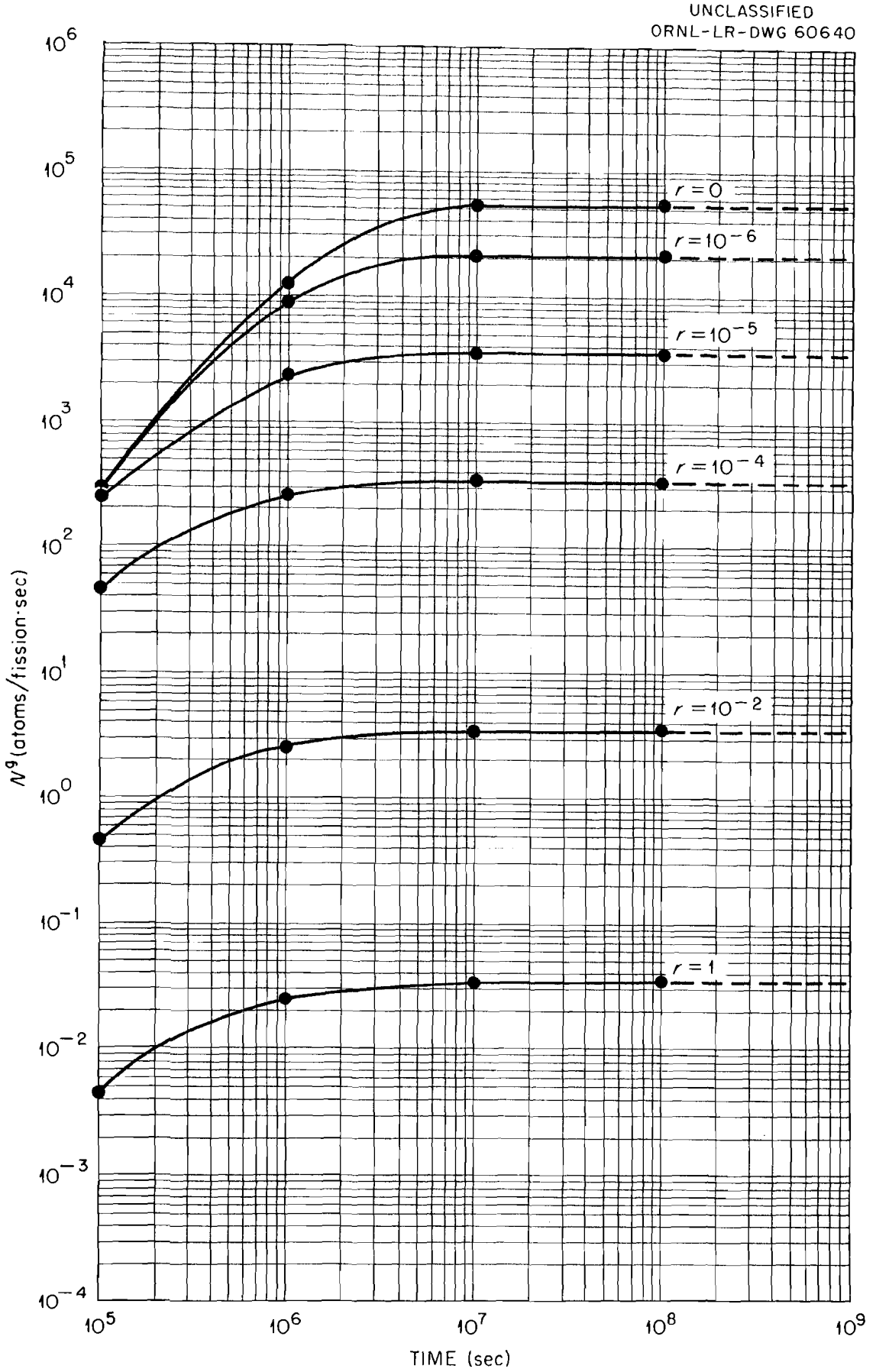

Fig. 23. Buildup of Fission Products in Coolant; Ba^{140}, Case I, $D' = 10^{-7}$, $r = 0$ to 1 (sec^{-1}).

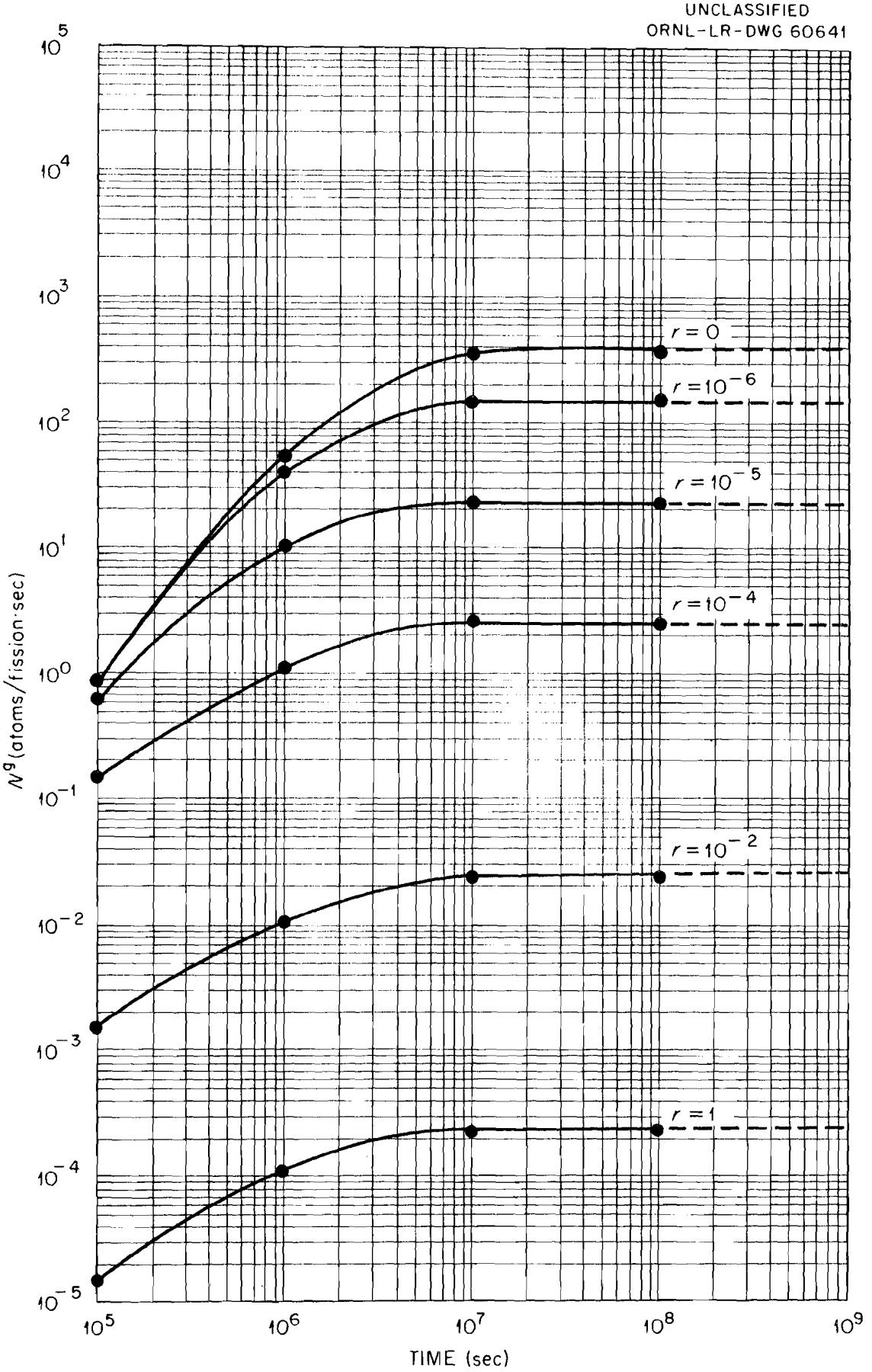

Fig. 24. Buildup of Fission Products in Coolant; Ba^{140}, Case I, $D' = 10^{-12}$, $r = 0$ to 1 (sec^{-1}).

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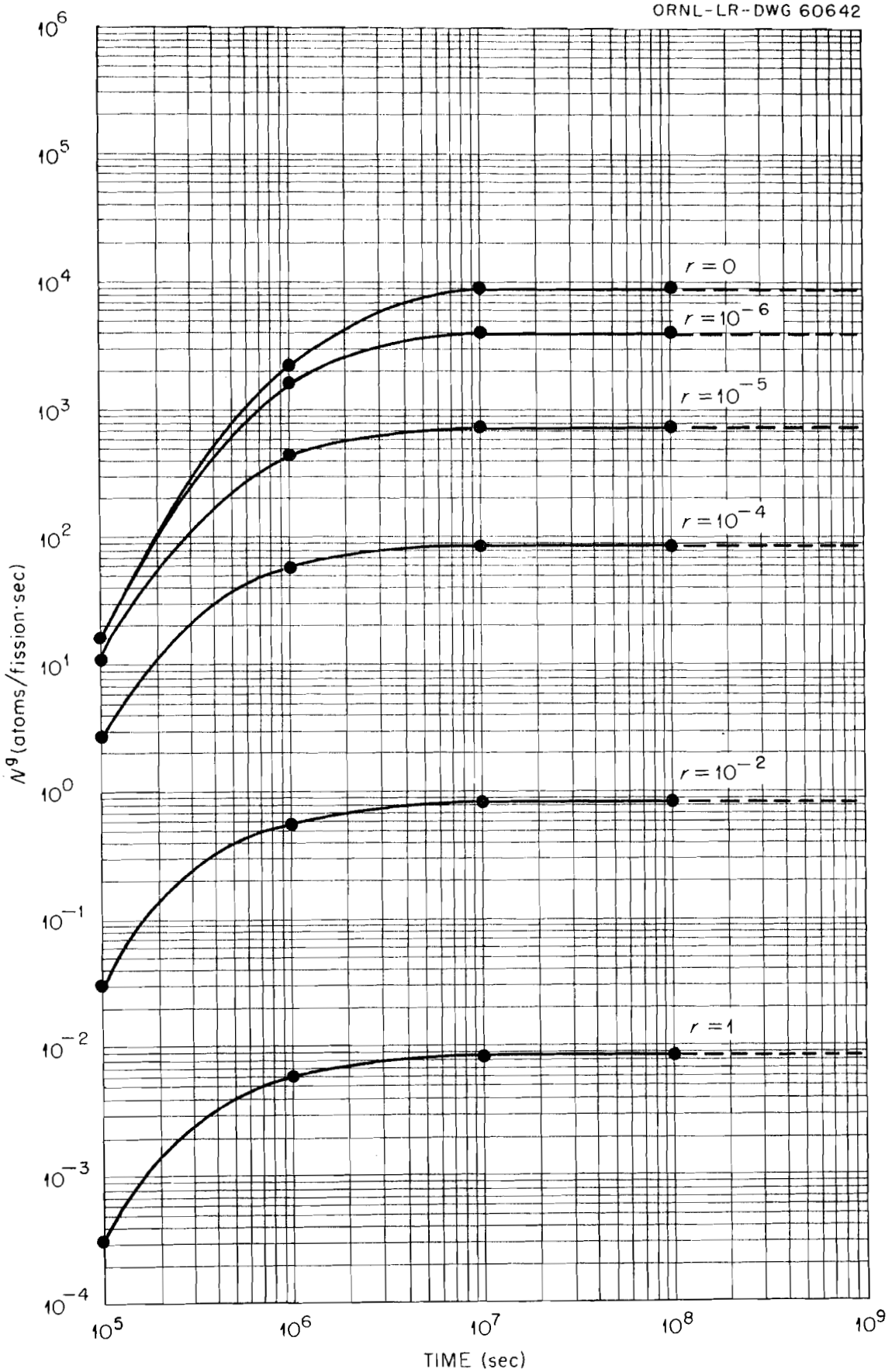


Fig. 25. Buildup of Fission Products in Coolant; La^{140}, Case I, $D' = 10^{-7}$, $r = 0$ to 1 (sec^{-1}).

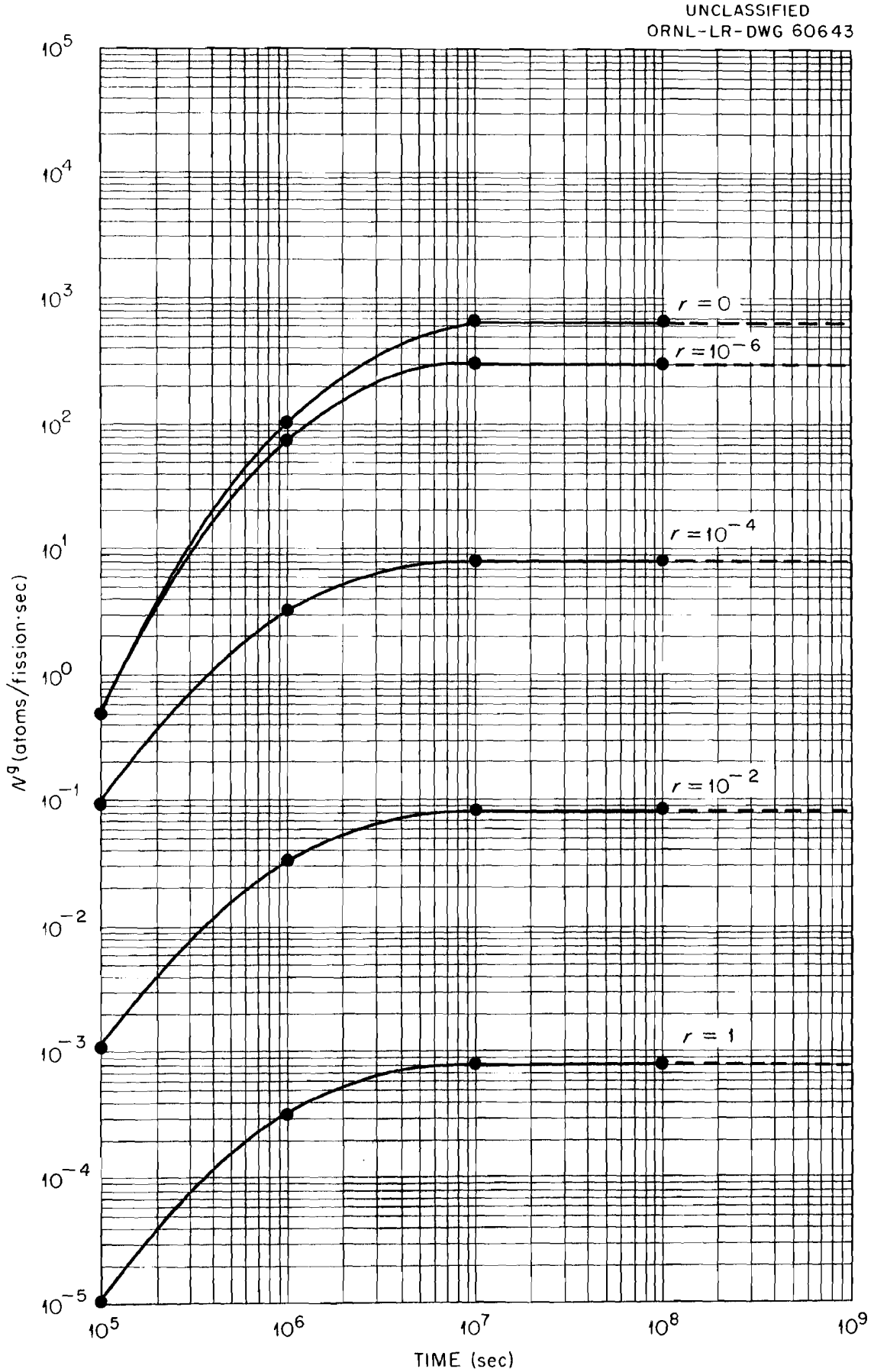


Fig. 26. Buildup of Fission Products in Coolant; La^{140}, Case I, $D' = 10^{-10}$, $r = 0$ to 1 (sec^{-1}).

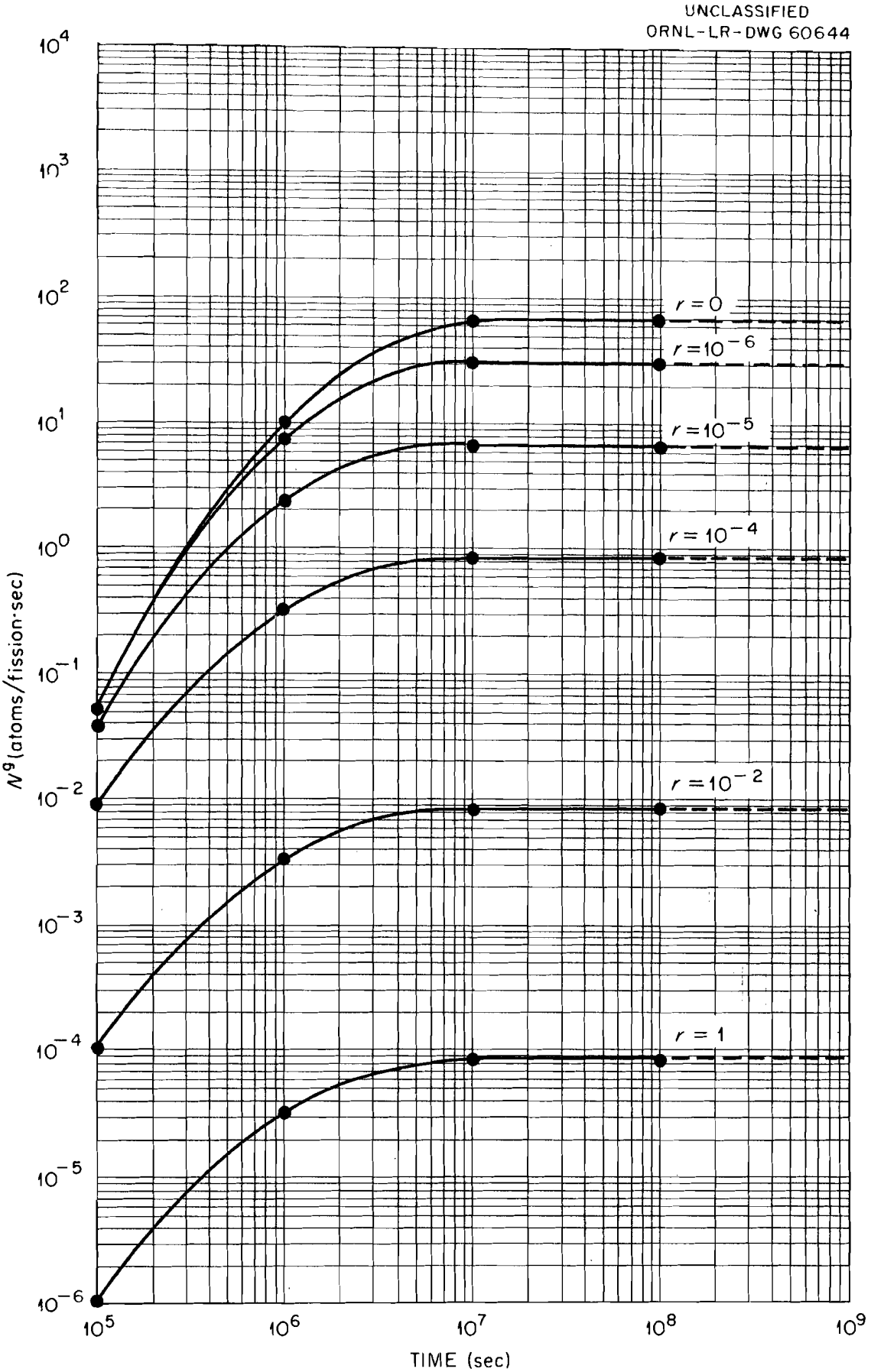


Fig. 27. Buildup of Fission Products in Coolant; La^{140}, Case I, $D' = 10^{-12}$, $r = 0$ to 1 (sec^{-1}).

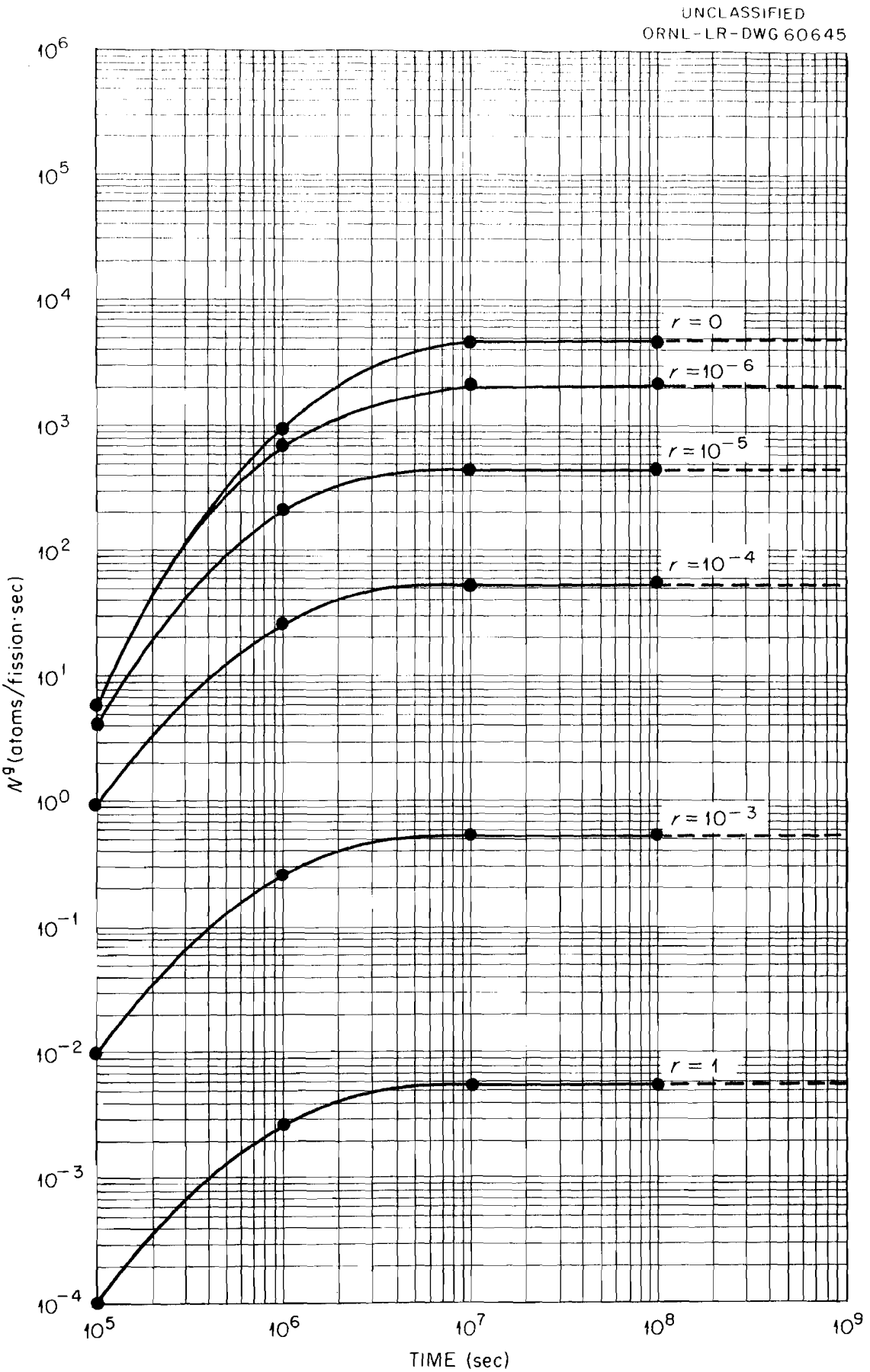
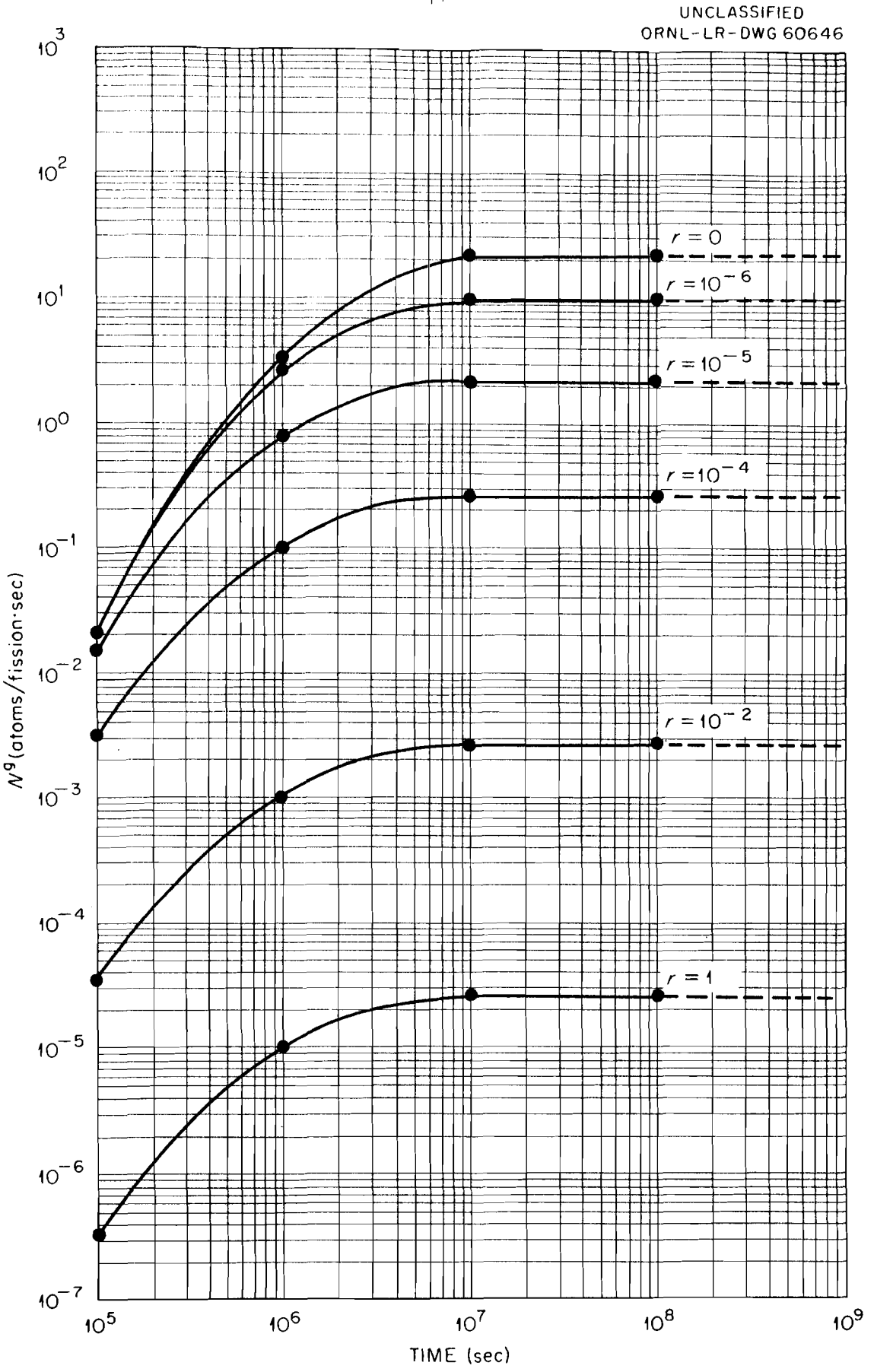


Fig. 28. Buildup of Fission Products in Coolant; La^{140}, Case II, $D_a' = 10^{-7}$, $r = 0$ to 1 (sec^{-1}).



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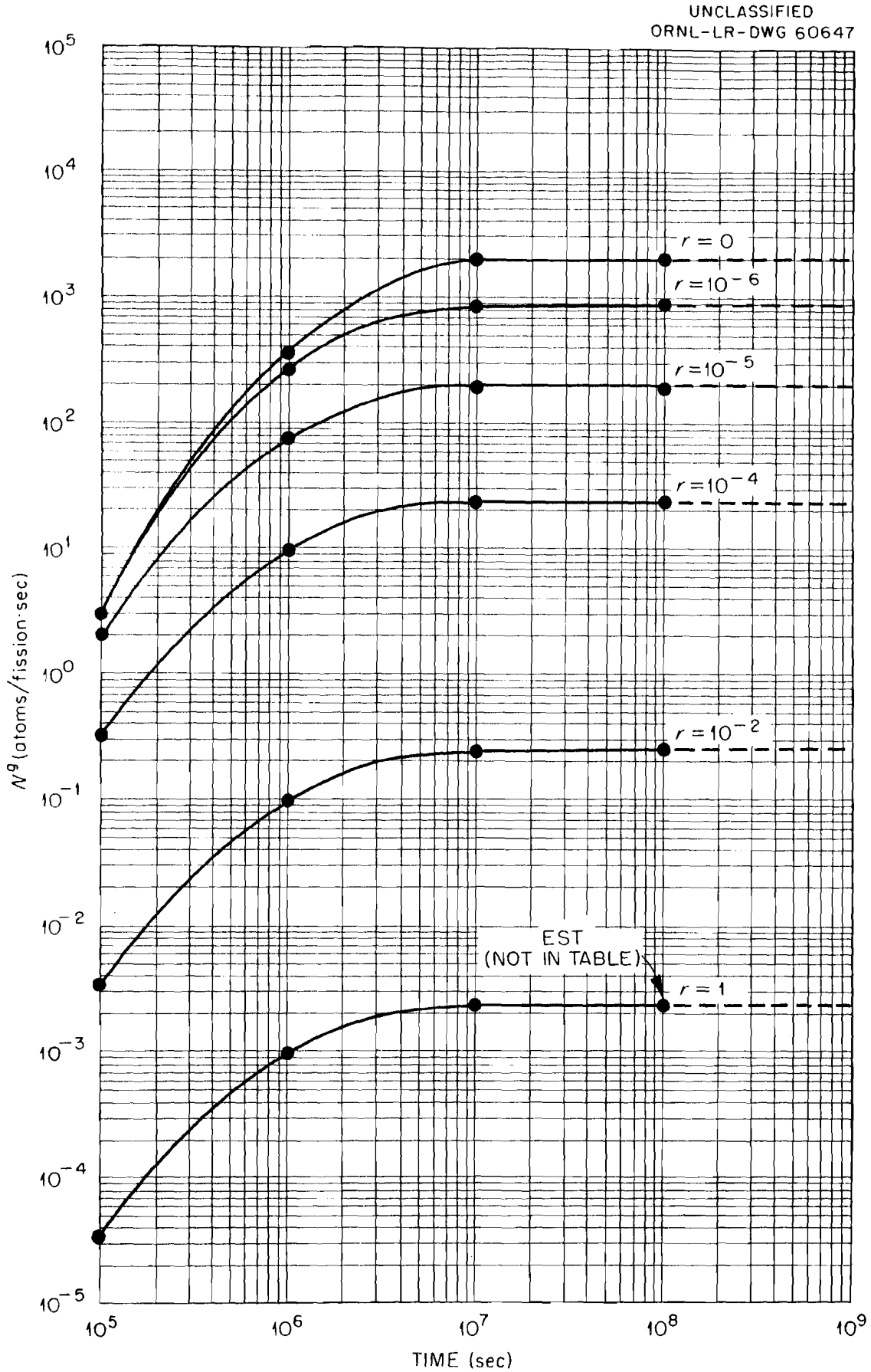

Fig. 30. Buildup of Fission Products in Coolant; La^{140}, Case III, $D_a' = 10^{-7}$, $r = 0$ to 1 (sec^{-1}).

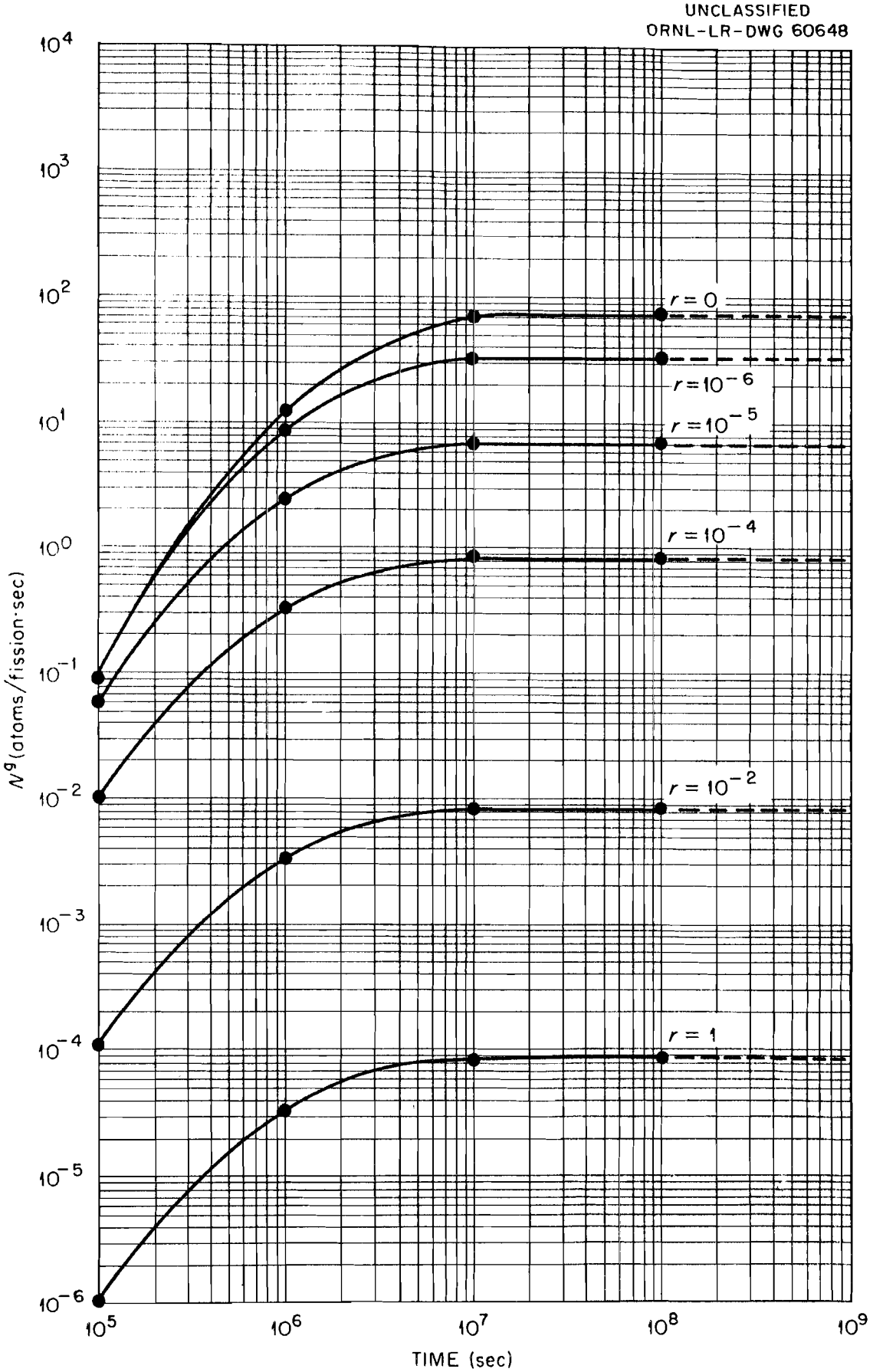

Fig. 31. Buildup of Fission Products in Coolant; La^{140}, Case III, $D_a' = 10^{-10}$, $r = 0$ to 1 (sec^{-1}).

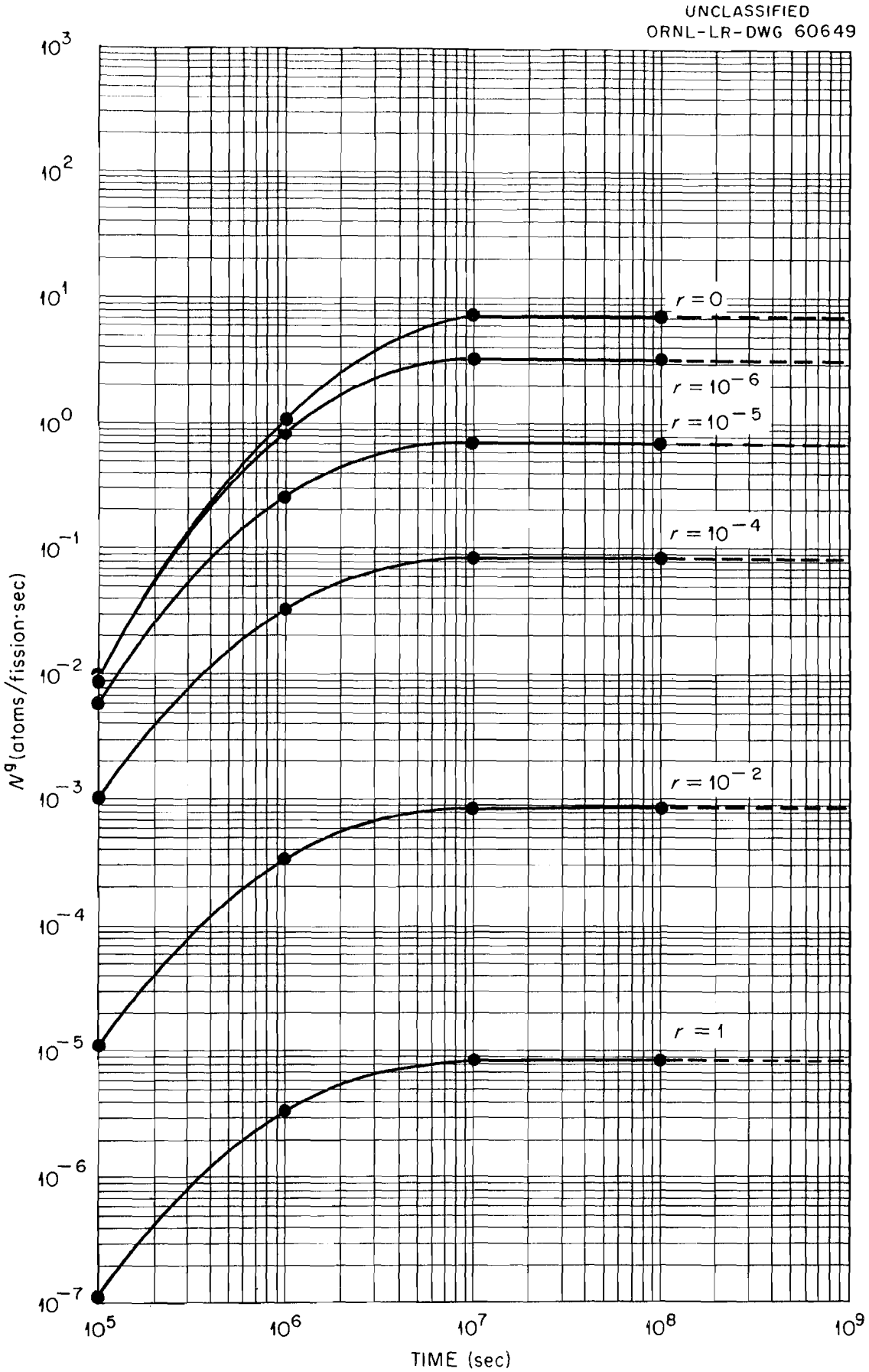

Fig. 32. Buildup of Fission Products in Coolant; La^{140}, Case III, $D_a' = 10^{-12}$, $r = 0$ to 1 (sec^{-1}).

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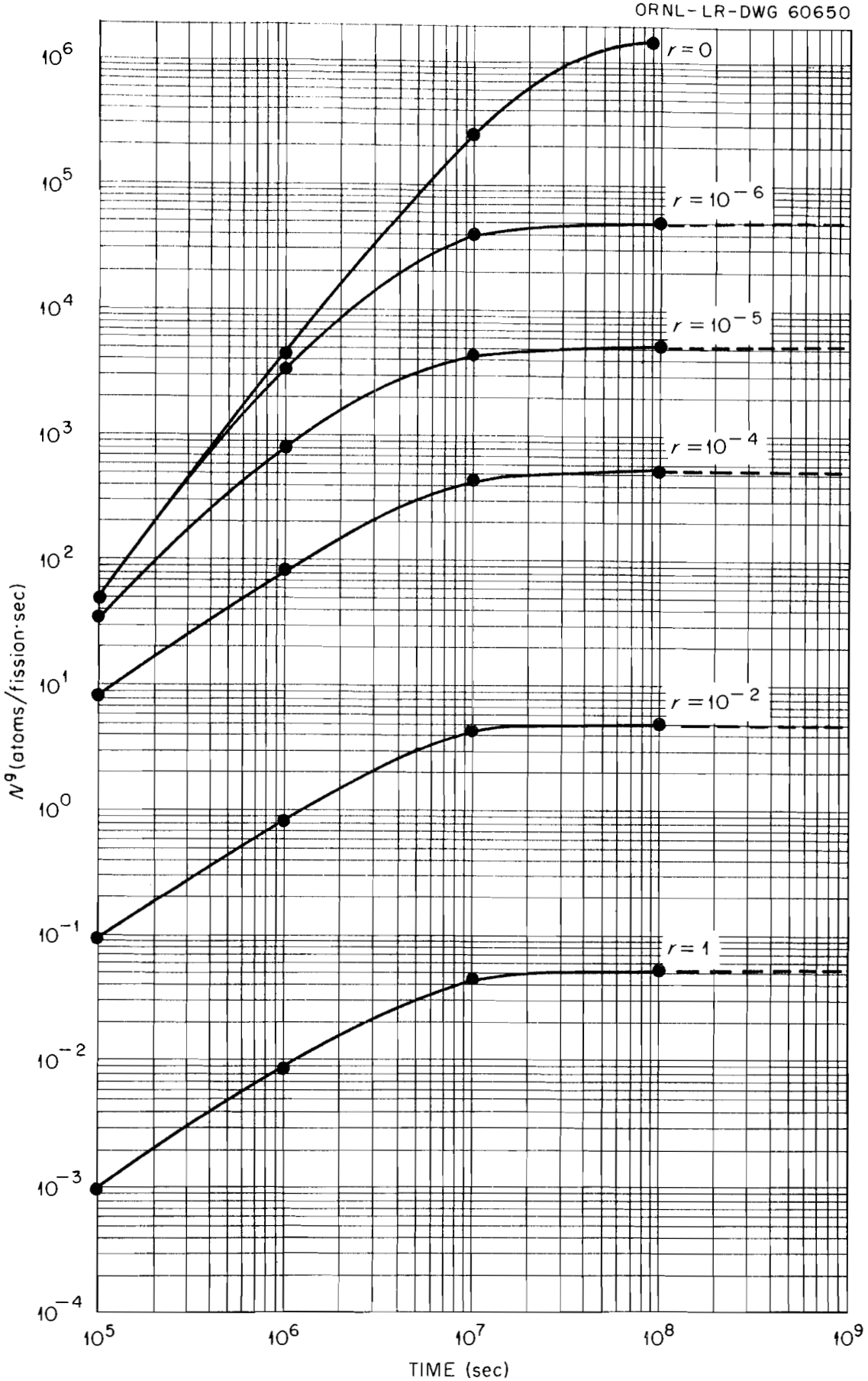

Fig. 33. Buildup of Fission Products in Coolant; Ce^{144}, Case I, $D' = 10^{-7}$, $r = 0$ to 1 (sec^{-1}).

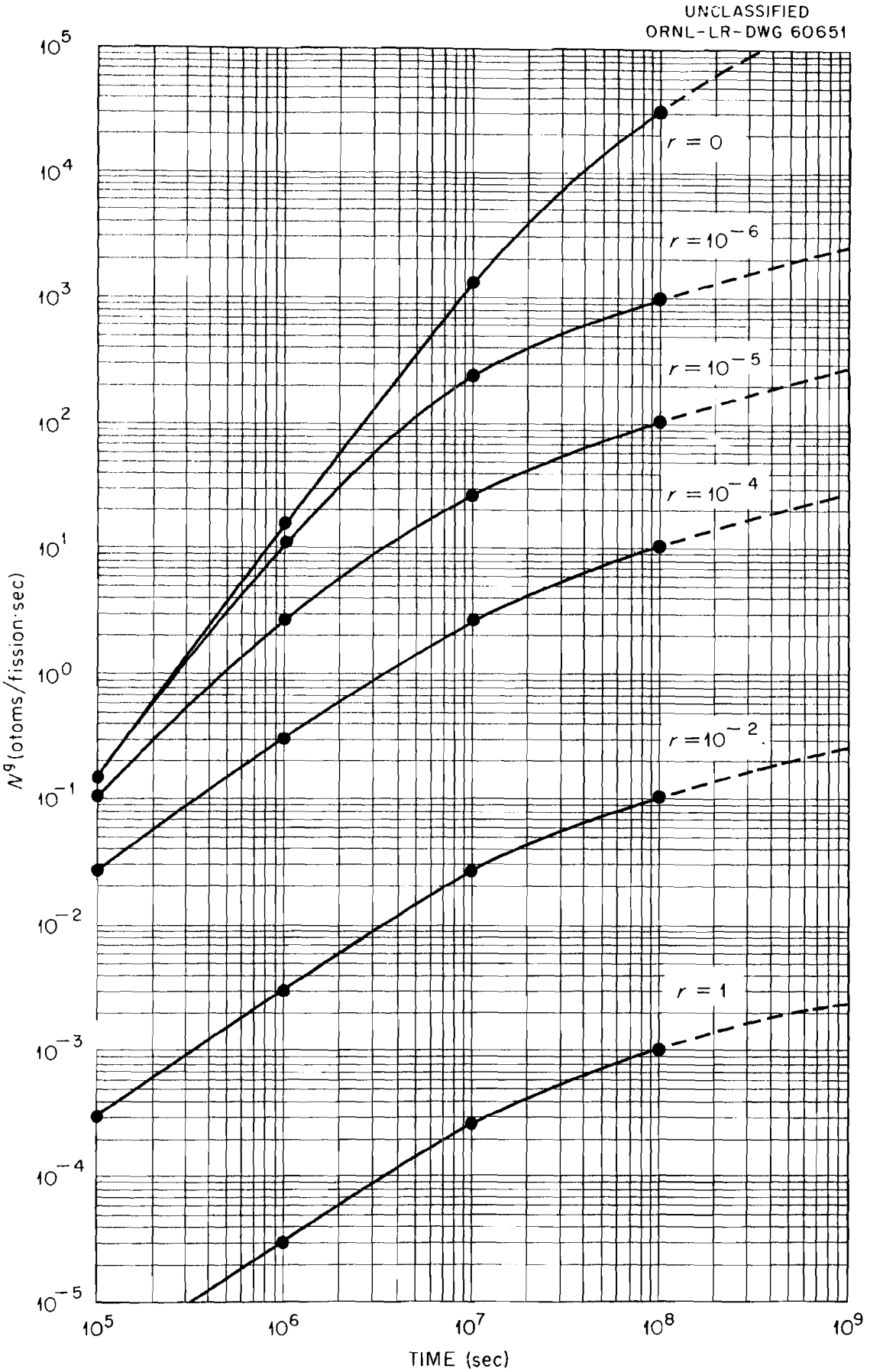

Fig. 34. Buildup of Fission Products in Coolant; Ce^{144}, Case I, $D' = 10^{-12}$, $r = 0$ to 1 (sec^{-1}).

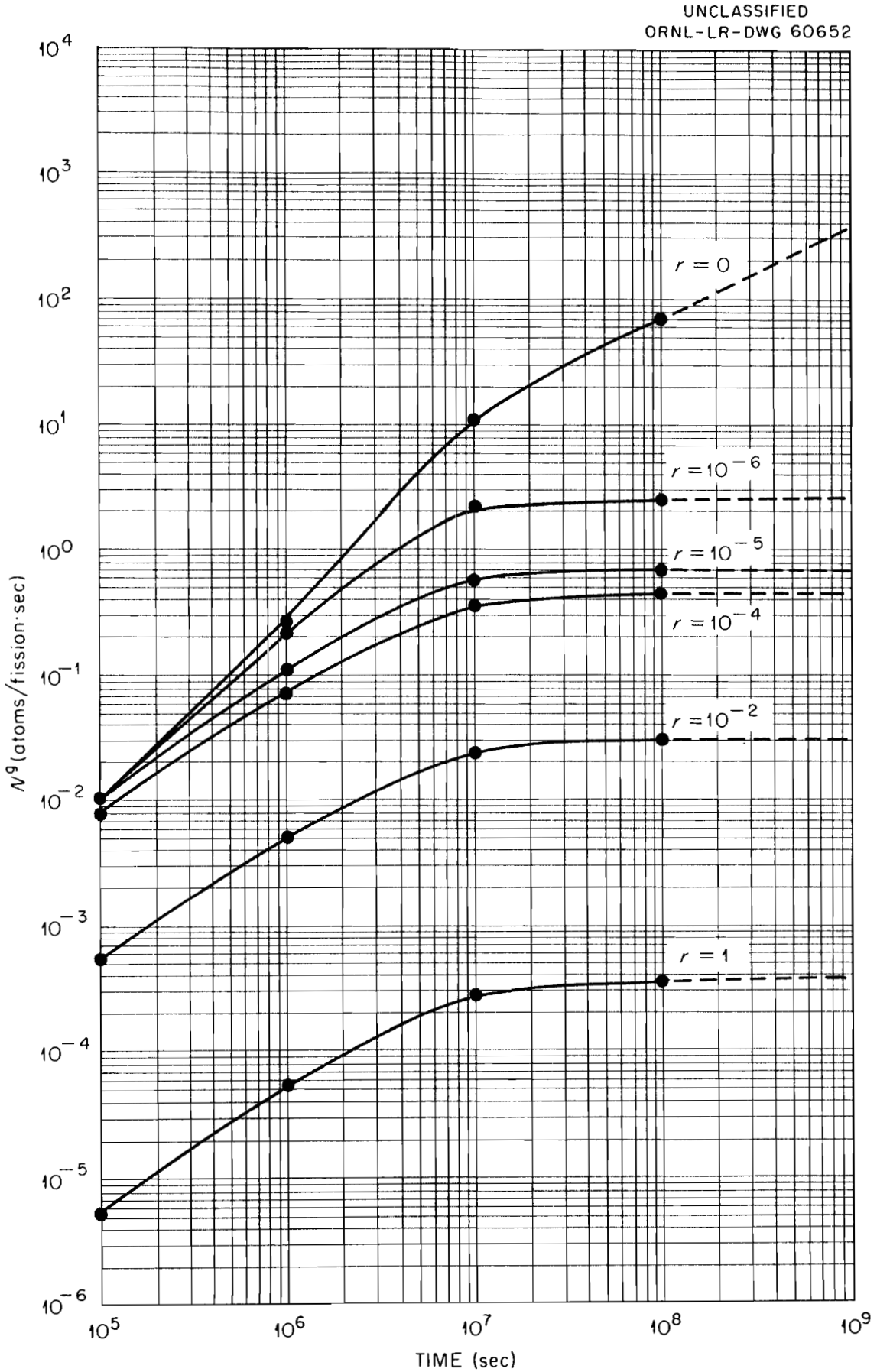

Fig. 35. Buildup of Fission Products in Coolant; Pr^{144}, Case I, $D' = 10^{-7}$, $r = 0$ to 1 (sec^{-1}).

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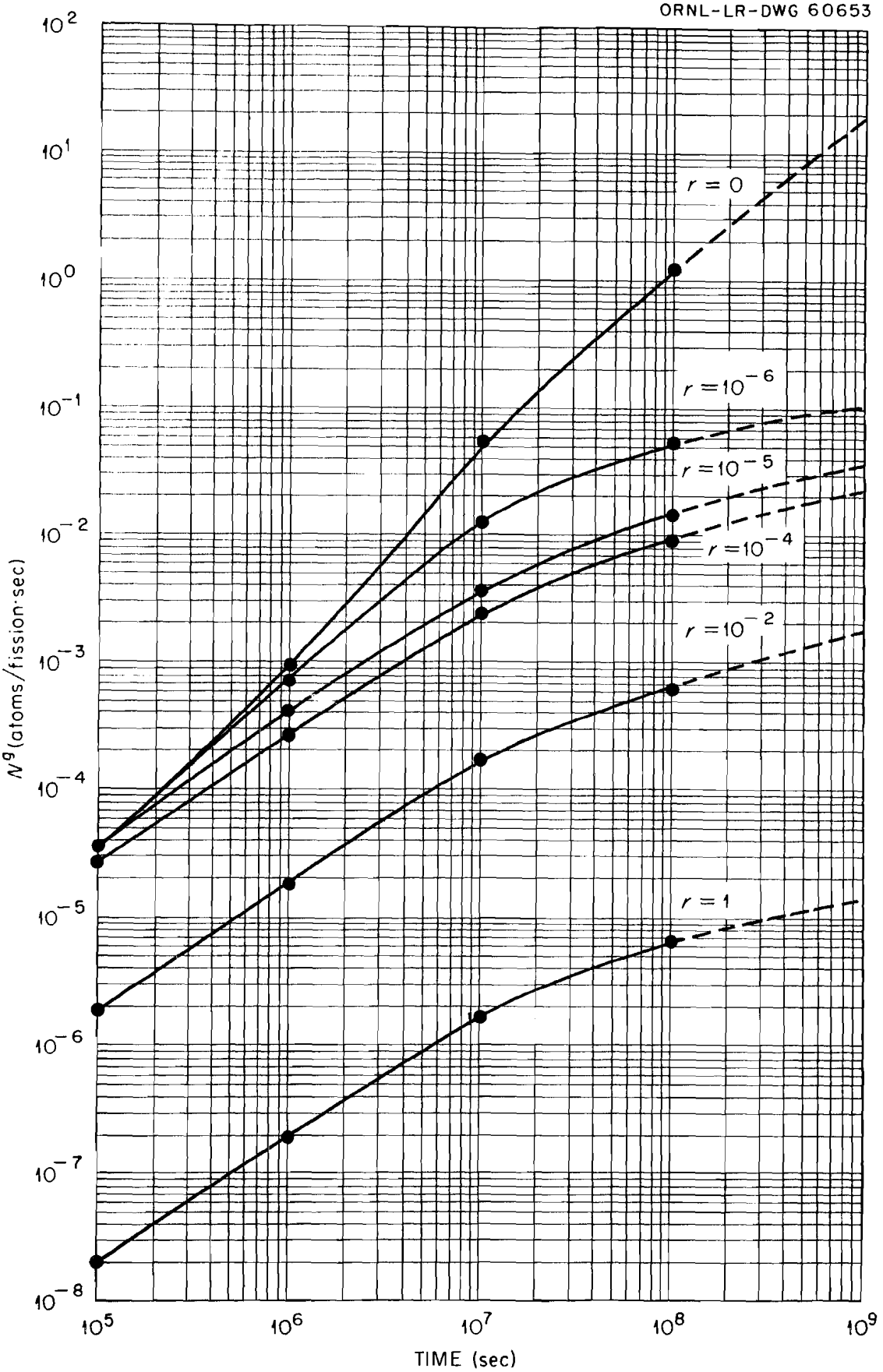

Fig. 36. Buildup of Fission Products in Coolant; Pr^{144}, Case I, $D' = 10^{-12}$, $r = 0$ to 1 (sec^{-1}).

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