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CALCULATION OF THE FRACTIONAL RELEASE OF GASES FROM A SPHERE AFTER RECOIL BOMBARDMENTS

by

G. DI COLA (EURATOM)
and Hj MATZKE (Nuclear and Radiochemistry Laboratory, Braunschweig Technical University)

1967

Joint Nuclear Research Center
Ispra Establishment - Italy

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Chemistry Department - Radiochemistry Service

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SUMMARY

The gas release from a recoil-bombarded sphere of inert material on annealing at a given temperature is calculated.

Two sources of recoil gas atoms are considered : one of infinite width, the other infinitesimally thin compared to the recoil range in the source. Some additional results are presented for the release from a recoil-bombarded cylinder.

Derived equations are given and values of the fractional release, F, are tabulated and reproduced in figures using results obtained from a standard 7090 IBM electronic computer.

Introduction

This report concludes the calculations started in a previous report (1) which gives the time dependence of the release of recoil gas atoms from a slab (infinite plane sheet) during post-bombardment annealing. In the present report, we will consider the release from a sphere in detail and will give some additional data for release from an infinite cylinder. Two different initial distributions are considered: the gas atoms are assumed to have recoiled into the sample either from an infinitely thick source (with respect to the recoil range in the source) or an infinitesimally thin source. The first distribution is experimentally verified by embedding the solid into a uranium bearing matrix. During reactor irradiation, the solid is labelled with the fission rare gases Kr and Xe. The second distribution is verified by adsorbing a thin layer of Ra-226 on the solid. During the α-decay, the daugther product Rn-222 is recoiled into the solid.

We will assume the following: inert material, no diffusion before annealing, reaching the prescribed temperature "instantaneously", constant diffusion coefficient, zero surface concentration at all times, stable recoil gas atoms.

The derived equations are presented. Using values obtained from a standard 7090 IBM electronic computer, the fractional release, F, is tabulated and shown graphically related to a dimensionless function of the diffusion coefficient. Comparison with literature and experiments is made.

The calculations can easily be extended to a radioactive nuclide by a simple correction factor.

Manuscript received on May 16, 1967.

I. The Mathematical Problem

1. The diffusion of gases following irradiation or bombardment in a solid sphere or an infinite cylinder is described by the following differential equation

$$\frac{\partial c}{\partial t} = D\left(\frac{\partial^2 c}{\partial r^2} - \frac{v}{r}\frac{\partial c}{\partial r}\right) \qquad (1)$$

with v = 2 for the cylindrical and v = 1 for the spherical geometry.
We assume the surface concentration to be zero, hence

$$c(a,t) = 0$$

where a is the radius of the solid.
Furthermore, we use the boundary condition

$$c(r,0) = f(r) = \text{distribution of recoil atoms in the solid}$$

The solution of these problems can be obtained by standard methods (2).

2. Solution for the sphere

$$c(r,t) = \frac{2}{ar}\sum_{n=1}^{\infty} \sin\frac{n\pi r}{a}\exp\left(-\frac{n^2\pi^2 DT}{a^2}\right)\int_0^a r'f(r')\sin\frac{n\pi r'}{a}\,dr' \qquad (2)$$

or

$$c(r,t) = \frac{1}{2r\sqrt{\pi Dt}}\sum_{n=-\infty}^{+\infty}\int_0^a r'f(r') \times$$

$$\times\left\{\exp\left(-\frac{2na+r'-r}{4Dt}\right)^2 - \exp\left(-\frac{2na+r+r'}{4Dt}\right)^2\right\}dr' \qquad (2')$$

Eq. (2') is useful for small values of Dt/a^2 .

3. Solution for the cylinder:

$$c(r,t) = \frac{2}{a^2} \sum_{n=1}^{\infty} \frac{J_o(h_n \frac{r}{a})}{J_1^2(h_n)} \exp\left(- h_n^2 \frac{Dt}{a^2}\right) \int_o^a r'f(r')J_o(h_n \frac{r}{a})dr' \tag{3}$$

where h_n are the roots of the equation, $J_o(h_n) = 0$.

II. Distribution of Recoil Atoms

The distribution of recoil atoms in a sphere of radius a may be derived in a way similar to that described in ref. (1) for the case of a slab. The fraction of recoil atoms which pass through the unit area of the sphere from those recoiled from the volume V of the source is given by

$$B = Q \int_V \frac{\cos \varphi}{4\pi r'^2} dV \tag{4}$$

where Q is the number of atoms in each unit of volume of the source. The region V from which the recoil atoms can strike the surface of the sphere is bounded by

$$0 < \varphi < \bar{\varphi} \quad \text{and} \quad r_1 < r' < r_1 + r_2 \tag{5}$$

where $\cos \bar{\varphi} = \frac{a^2-R^2-r^2}{2Rr}$

$$r_1 = \sqrt{a^2-r^2\sin^2\varphi} - r \cos \varphi$$

$$r_2 = \begin{cases} R'(1 - \frac{r_1}{R}) & \text{infinite source with the assumption of linear energy loss} \\ \frac{a\Delta a}{\sqrt{a^2-r^2\sin^2\varphi}} & \text{infinitesimal source of depth } \Delta a. \end{cases}$$

R and R' (R' < a) are, respectively, the recoil ranges in the sphere and in the source.

The distribution of recoil atoms is given by the concentration of recoil atoms coming to rest at a given location:

$$f(r) = \frac{1}{r^2} \frac{d}{dr} (r^2 B) \qquad (6)$$

After some lenghty calculations the following expressions for f(r) result

$$f(r) = \left\{ \begin{array}{ll} \frac{QR'}{4R^2} \left[\frac{(R^2-a^2)}{r} + 2R + r\right] & \text{(infinite source)} \\ \frac{Qa\Delta a}{2R} \cdot \frac{1}{r} & \text{(infinitesimal source)} \\ & \text{for } a - R < r < a \\ f(r) = 0 & \text{for } 0 \leq r \leq a - R \end{array} \right. \qquad (7)$$

As an approximation for R << a, a uniform concentration might be assumed in the bombarded region, i.e.

$$f(r) = \text{const.} \quad \text{for} \quad a - R < r < a$$

$$f(r) = 0 \quad \text{for} \quad 0 \leq r \leq a - R$$

For the infinite cylinder, only this case will be treated as the above type of derivations become more complicated. Furthermore, less experimental work is done using the cylindrical geometry. Hence, the above case can be considered a useful approximation for this case.

III. The fractional Gas Release

The fractional gas release, $F(t)$, is defined by

$$F(t) = 1 - \frac{\bar{c}(t)}{\bar{c}(0)} \tag{8}$$

where $\bar{c}(t) = \int_V c(r,t)dV$ with V = volume of the solid.

After straight forward but lengthy calculations and with the use of dimensionless parameters $\beta = R/a$ and $\tau^2 = Dt/a^2$, we have;

for a sphere and the infinite source

$$F(t) = 1 - \frac{24}{\beta^2(\beta^2-12)\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \left\{\frac{2}{n\pi}\left(\sin n\pi\beta + \frac{1-\cos n\pi\beta}{n\pi}\right) - \beta^2-2\beta\right\} \times$$

$$\times \exp(-n^2\pi^2\tau^2) \tag{9}$$

for a sphere and the infinitesimal source

$$F(t) = 1 - \frac{4}{\beta(2-\beta)\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} (1-\cos n\pi\beta) \exp(-n^2\pi^2\tau^2) \tag{9'}$$

for a sphere and a rectangular profile

$$F(t) = 1 - \frac{6}{\pi^3(\beta^3-3\beta^2+3\beta)} \sum_{n=1}^{\infty} \frac{1}{n^3} \left\{n\pi-(1-\beta)n\pi \cos n\pi\beta - \sin n\pi\beta\right\} \times$$

$$\times \exp(-n^2\pi^2\tau^2) \tag{9''}$$

For $\beta = 1$, i.e. a uniform distribution eq. (9") is easily seen to yield the usual solution (2) for a homogeneously labelled sphere

$$F(t) = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp\left(- n^2\pi^2\tau^2\right)$$

For the infinite cylinder having a rectangular profile

$$F(t) = 1 - \frac{4}{2\beta-\beta^2} \sum_{n=1}^{\infty} \frac{1}{h_n^2} \left\{1 - \frac{J_1(h_n(1-\beta))}{J_1(h_n)} (1-\beta)\right\} \exp\left(- h_n^2\tau^2\right) \tag{10}$$

Again, for $\beta = 1$, eq. (10) yields the known (2) solution for a homogeneously labelled cylinder

$$F(t) = 1-4 \sum_{n=1}^{\infty} \frac{1}{h_n^2} \exp\left(- h_n^2\tau^2\right)$$

IV. Comparison with literature

This report concludes the calculations started previously. These earlier calculations are summarized in ref. (1) which gives greater details. A large amount of experimental work has been done on the release of gases from spheres when the distribution was uniform, as occurs, for example, by irradiation in a reactor. Therefore, the kinetics for the release for this case have been tabulated before (3, 4). In contrast, very little calculations have been done with types of concentration profiles used here though a variety of experiments has been published (5 - 8). The gas release from a sphere surrounded by an infinitely thick source has been calculated elsewhere (7, 9) and has yielded results similar to these obtained here.

V. Comparison with experiments

The concentration profiles considered here have been obtained either by adsorbing a thin layer of Ra-226 on the surface of the solid (8, 10, 11) or by irradiating the solid in a matrix of U or UO_2 (e.g. 5-7, 10, 11). In the first case, the daughter product Rn-222 penetrates the solid by means of the α-recoil energy of about 85 keV; in the second case, the fission product gases Kr or Xe enter the solid by recoil with the fission energy of about 80 MeV.

The distribution of the injected atoms could not be determined experimentally because of the difficulty of applying any sectioning or stripping technique to a solid having spherical (or cylindrical) geometry. However, the experimentally observed gas release curves for both Rn-222 and fission-Xe-133 show good agreement with the theoretical curves, as long as the gas concentration is low enough to exclude trapping phenomena (see refs. (12, 13). As examples, fig.5 gives experimental results for the infinite, fig.6 for the infinitesimal source. In both cases, the experimental release is seen to follow satisfactorily the theoretically expected time dependence. The shift of release towards higher values of t for $\beta < 1$ represents the diffusion into the unlabelled interior of the solid.

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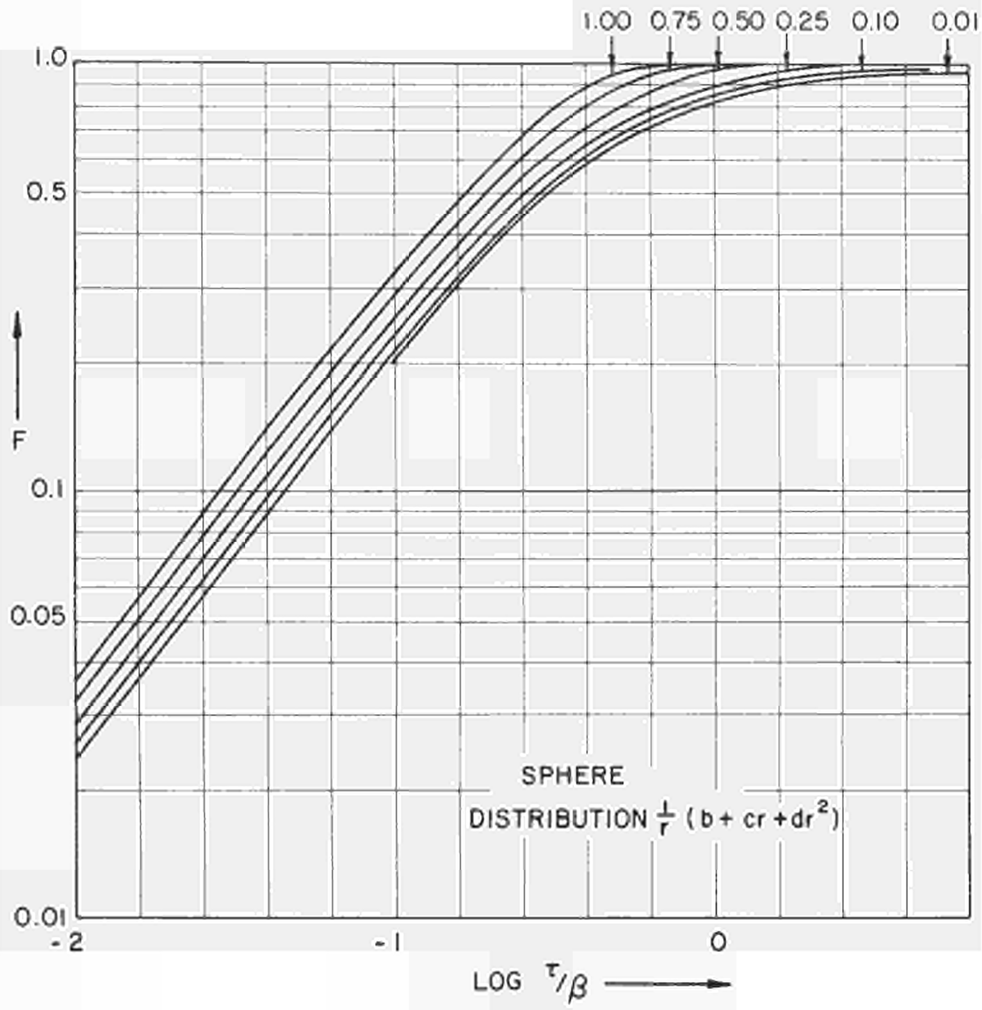

Fig. 1

Fig. 1 : Fractional release, F, as function of τ/β for a sphere with a concentration of the type $f(r) = \frac{1}{r}(a+br+cr^2)$. This case corresponds to an infinite source of recoil atoms.
(β = 1.00, 0.75, 0.50, 0.25, 0.10, 0.01 $\approx$ 0.001)

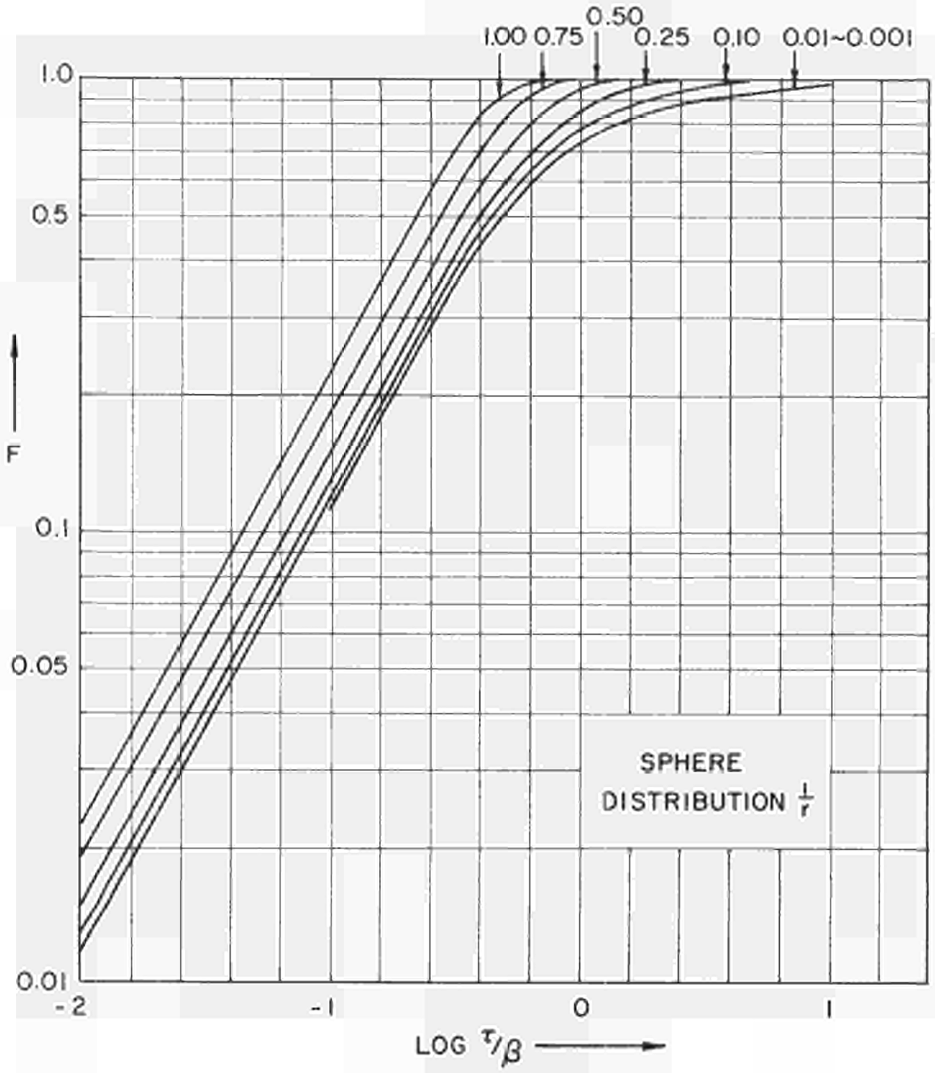

Fig. 2

Fig. 2 : F as function of τ/β for a sphere with an initial concentration profile of the type $f(r) = 1/r$. This case corresponds to an infinitesimally thin source of recoil atoms.
(β = 1.00, 0.75, 0.50, 0.25, 0.10, 0.01)

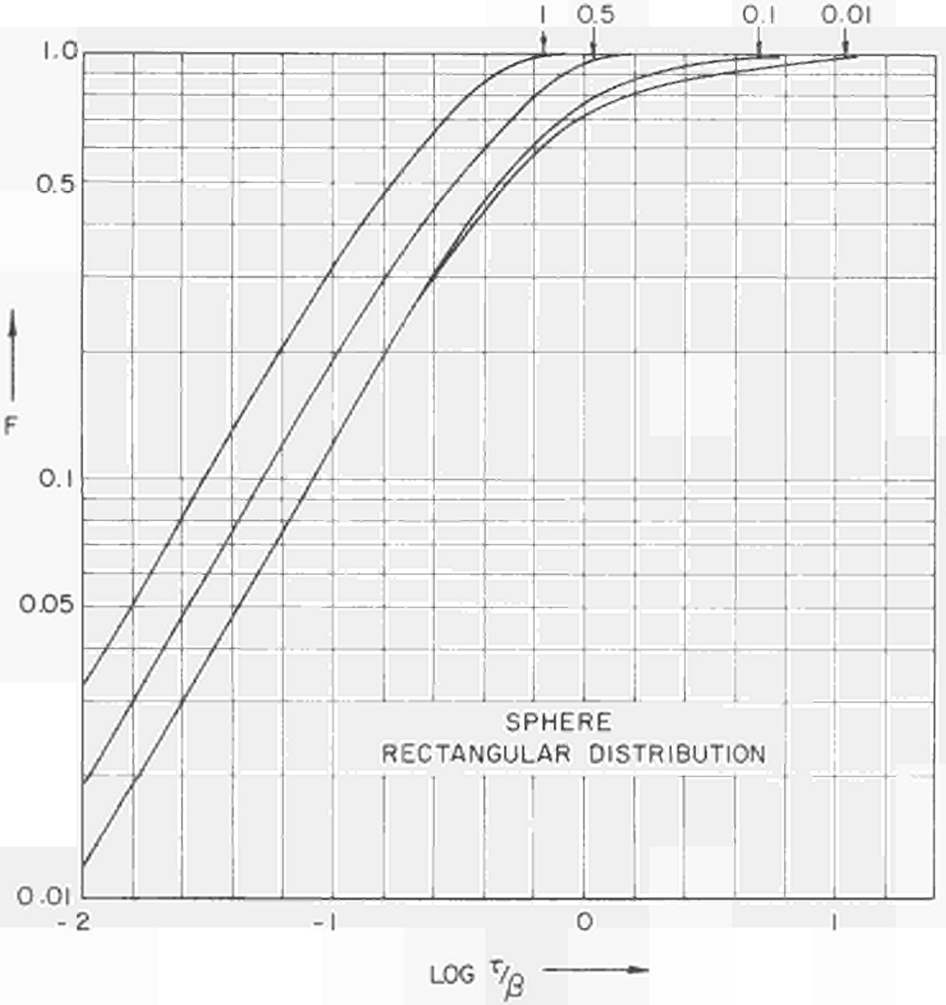

Fig. 3

Fig. 3 : F as function of τ/β for a sphere with a concentration profile of the type $f(r)$ = const. for $a - R < r < a$. This case may be used as an approximation for $R \ll a$. (β = 1.00, 0.50, 0.10, 0.01)

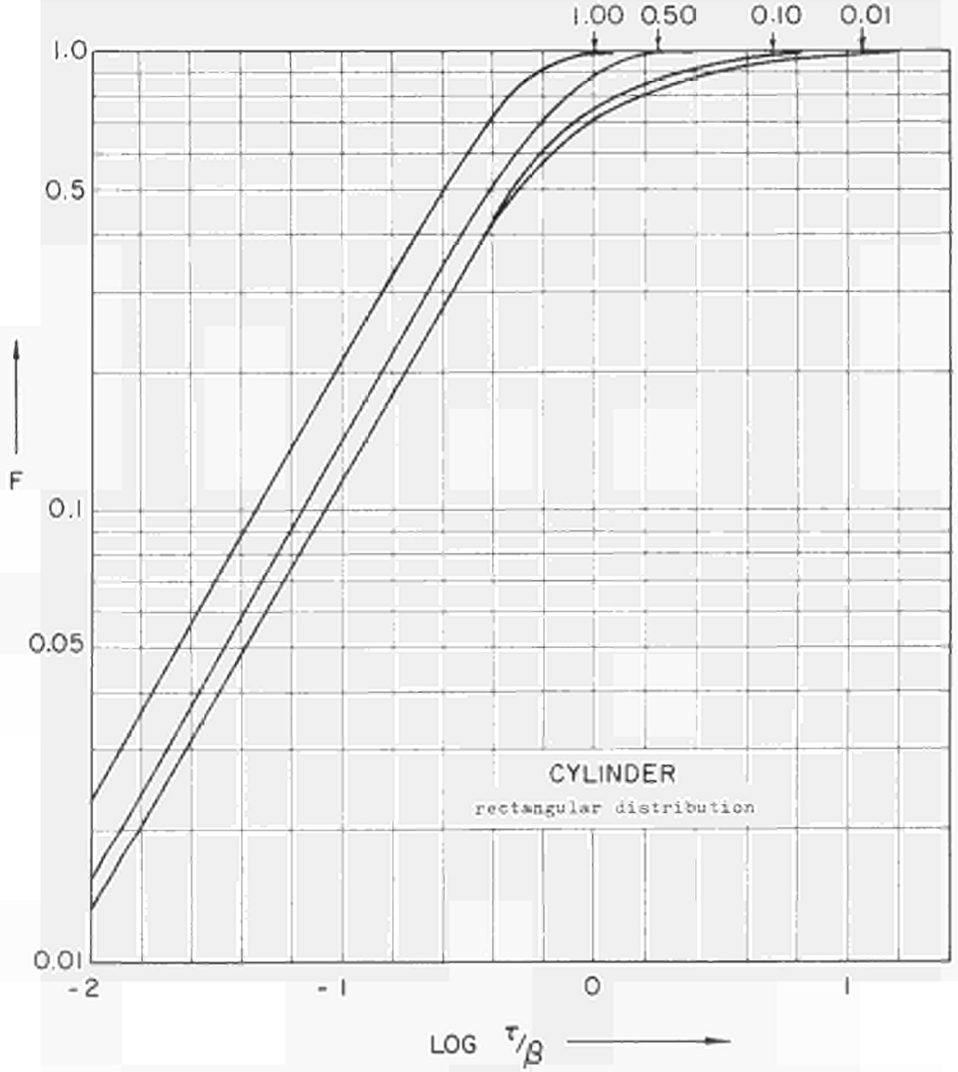

Fig. 4

Fig. 4 : F as function of τ/β for an infinite cylinder with a concentration profile of the type $f(r)$ = const. for $a - R < r < a$. This case may be used as an approximation for $R \ll a$.
(β = 1.00, 0.50, 0.10, 0.01)

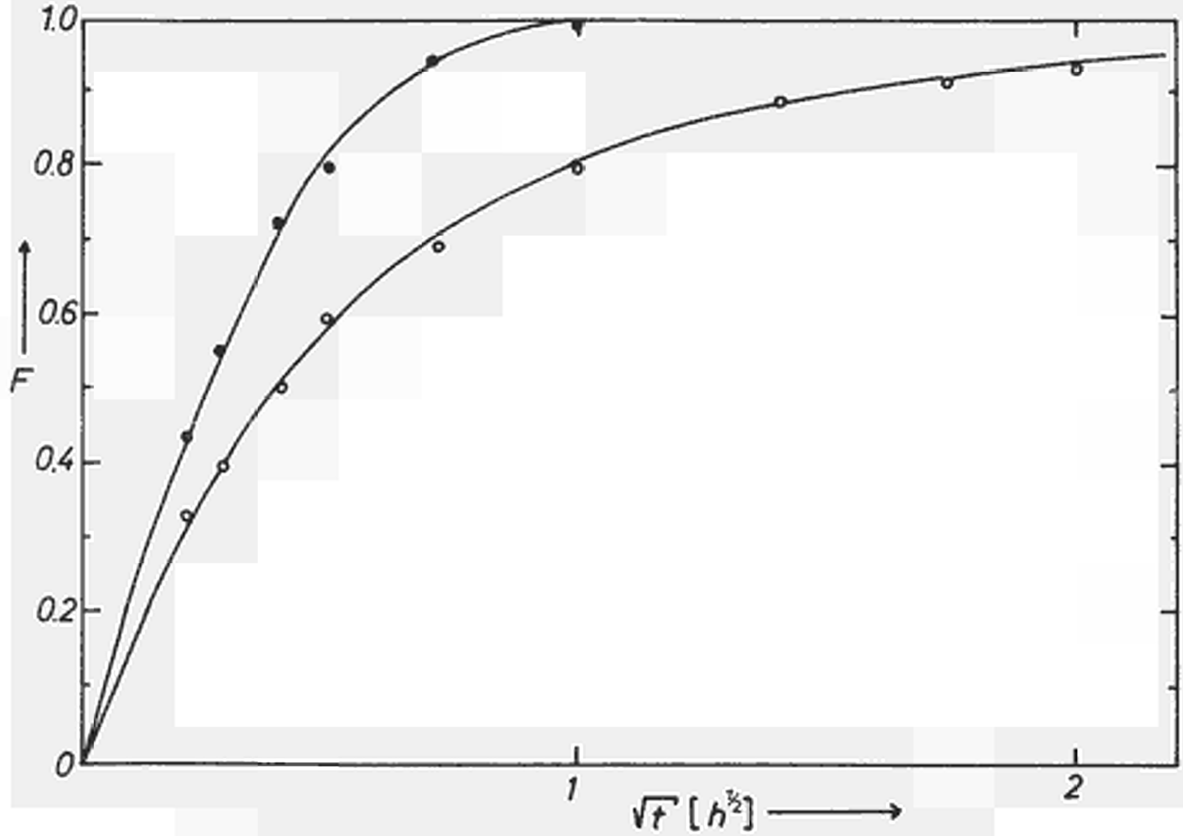

Fig. 5 : Observed fractional release, F, of Xe - 133 from (spherical) CaF_2 particles irradiated in a UO_2 matrix. The full symbols represent the condition $\beta = 1$, the open symbols are for $\beta = 0.25$. The drawn lines are the theoretical curves.

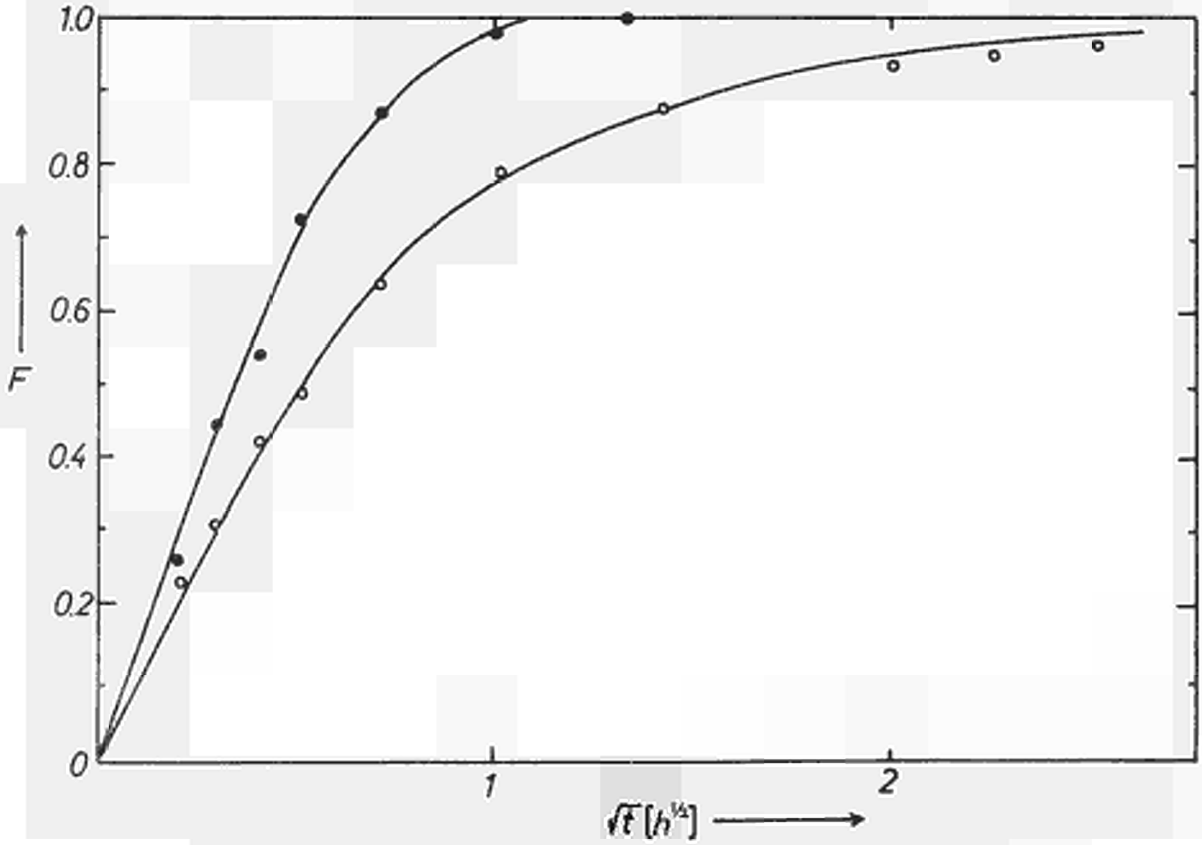

Fig. 6 : Observed fractional release, F, of Rn - 222 from (spherical) U_3O_8 particles labelled by α-recoil from an adsorbed thin layer of Ra-226. The full symbols represent the condition $\beta = 0.75$, the open symbols are for $\beta = 0.25$. The drawn lines are theoretical curves

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Table 1 : F as function of τ/β for a sphere and a concentration of the type $f(r) = \frac{1}{r}(a+br+cr^2)$.

$\log \frac{\tau}{\beta}$ \ β	1.00	0.75	0.50	0.25	0.10	0.01
-2.00	0.0365	0.0322	0.0285	0.0253	0.0235	
-1.95	0.0409	0.0361	0.0319	0.0283	0.0263	
-1.90	0.0458	0.0404	0.0358	0.0317	0.0295	
-1.85	0.0513	0.0453	0.0401	0.0356	0.0331	
-1.80	0.0574	0.0507	0.0449	0.0398	0.0370	
-1.75	0.0643	0.0567	0.0503	0.0446	0.0415	
-1.70	0.0720	0.0635	0.0563	0.0499	0.0464	
-1.65	0.0805	0.0711	0.0630	0.0559	0.0520	
-1.60	0.0900	0.0795	0.0704	0.0625	0.0582	
-1.55	0.1007	0.0889	0.0788	0.0699	0.0651	
-1.50	0.1125	0.0993	0.0881	0.0782	0.0728	
-1.45	0.1256	0.1109	0.0984	0.0874	0.0814	
-1.40	0.1402	0.1239	0.1099	0.0976	0.0909	
-1.35	0.1564	0.1382	0.1226	0.1090	0.1016	
-1.30	0.1743	0.1541	0.1368	0.1216	0.1133	
-1.25	0.1942	0.1717	0.1525	0.1356	0.1264	
-1.20	0.2160	0.1911	0.1698	0.1511	0.1409	
-1.15	0.2401	0.2125	0.1889	0.1682	0.1569	
-1.10	0.2666	0.2360	0.2099	0.1871	0.1746	
-1.05	0.2956	0.2618	0.2330	0.2078	0.1940	
-1.00	0.3273	0.2900	0.2582	0.2305	0.2153	0.2066
-0.95	0.3617	0.3207	0.2858	0.2553	0.2386	0.2291
-0.90	0.3990	0.3540	0.3157	0.2822	0.2230	0.2536
-0.85	0.4392	0.3899	0.3480	0.3114	0.2915	0.2801
-0.80	0.4822	0.4285	0.3827	0.3429	0.3212	0.3088
-0.75	0.5279	0.4695	0.4198	0.3765	0.3530	0.3395
-0.70	0.5761	0.5128	0.4590	0.4122	0.3867	0.3722
-0.65	0.6264	0.5581	0.5001	0.4497	0.4223	0.4067
-0.60	0.6783	0.6049	0.5426	0.4886	0.4593	0.4426
-0.55	0.7308	0.6527	0.5862	0.5285	0.4972	0.4795
-0.50	0.7830	0.7008	0.6300	0.5688	0.5356	0.5168
-0.45	0.8333	0.7484	0.6736	0.6089	0.5739	0.5541
-0.40	0.8797	0.7949	0.7162	0.6482	0.6115	0.5907
-0.35	0.9199	0.8393	0.7574	0.6862	0.6478	0.6261
-0.30	0.9519	0.8805	0.7966	0.7225	0.6825	0.6599
-0.25	0.9747	0.9171	0.8336	0.7567	0.7153	0.6919
-0.20	0.9887	0.9475	0.8680	0.7887	0.7459	0.7217
-0.15	0.9959	0.9704	0.8996	0.8182	0.7743	0.7494
-0.10	0.9989	0.9856	0.9279	0.8454	0.8004	0.7749
-0.05	0.9998	0.9942	0.9520	0.8703	0.8242	0.7982
0.00	0.9999	0.9981	0.9712	0.8929	0.8459	0.8194
0.05		0.9996	0.9848	0.9134	0.8655	0.8386
0.10		0.9999	0.9932	0.9319	0.8833	0.8559
0.15			0.9975	0.9484	0.8993	0.8715
0.20			0.9993	0.9630	0.9137	0.8855
0.25			0.9999	0.9754	0.9265	0.8982
0.30				0.9852	0.9381	0.9094
0.35				0.9922	0.9484	0.9195
0.40				0.9964	0.9577	0.9285
0.45				0.9987	0.9660	0.9367
0.50				0.9996	0.9733	0.9439
0.70				0.9999	0.9943	0.9658
0.90					0.9999	0.9796
1.10						0.9884

Table 2 : F as function of τ/β for a sphere and a concentration of the type $f(r) = 1/r$.

$\log \frac{\tau}{\beta}$ \ β	1.00	0.75	0.50	0.25	0.10	0.01
-2.00	0.0226	0.0181	0.0150	0.0129	0.0119	
-1.95	0.0253	0.0203	0.0169	0.0145	0.0133	
-1.90	0.0284	0.0227	0.0189	0.0162	0.0150	
-1.85	0.0318	0.0255	0.0213	0.0182	0.0168	
-1.80	0.0358	0.0286	0.0238	0.0204	0.0188	
-1.75	0.0401	0.0321	0.0267	0.0229	0.0211	
-1.70	0.0450	0.0360	0.0300	0.0257	0.0237	
-1.65	0.0505	0.0404	0.0336	0.0289	0.0266	
-1.60	0.0567	0.0454	0.0378	0.0324	0.0298	
-1.55	0.0636	0.0509	0.0424	0.0363	0.0335	
-1.50	0.0714	0.0571	0.0476	0.0408	0.0376	
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-1.40	0.0898	0.0719	0.0599	0.0513	0.0473	
-1.35	0.1008	0.0806	0.0672	0.0576	0.0531	
-1.30	0.1131	0.0905	0.0754	0.0646	0.0595	
-1.25	0.1269	0.1015	0.0846	0.0725	0.0668	
-1.20	0.1424	0.1139	0.0949	0.0814	0.0749	
-1.15	0.1598	0.1278	0.1065	0.0913	0.0840	
-1.10	0.1793	0.1434	0.1195	0.1024	0.0943	
-1.05	0.2011	0.1609	0.1341	0.1149	0.1059	
-1.00	0.2257	0.1805	0.1505	0.1290	0.1188	0.1134
-0.95	0.2532	0.2026	0.1688	0.1447	0.1333	0.1272
-0.90	0.2841	0.2273	0.1894	0.1623	0.1495	0.1428
-0.85	0.3188	0.2550	0.2125	0.1822	0.1678	0.1602
-0.80	0.3577	0.2861	0.2384	0.2044	0.1882	0.1797
-0.75	0.4013	0.3210	0.2675	0.2293	0.2112	0.2017
-0.70	0.4502	0.3602	0.3002	0.2573	0.2370	0.2262
-0.65	0.5047	0.4040	0.3366	0.2885	0.2658	0.2537
-0.60	0.5648	0.4527	0.3772	0.3233	0.2978	0.2843
-0.55	0.6298	0.5063	0.4220	0.3617	0.3331	0.3181
-0.50	0.6979	0.5646	0.4705	0.4033	0.3715	0.3547
-0.45	0.7660	0.6265	0.5222	0.4476	0.4123	0.3936
-0.40	0.8304	0.6907	0.5762	0.4939	0.4549	0.4343
-0.35	0.8869	0.7549	0.6313	0.5411	0.4984	0.4758
-0.30	0.9321	0.8166	0.6864	0.5883	0.5419	0.5174
-0.25	0.9642	0.8724	0.7404	0.6346	0.5845	0.5581
-0.20	0.9841	0.9190	0.7923	0.6793	0.6257	0.5974
-0.15	0.9942	0.9543	0.8412	0.7219	0.6649	0.6348
-0.10	0.9984	0.9798	0.8856	0.7618	0.7017	0.6699
-0.05	0.9996	0.9910	0.9238	0.7990	0.7359	0.7026
0.00	0.9999	0.9971	0.9542	0.8333	0.7675	0.7328
0.05		0.9993	0.9758	0.8647	0.7964	0.7604
0.10		0.9999	0.9892	0.8933	0.8228	0.7856
0.15			0.9961	0.9191	0.8467	0.8084
0.20			0.9989	0.9419	0.8683	0.8291
0.25			0.9998	0.9613	0.8878	0.8477
0.30			0.9999	0.9767	0.9053	0.8644
0.35				0.9877	0.9211	0.8794
0.40				0.9945	0.9352	0.8929
0.45				0.9980	0.9478	0.9050
0.50				0.9994	0.9591	0.9157
0.70				0.9999		0.9485
0.90						0.9694
1.10						0.9825

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Alfred Nobel

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