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EUROPEAN ATOMIC ENERGY COMMUNITY - EURATOM

KNOCK-OUT COEFFICIENT OF SINTERED URANIUM DIOXIDE

by

O. GAUTSCH and C. MUSTACCHI

1966

Joint Nuclear Research Center
Ispra Establishment - Italy

Physical Chemistry

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CONTENTS

SUMMARY

Sintered UO_2 wafers, facing a metal collector were irradiated at thermal neutron doses of about $5 \cdot 10^{18}$ n/cm^2. Quantitative analysis of the U deposits on the collector yielded a knock-out coefficient of 260 atoms (U + O) per average recoil fragment.

1. Introduction

Fission fragments escaping from the surface of a fuel element knock out some fuel material (1) (2) (3). This phenomenon may lead to gradual disintegration of fine sized fuel particles at high burn-up rates.

Scope of the present experiments was to asses the knock-out coefficient for sintered UO_2. Similar measurements were performed by M.D. Rogers (1) and G. Nilsson (4) leading to rather scattered results, i.e., 630 and 10 atoms ejected per average fission fragment.

The knowledge of the knock-out coefficient is important whenever the stability and duration of suspended fuel in homogeneous reactors is investigated. It is also important when the interaction fuel-clad or anomalies in the release behaviour of fission gases are studied.

Besides the knock-out coefficient the effect of distance from the emitting UO_2 surface on the collected amount of U and Zr^{95} was determined. The purpose of this double determination was to check whether knocked out particles had energies other than those of recoil fragments, since in this case their vertical distribution on the collector would differ from that of fission products.

Manuscript received on May 17, 1966.

2. Experimental procedure

Quartz vials containing a sintered UO_2 pellet (10.74 g/cm^3, 1.5 mm thick, surface roughness below 0.1 μm, see also Appendix 5.1) and a cylindrical Al collector were irradiated with thermal neutron doses of about $5 \cdot 10^{18}$ n/cm^2. The vials (Fig. 1) were filled with known amounts of Kr. Because of the presence of Kr, the cylindrical shape of the collector, and the low value of the ratio "emitter surface/collector surface" (0.28 $cm^2/6$ cm^2), return of deposited matter from the collector to the emitter was believed negligible. Experimental data are summarized in table 1.

Table 1

Experimental data

Sample	1	2	3	4
Volume of Kr in the vial, cm^3 NTP	4.6±2%	4.6±2%	4.6±2%	-
Kr pressure at R.T. atm	1.3±3%	2.0±3%	1.7±3%	-
Irradiation time, sec	$1.27.10^6$	$1.39.10^6$	$1.40.10^6$	$0.95.10^6$
Flux, n/cm^2 sec	$5.8.10^{12}$±7%	$4.1.10^{12}$±7%	$5.8.10^{12}$±7%	-
Decay time, days	191	147	105	9

After irradiation and the indicated cooling time the irradiated capsules were opened and the collectors were cut into rings of approximately the same height. These rings yielded gamma spectra of the deposited fission products and the amounts of collected U, which was determined by activation analysis[*], with a maximum error of ± 10%.

The gamma spectra were obtained by placing the rings (collector elements) on a 60 mm dia. scintillator and by integration of the photopeaks with a 400 channel pulse height analyzer. The maximum counting error was ± 2%. The spectrum of the collector material (sample 4, Table 1) showed photopeaks belonging to impurities in the Al. These activity rates were constant for all collector elements. On samples 1, 2, and 3 an additional photopeak appeared at 0.76 MeV. It was attributed to Zr^{95} + Nb^{95} (see Appendix 5.2.), and the area below it decreased with increasing distance between collector element and emitter.

The amounts of U and the activity rates of Zr^{95} + Nb^{95} found on the collector rings are given in table 2.

Estimates were made showing that the quantities of Zr^{95} knocked out from the UO_2 pellet and/or generated on the collector were negligible as compared with the amounts of Zr^{95} reaching the collector as Zr^{95} recoil fragment or as its precursor in the fission chain.

[*] Dr. Carla Bigliocca, Nuclear Chemistry Service.

3. Results

3.1. The knock-out coefficient was calculated from the amounts of U found on the collector elements assuming that O_2 is knocked-out proportionally to its concentration in UO_2.

Results are shown in table 3.

Table 2

Results of Zr^{95} + Nb^{95} and U determination.

Collector ring	Average distance from UO_2 emitter, mm	Zr^{95} + Nb^{95} activity rate under the 0.76 MeV photopeak		Collected amount of U	
		cpm/collector element	cpm/cm^2	g/collector element 10^6	g/cm^2.10^6
1/0	21.6	-	-	0.62	0.96
1/1	18.7	-	-	0.54	0.40
1/2	14.4	-	-	0.37	0.35
1/3	10.5	-	-	0.31	0.26
1/4	6.3	-	-	0.46	0.38
1/5	2.1	-	-	0.46	0.38
				2.76	
2/0	22.8	6100	9550	0.46	0.72
2/1	19.2	7760	4370	0.42	0.23
2/2	13.6	10300	7410	0.16	0.11
2/3	9.0	39200	33820	0.19	0.16
2/4	3.5	87600	77600	0.29	0.26
2/5	1.5	66000	77700	0.22	0.26
		216960		1.74	
3/0	20.5	11350	17700	-	-
3/1	17.5	13550	9600	-	-
3/2	13.5	23200	26950	-	-
3/3	9.8	91500	72000	-	-
3/4	4.2	286000	155500	-	-
		425600			

Table 3

Results

Sample	1	2
Neutron dose, n/cm^2	7.4×10^{18}	5.7×10^{18}
Atoms of U per collector	7.0×10^{15}	4.4×10^{15}
Atoms of U per collector normalized to flux-time data of sample 1	7.0×10^{15}	5.7×10^{15}
Atoms (U+O) knocked out	2.1×10^{16}	1.3×10^{16}
Recoil fragments per sample from flux-time data	7.2×10^{13}	5.6×10^{13}
Knock-out coefficient (atoms U+O per recoil fragment)	290±17%	230±17%

These knock-out rates are such to lead one to expect a substantially complete disintegration of UO_2 particles with a diameter less than about 7.10^{-4} cm at thermal neutron doses in excess of 10^{21} n/cm^2.

3.2. Fig. 2 shows the vertical distribution of knocked-out U on the collector. The distribution curve for Zr^{95} is given for comparison (see also 5.1. Appendix). About 10^{15} atoms of U/cm^2

were deposited at a distance of 2 and 18 mm from the UO_2 wafer, indicating random walk of knocked-out particles. About 20 times less Zr^{95} fragments/cm^2 were found at 19 mm from the emitter than at a distance of 2 mm, indicating that fission fragments are not to be considered deposited from a vapor phase, but are endowed with sufficient energy to follow a range-type of deposition. However, although the mean range of recoil fragments in Kr at 2 atm is only about 13 mm, Zr^{95} was found also at higher distances.

4. Conclusions

4.1. A knock-out coefficient of 260 atoms (U+O)/recoil fragment was obtained for sintered UO_2.

4.2. The different vertical distribution of knocked-out U and escaped Zr^{95} fragments indicates that knocked-out particles have energies not far above thermal range.

Appendix 5.1.

Knock-out from rough surfaces.

In a plain surface the angle of escape for knocked-out particles is 3.14 radians and the probability for a particle to escape from the surface is unity. In case of rough surfaces the escape probability will be less than unity. (Fig. 3)

Calculations were made for several ratios of "roughness amplitude/wave length" (5) and the results are shown in Table 4. The escape probability is seen to decrease only by a few %.

Table 4

Results (see also Fig.3)

$\frac{h}{a}$	mean angle of escape, radian	escape probability
0.025	3.07	0.976
0.05	3.00	0.955
0.1	2.87	0.931
0.5	2.00	0.903

By approximating the surface waviness with pyramids of height h and base length a, the ratio of the rough to the plain surface is given by $\left(\left(\frac{2h}{a}\right)^2+1 \right)^{1/2}$. Common values $\frac{h}{a}$ are about 0.01 to 0.1(5). For the wafers used in the foregoing experiments h was about 1 μm and a was 25 μ. Thus the true surface was about 0.3% bigger than the plain one.

Appendix 5.2.

Counting of Zr^{95}.

The activity rate of Zr^{95} at the end of irradiation was calculated for samples 2 and 3 to show that the 0.76 MeV activity belonged to Zr^{95} + Nb^{95}. Both samples must have the same activity rate after correction for decay and normalization to the neutron dose if the observed 0.76 MeV peak is due to Zr^{95} + Nb^{95}.

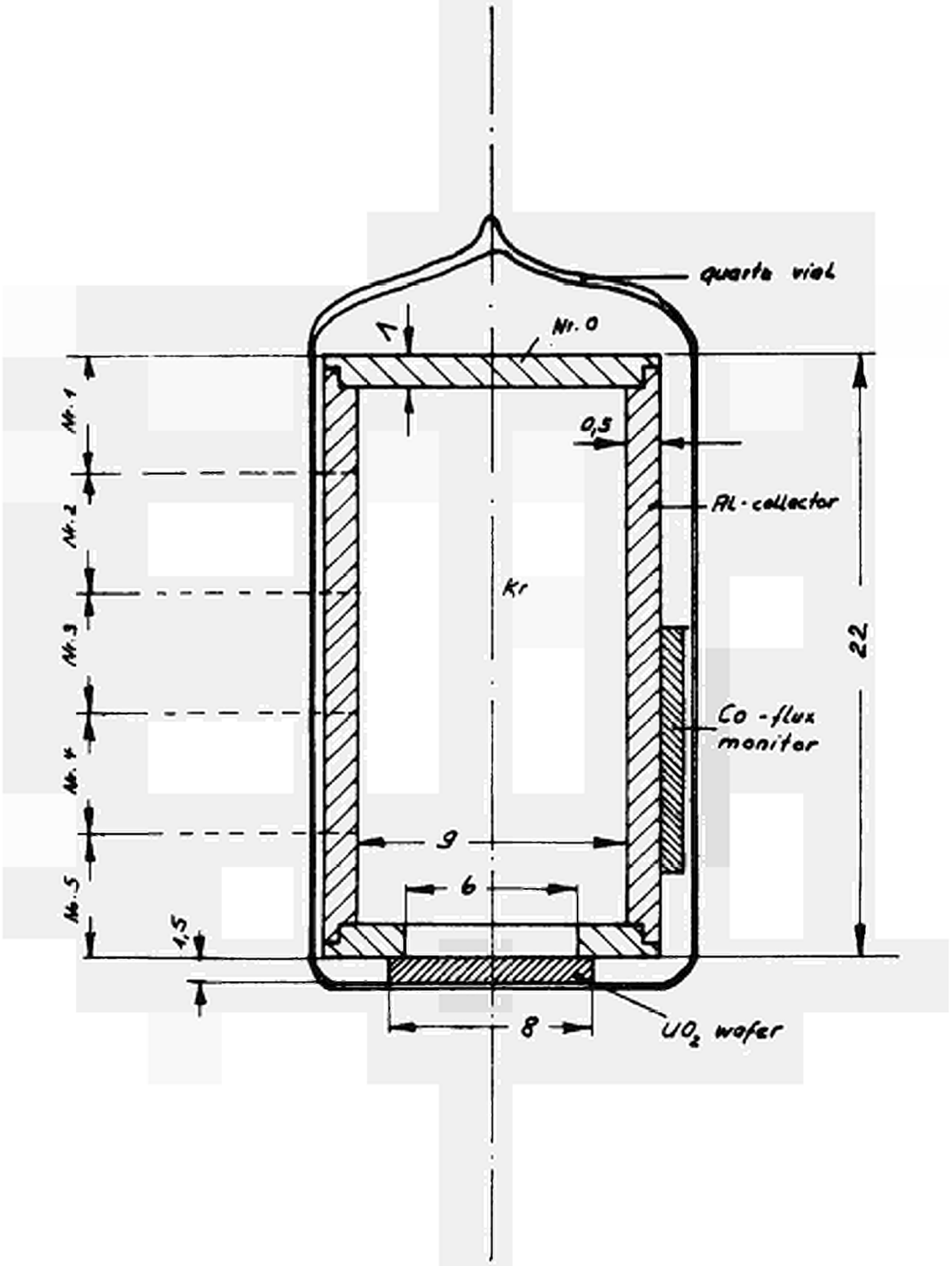
Zr^{95}, with a cumulative fission yield of 6.2 %, has a short-lived precursor chain and a half life of 65 days. It decays with 49 % gamma radiation of 0,76 MeV, 49 % of 0.72 MeV, and 2 % of 0.23 MeV. Nb^{95} has a half life of 35 days and 100 % of 0.77 MeV radiation.

Neglecting the 2 % of isomeric transition and the amount of Nb^{95} present at the end of irradiation, the activity rates of Zr^{95} + Nb^{95} observed after a given decay time were estimated, assuming the same detection coefficient for both nuclides, by

$$(1)\quad A_2 = \frac{T_1}{T_1 - T_2}\ A_1^{\circ}\ \left(\exp\left(-\frac{0.693\ t}{T_1}\right) - \exp\left(-\frac{0.693\ t}{T_2}\right)\right)$$

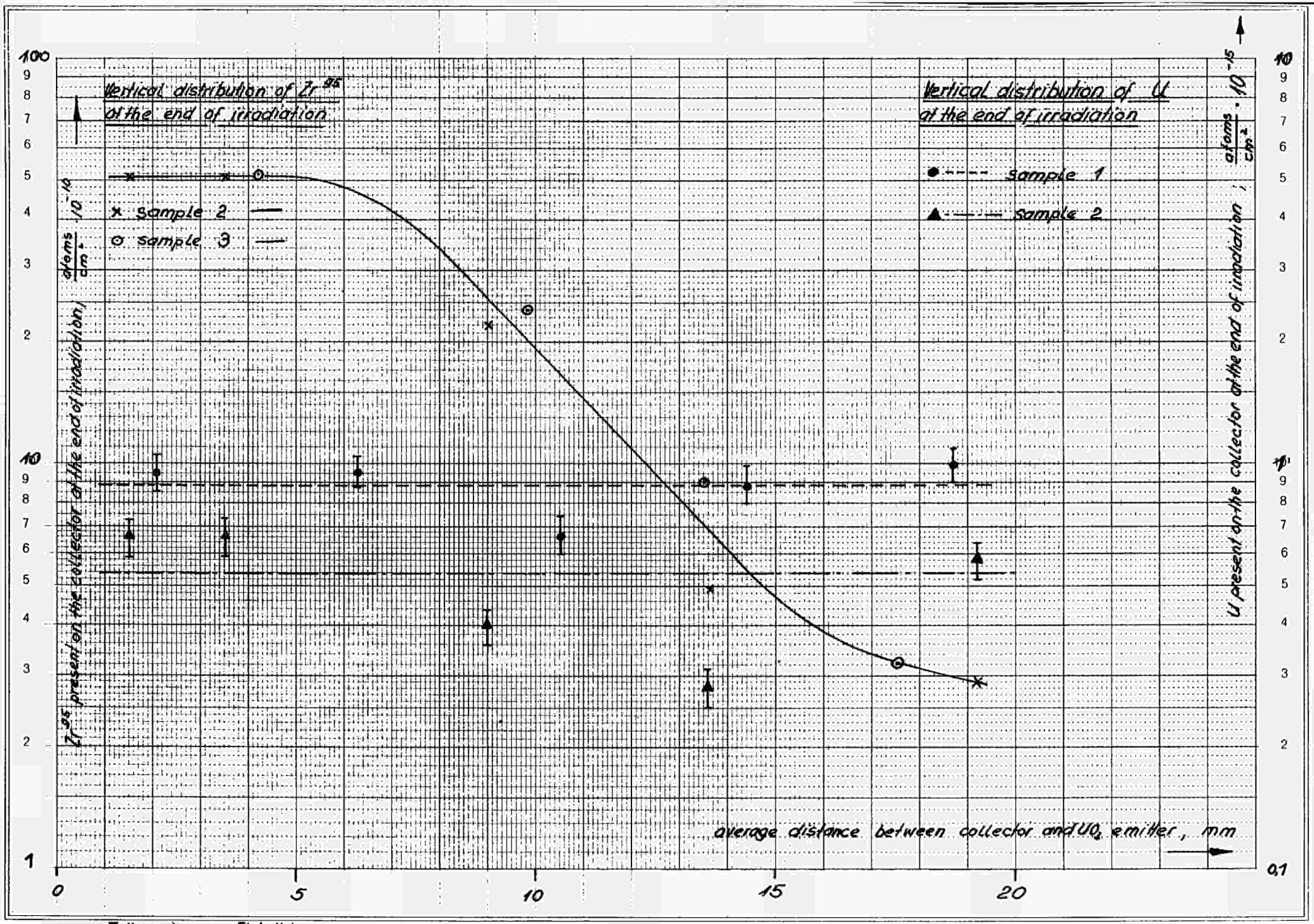
$$(2)\quad A_1 = A_1^{\circ} \exp\left(-\frac{0.693\ t}{T_1}\right)$$

where A_2 is the activity rate of Nb^{95} at time t, A_1° the activity rate of Zr^{95} at the end of irradiation, T_1 and T_2 the half-lives of Zr^{95} and Nb^{95}.



Irradiation vial

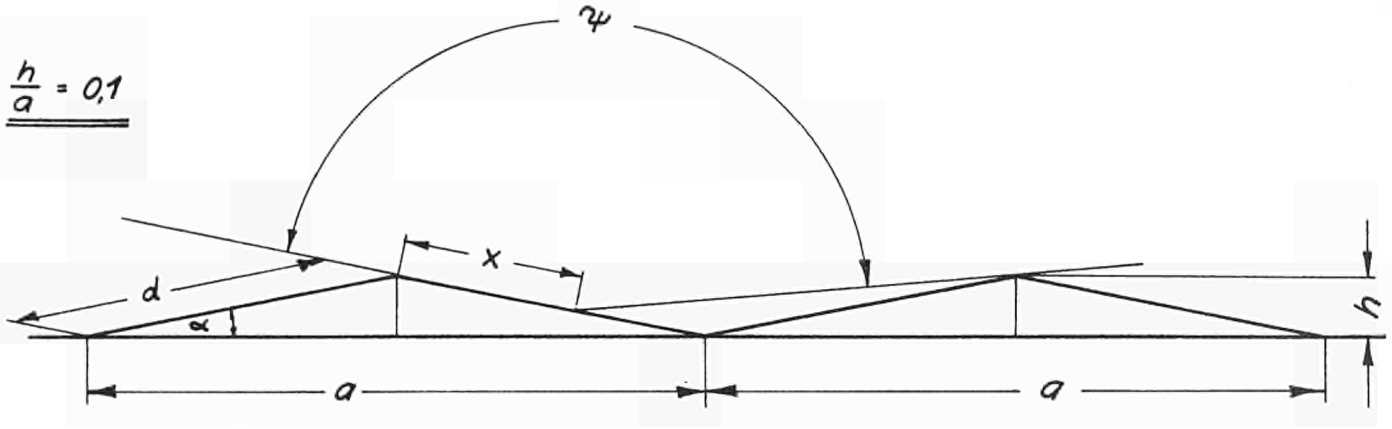
Fig. 1



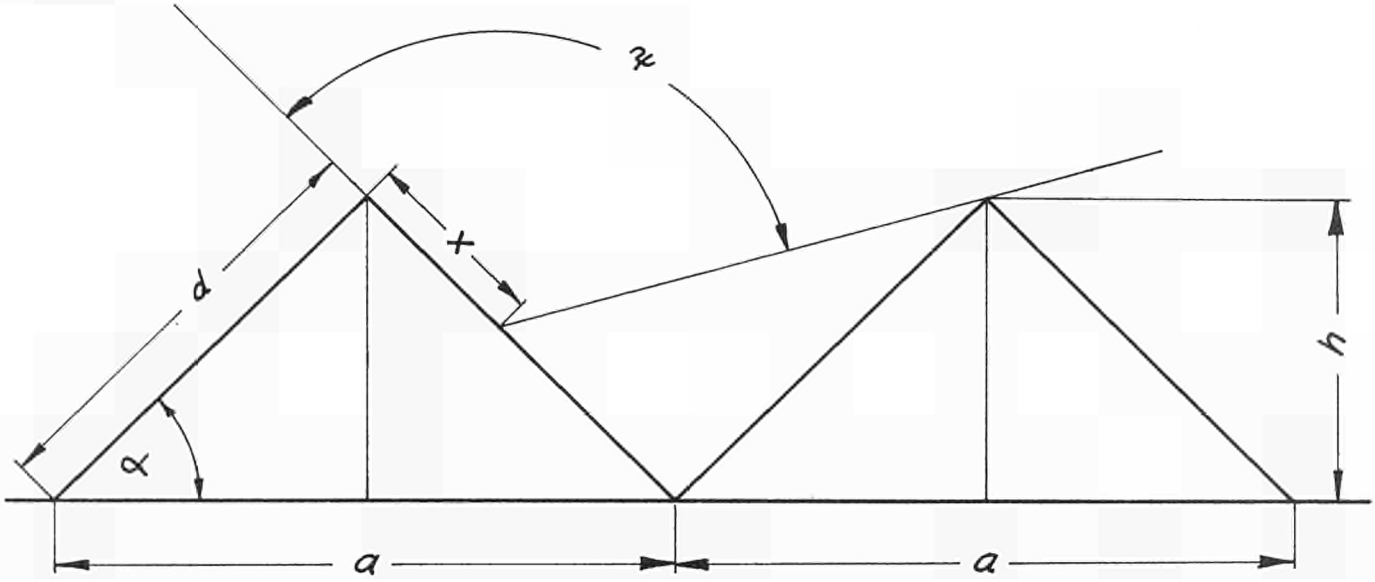
Logar. Teilung / Division 1 - 100, Einheit / Unité 83,33 mm

Fig. 2

$\frac{h}{a} = 0{,}1$



$\frac{h}{a} = 0{,}5$

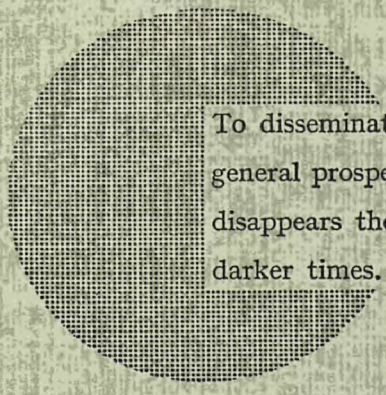


$$\psi_{(x)} = \frac{\pi}{2} - \alpha + \operatorname{arc\,tg} \frac{a - x \cdot \cos\alpha}{x \cdot \sin\alpha}$$

$$\bar{\psi} = \frac{1}{d} \int_0^d \psi_{(x)}\, dx$$

Escape probability : $P = \frac{\bar{\psi} \cdot 2 \cdot d}{\pi \cdot a}$

Fig. 3



To disseminate knowledge is to disseminate prosperity — I mean general prosperity and not individual riches — and with prosperity disappears the greater part of the evil which is our heritage from darker times.

Alfred Nobel

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