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Dragon Project Report

FISSION PRODUCT RETENTION IN H.T.R. FUEL ELEMENTS

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Fission Product Retention in H.T.R. Fuel Elements

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ABSTRACT

The retention of metallic fission products by the structural parts of Dragon-type fuel elements can be very much greater than that of the fuel, because the distance to be travelled by every escaping atom is millimetres instead of microns.

For experiments lasting up to six months this apparent failure of strontium and barium in particular to penetrate fuel tubes significantly has led to some remarkably clean coolant circuits Designers need to know how the overall release of fission products is likely to vary with time and assessment studies demand some sort of model for fission product transport, which will permit extrapolation to longer times.

This interim report describes an attempt to apply the equations describing heat flow through a slab into a region at zero temperature to measurements of radial fission product concentration profiles and coolant circuit activity from Pluto Loop A experiments

We have examined the retention of Group I, II and III fission products in the structural components of Pluto Loop A Fuel Elements. The fission product retention in the fuel tubes and fuel free zones has been compared with that in the fuel particles. For strontium and barium in particular the retention was good in the times involved The problem is to know whether this retention will last the life-time of a fuel element, and under what conditions the fuel tube remains useful as a method of fission product retention. At the same time assessment studies demand that one can make some sort of model of the f p. transport, so that extrapolation to longer times is possible.

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1.0 Introduction

The Pluto Loop A series of irradiations has demonstrated that in times up to six months for burn-up up to 6% FIMA H.T.R. fuel elements release only small fractions of their fission products into the coolant (1-6).

The General Atomic GAIL loop programme has extended this low releasing period to 500 days$^{(7)}$, which approaches the life of a "feed" element in a feed and breed fuel cycle. However, reactor designers wish to predict with certainty the releases of fuel elements after from 5 to 15 years irradiation, to burn-ups approaching 15% FIMA.

In this paper this problem is treated as the estimation of how long a non-equilibrium situation, fission products in a leaky can, may be expected to exist.

2.0 The Fuel Tube as a Fission Product Delay Line

The fuel tube, or fuel free zone, plays a large part in the delay of metallic fission products because it is ~100 times thicker than a particle coating and because every atom that leaves the fuel must pass through it to reach the coolant. Table 1 shows examples of the fuel tubes preventing fission products which have left the fuel from entering the coolant.

To explore the time dependance of this virtue and to permit comparisons between experiments of different length a simple model of fuel tube behaviour based on heat flow in a slab was made.

A constant flux of atoms, Fo, enters from one side of a parallel sided slab; from the opposite side evaporation occurs at a rate proportional to concentration (C_1). Initially the slab is empty; at equilibrium

$$C_1 = Fo/H$$

and the concentration gradient between faces is linear

At this stage no attempt has been made to match the input rate to the rate of release from the fuel. This is being done by Dragon using far more elegant mathematics(8). Our justification is that a constant flux represents the worst case and calculations(10) show that the rate of release from a particle approaches a constant value after $\sim 10^{4}$ seconds for the diffusion coefficient being discussed.

A far more tenuous assumption is that D has single value for the range of concentrations (and hence times) under discussion, and that the diffusing species remains the same. D is known to vary with concentration rapidly at (10) concentrations of the order of 5×10^{18} atoms cm^{-3} (1 mg cm^{-3}) above which it is typically 10^{-5} or 10^{-6} for a group IIA metal at 1000°C(11). The measured concentration profiles show that this concentration region has not been reached for strontium and barium and all group III fission products.

Finally the preliminary study has been confined to stable nuclides. Radioactive decay will produce steeper concentration gradients for a given diffusion coefficient.

Rather than discuss purely hypothetical situations we have attempted to relate our calculated concentration profiles to some profiles measured on old Pluto Loop A samples. The constants obtained have then been used to calculate the behaviour of fuel elements after different times.

3.0 Calculated Concentration Profiles

Radial concentration profiles of ^{137}Cs, ^{90}Sr, ^{155}Eu and ^{144}Ce are being measured at Harwell and we have used some data obtained by Paul at Seibersdorf(12).

The estimation of H the evaporation coefficient is difficult. Evaporation has been assumed to be confined to that region of the fuel tube within 100°C of the maximum temperature, unless otherwise stated, and over that region the rate has been assumed uniform. This makes it possible to associate a release measurement with a profile in this region.

Examination of typical profiles shows that H can only be determined from a profile if $H \ll D$, unless D is very large (10^{6}) when $H < D$ yields profiles from which

H can be found if Fo is known. In all other circumstances H cannot be deduced from concentration profiles measured after relatively short times, unless the number of fission product atoms released (per cm^2-sec) is known. (See Fig. 1.) A plot of fractional release from the fuel tube ($\int_0^T$ Ho dt/Fo) against D after time T is given in Fig. 2 for various values of H.

4.0 Extrapolation to Long Irradiation Times

Fig.3 shows how fission product concentration varies with time at the coolant surface of our model element. The dangers of extrapolation from experiments lasting less than 10^7 seconds are apparent.

Short half-life nuclides will come to equilibrium at a rate determined primarily by their half-lives. However, rapid diffusion of ^{140}Ba over stable ^{138}Ba must not be excluded at high ^{138}Ba concentrations.

The absolute values of Cxt are artbitrary, in fact they lie above experimentally determined values.

Figs.5 and 6 illustrate the changes that would occur in the radial fission product concentration profile after times up to 10 years for D = 10^{-7} and 5×10^{-9}. Again the arbitrary input rate needs to be stressed, because a 100-fold reduction in Fo would reduce all concentrations by a corresponding factor and what is perhaps more important, increase the time taken to reach any critical concentration at which D changes due to a change in diffusion mechanism(12). This reduction in Fo is best made as close to the fuel as possible by introducing a barrier such as silicon carbide The barrier must not exhibit concentration dependant diffusion or if it does it must contain only a very small number of defect paths.

5.0 Fuel Tube Thickness

Fig.4 compares a 1.25 mm fuel tube with one 8.4 mm thick.

In the thin tube D needs to be very small before there is any increase in useful delay with decreasing D The longer equilibrium transit time of the thicker tube produces far lower fractional releases after short times Comparison of the releases to be expected from a 1.25 mm tube after 1.5×10^7 secs (Pluto 8A) with those to be expected from an 0.84 mm tube after 5×10^6 secs (Pluto 4A) explains why, other things being equal, Pluto 8A fuel free zone released such a large fraction of its fission products. The temperature difference between the two tubes would account for only a factor of 10 if the activation energy for evaporation were 40 k cal mole^{-1}, and 100 if it were 80 k cal mole^{-1}

6.0 Fuel Tube Temperature

To represent the effect of temperature by only a heat of evaporation is an oversimplification because the temperature variation of D and perhaps Fo would have been ignored. For reasons described above large changes in fission product release could therefore result from modest temperature reductions, provided a thick enough fuel tube were used.

7.0 Summary

The role of the fuel element structure in fission product retention has been outlined.

Neglect of these arguments can lead to the production of misleadingly low assessments of fission product release after very long times.

The study of radial concentration profiles in Dragon fuel tubes and particles will enable the approach to release equilibrium to be monitored, but it will not yield releases from individual elements since H will be difficult to measure, this is a task for a loop or better a reactor in which all fuel elements are identical.

TABLE 1

Fractional Release from Fuel Tubes and Fuel Free Zones

Nuclide	Fractional Release			
	Charge No. 3	4	6	8
134 Cs	4×10^{-1}	2×10^{-1}		.95
137 Cs	2×10^{-1}	2×10^{-1}	1	.97
89 Sr	2×10^{-4}	1×10^{-5}	5×10^{-2}	.25
90 Sr	3×10^{-4}	2×10^{-4}	1×10^{-2}	.29
140 Ba	5×10^{-3}	2×10^{-3}	6×10^{-2}	.045
91 Y	1×10^{-3}	3×10^{-2}	2×10^{-2}	.12
141 Ce		5×10^{-1}	2×10^{-2}	.12
144 Ce	2×10^{-2}	2×10^{-1}		.05
131 I	2×10^{-2}	1×10^{-2}	3×10^{-2}	1

TABLE 2

Diffusion and Evaporation Coefficients obtained by Comparing Observed and Calculated Concentration Profiles

Nuclide	Temperature °C	D	H
137 Cs	1000	10^{-7}	10^{-7}
90S Sr	1000	5×10^{-9}	-
140 Ba^{+}	1150	2×10^{-10}	-

$^{+}$Radioactivity not taken into account

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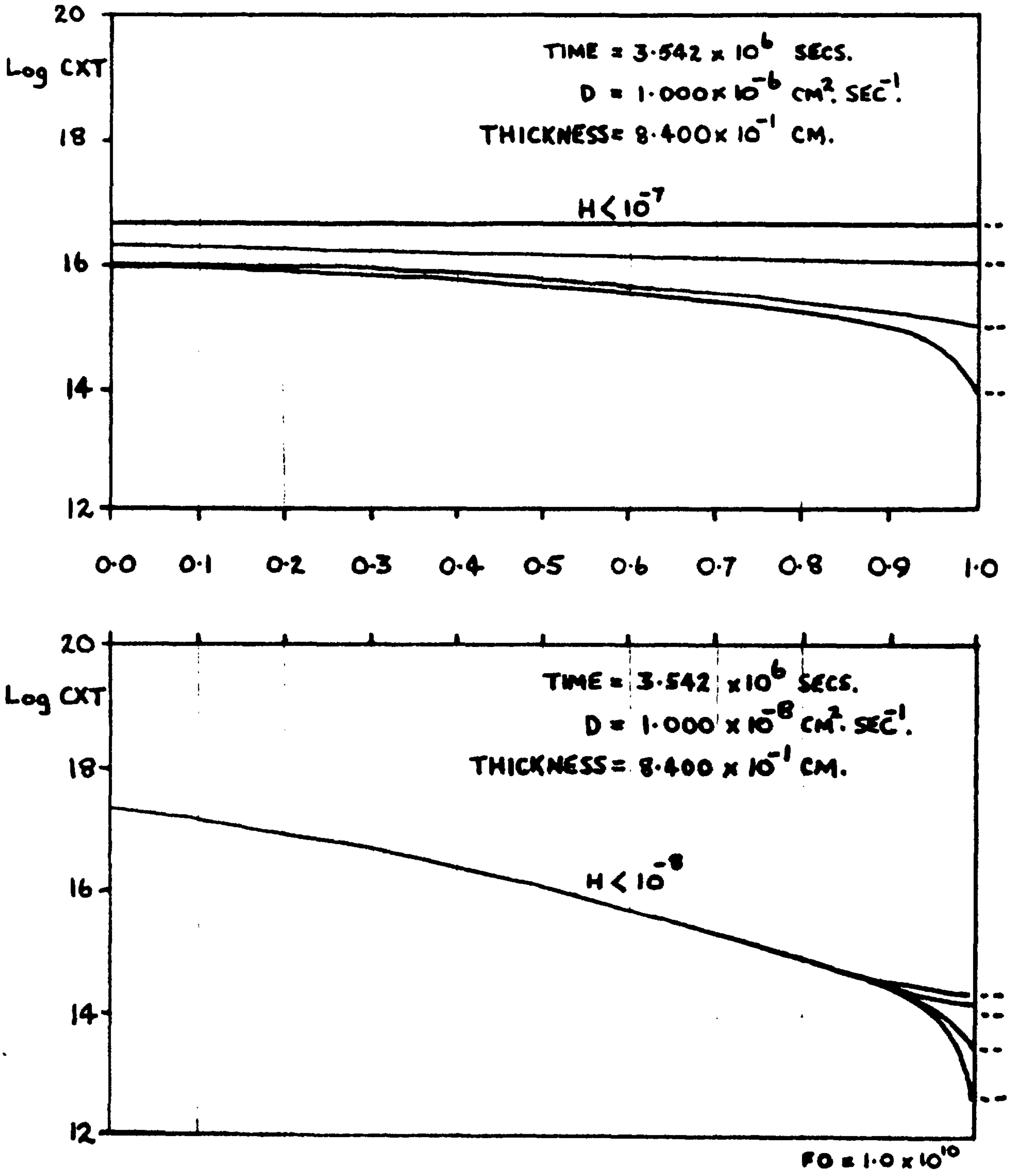
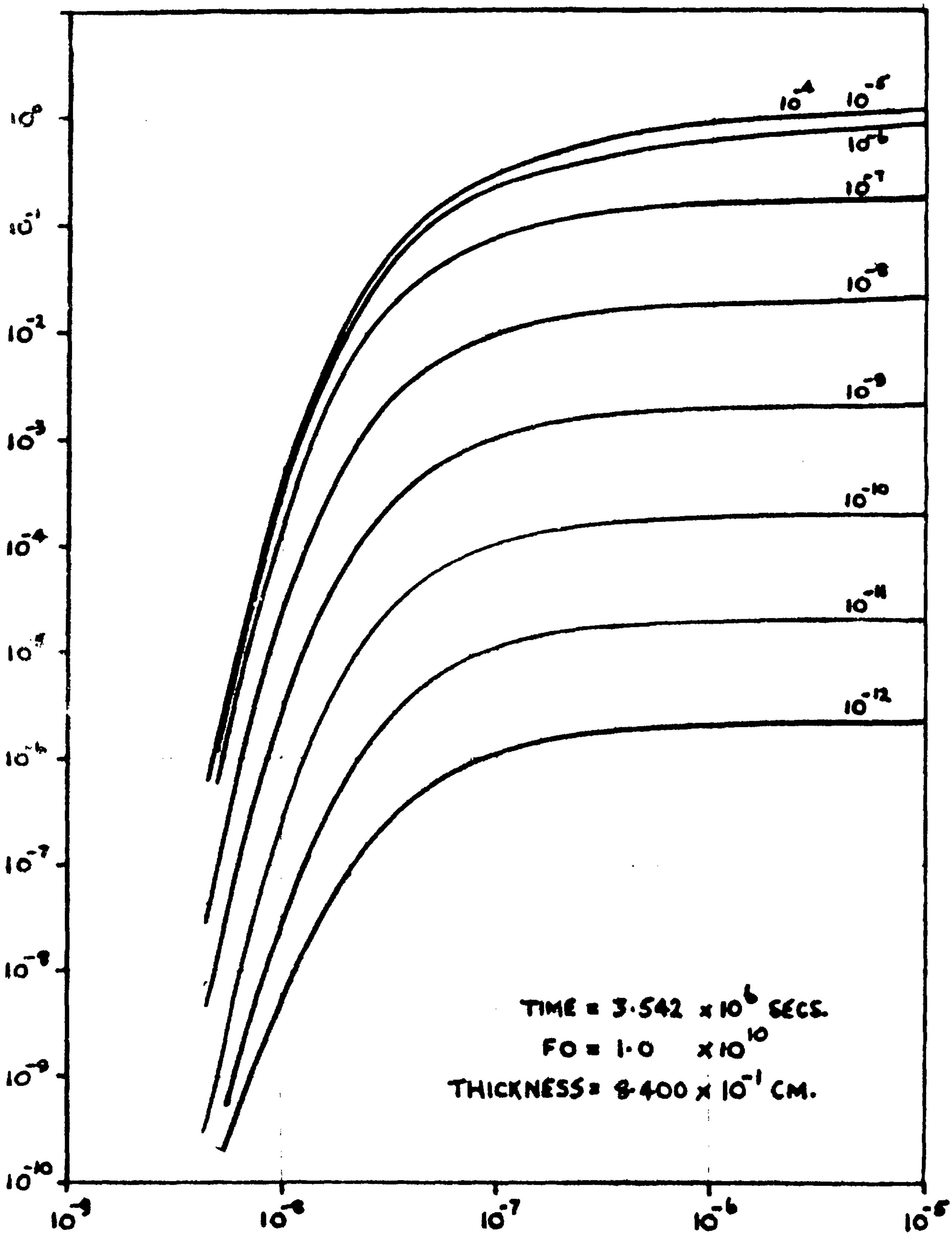
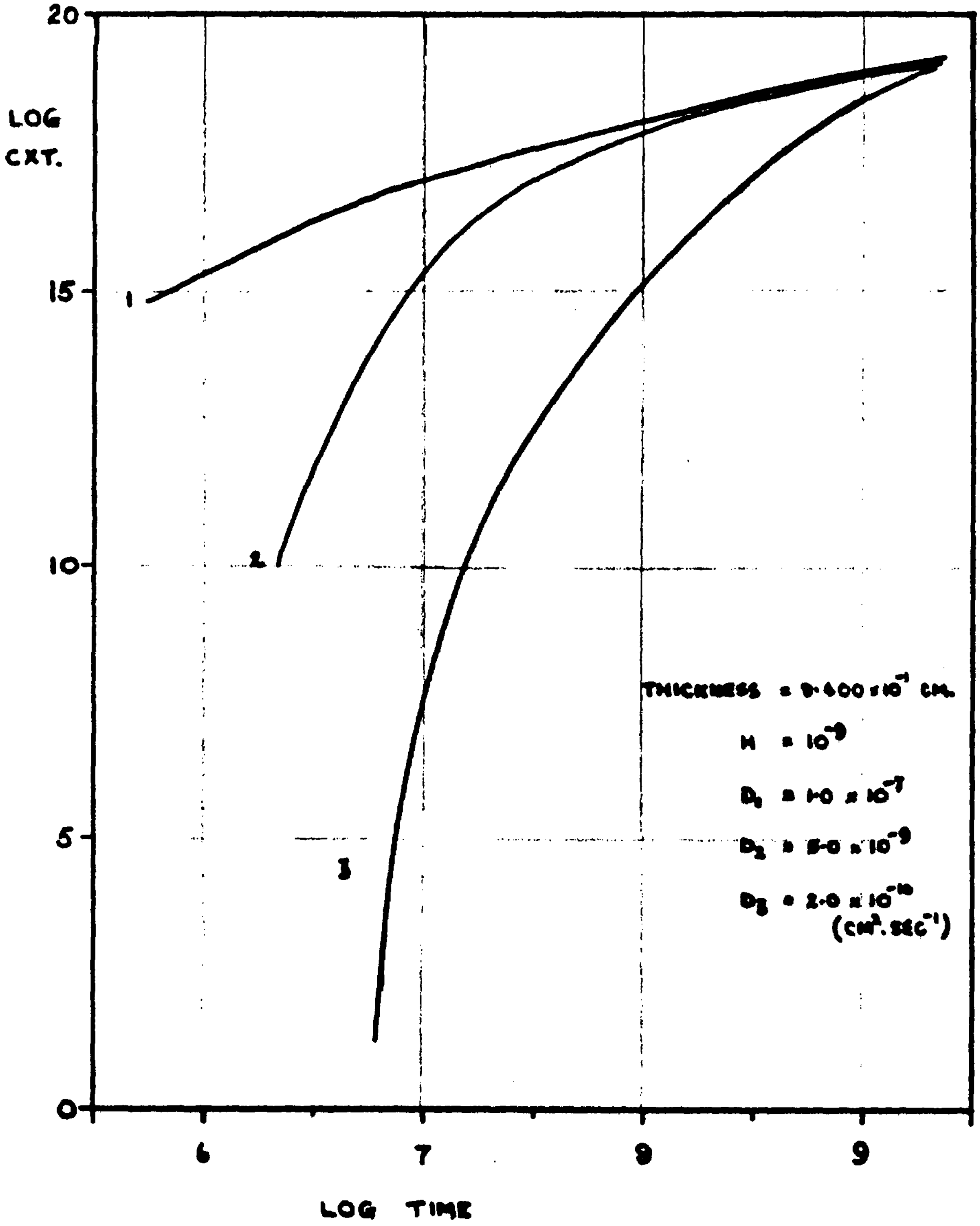


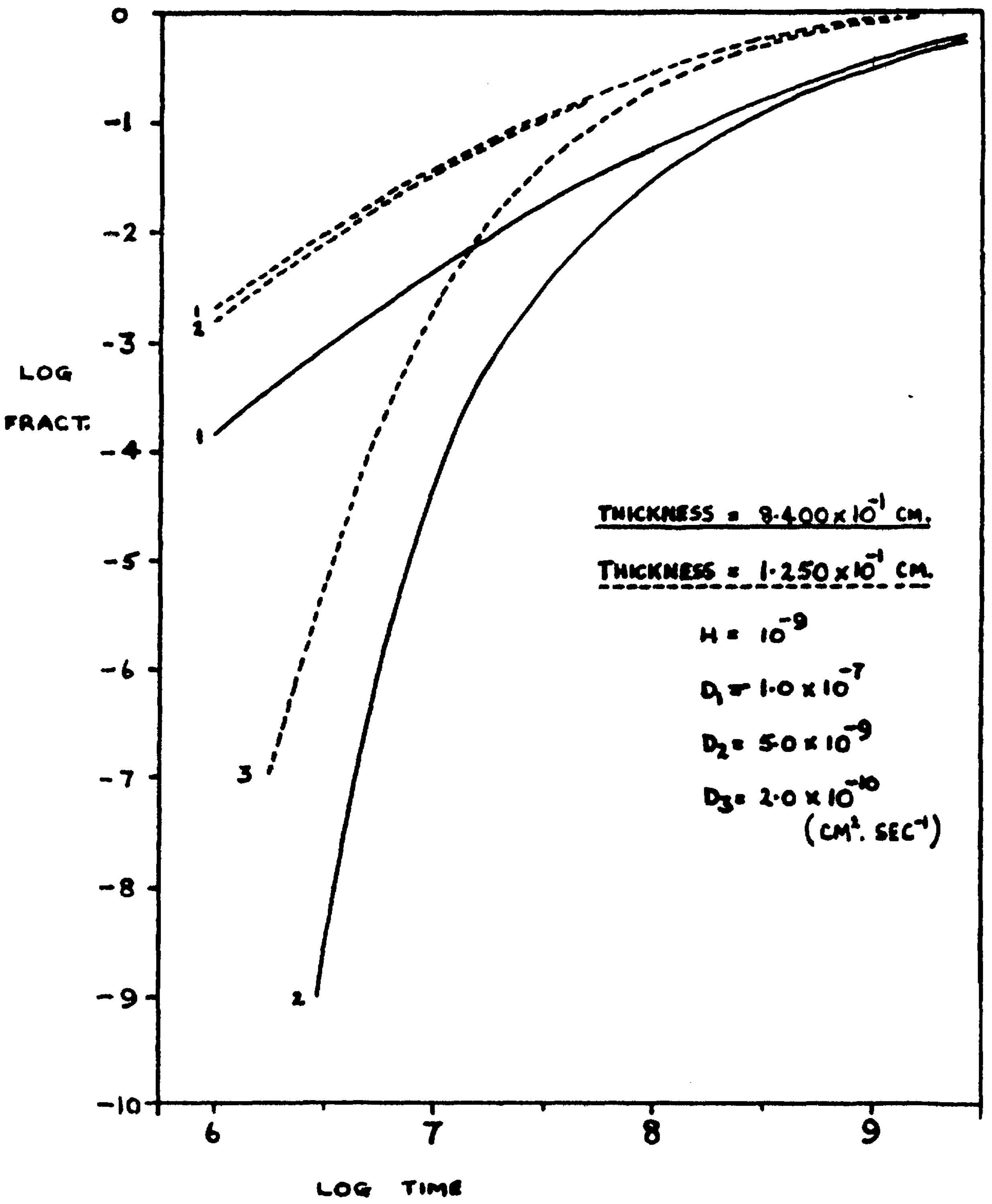
AERE-M1723 FIG. 1 CALCULATED CONCENTRATION PROFILES



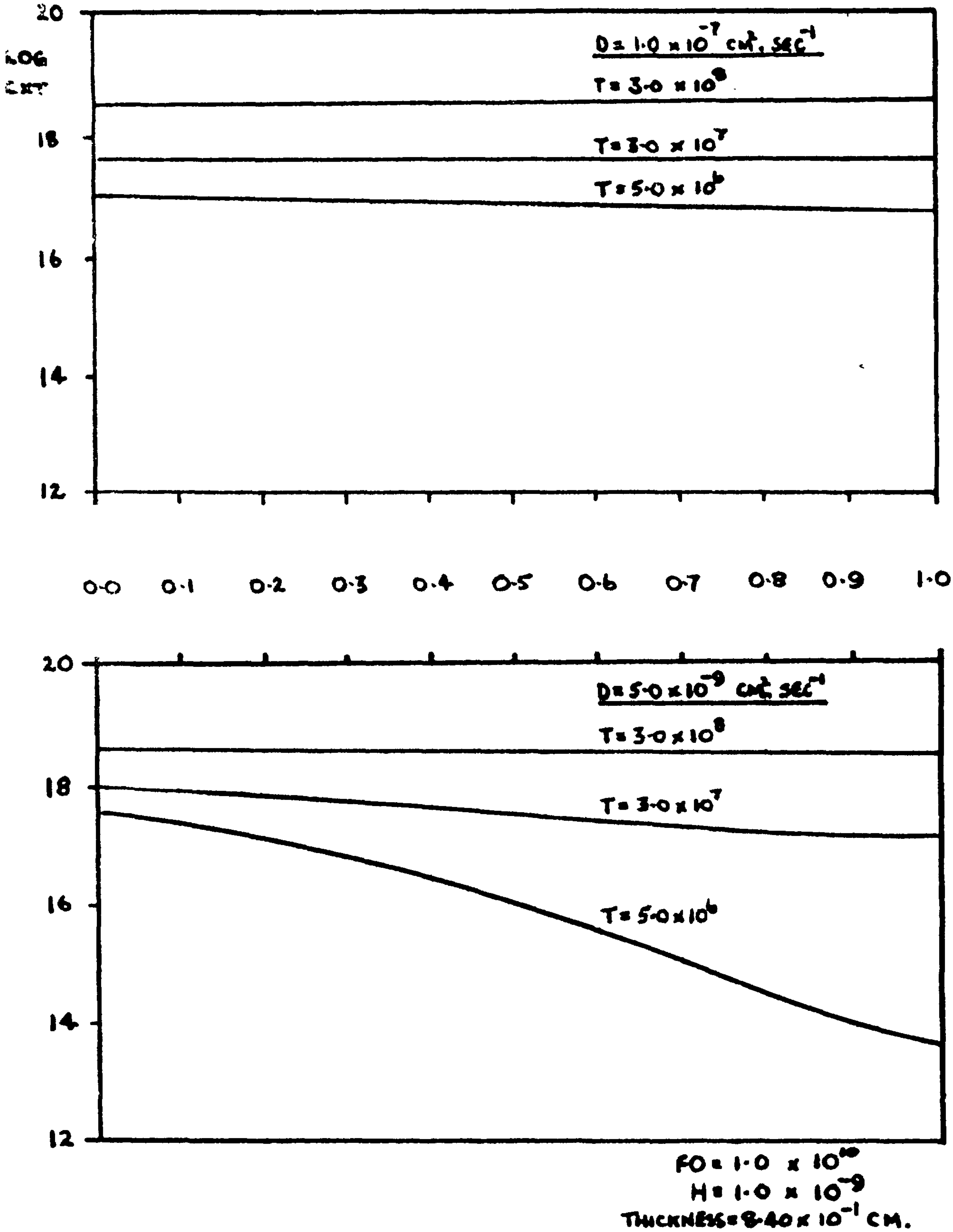
AERE-M1723 FIG.2 FRACTIONAL RELEASE v D



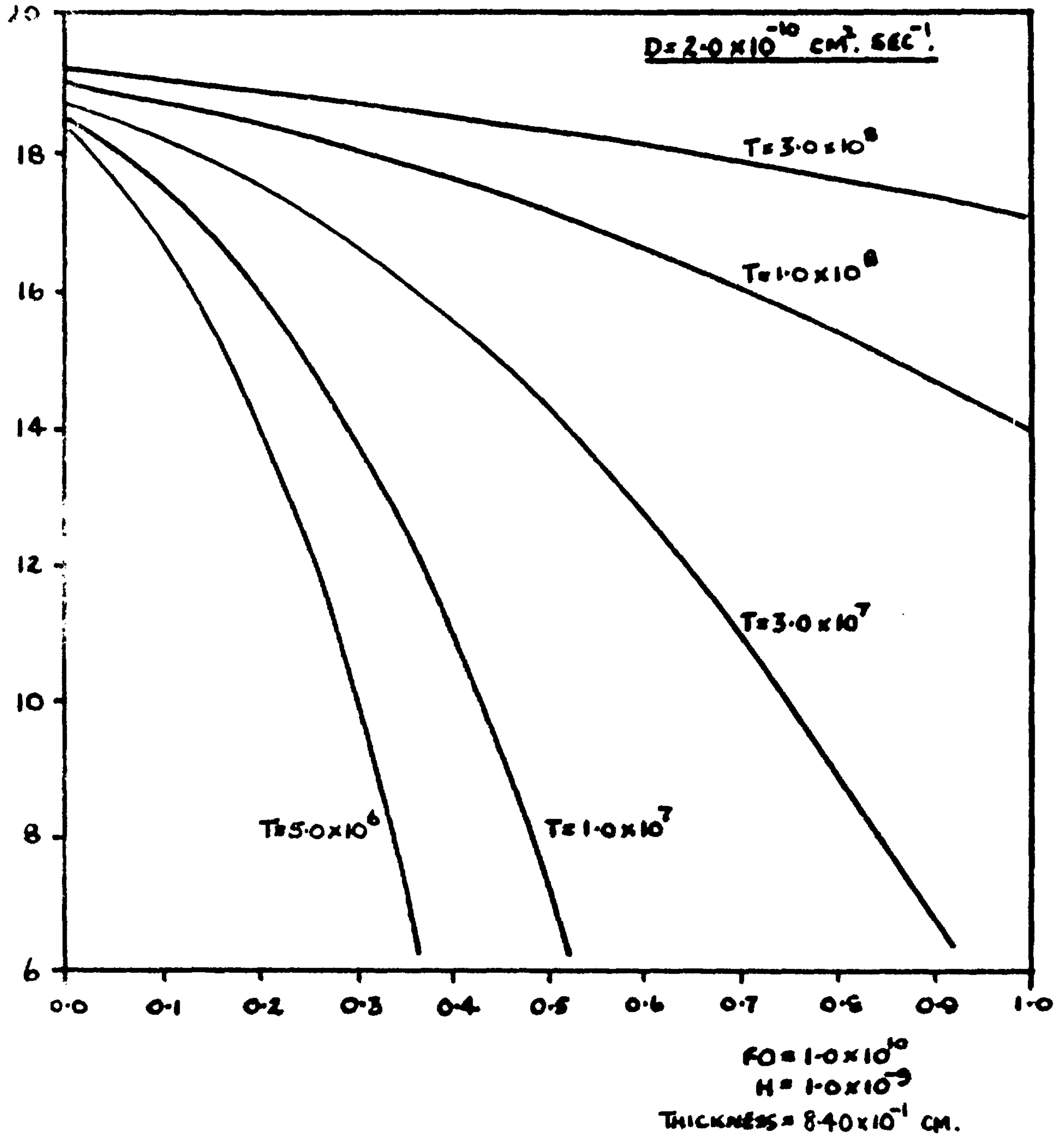
AERE - M1723 FIG.3 LOG CXT. v LOG TIME



AERE - M 1723 FIG. 4 LOG FRACT. v LOG TIME



AERE-M1723 FIG.5 CALCULATED CONCENTRATION PROFILES



AERE-M1723 FIG.6 CALCULATED CONCENTRATION PROFILES.

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