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Section P-7

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FISSION PRODUCT POISONING IN A PILE

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ABSTRACT

The effect of the neutron flux on the relative importance of different fission product poisons is discussed. The general expression for the poisoning of a fission product is given with the various equations for special cases of interest. The formulae are applied to the problem of finding the poisoning due to fission products at the end of one day in a pile operating with a flux of 4×10^{14} neutrons/cm^2/sec. These results are summarized in the table on page 12.

Appendix I contains a revised list of cross sections x yields for all stable isotopes. Such a list was first given by Wheeler in CP-889. Appendix II contains a list of radioactive fission products with half lives greater than one day with data about yields, parents, gaseous ancestors, and numbers of neutrons.

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FISSION PRODUCT POISONING IN A PILE

Katharine Way

Introductory Discussion

The amount of poisonous fission products produced in a pile depends, of course, upon the neutron flux since this determines the number of fissions per second. However, the poisons are themselves destroyed when they absorb neutrons so that their rate of disappearance depends also on the power level. The maximum possible amount of any poison will be the "equilibrium" value or the amount present when the rate of destruction just equals the rate of creation. If the poison is itself a stable nucleus and can be considered to be created directly in the fission process

$$\frac{dn}{dt} = yF(n\sigma)_{metal} - F(n\sigma)_{poison}$$

where n is the number of atoms with absorption cross section σ, F is the slow neutron flux which is assumed to be constant in time, and y is the yield* of the fission product in question. Then if the poisoning p be defined as the ratio of the number of slow neutrons absorbed by the poison to that absorbed in the metal

$$p = \frac{(n\sigma)_{poison}}{(n\sigma)_{metal}} = y\left[1 - e^{-F\sigma t}\right].$$

The "equilibrium" value of the poisoning is then just equal to the yield, y. When $F\sigma t$ is small, the poison grows linearly with time

*or branching ratio, i.e., the number of times per fission that a given fission product is formed.

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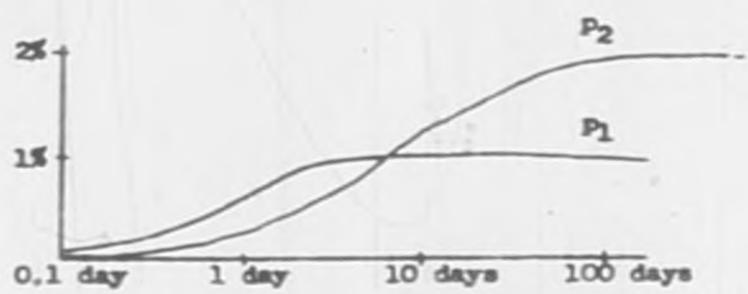
and the poisoning effect then depends on the cross section as well as the yield.

If the flux, F, be expressed in neutrons/barn/day, it is easy to see how long it will take any given poison to come near to its maximum value, or the time at which $F \sigma t = 1$. A flux of 1.16×10^{13} neutrons/ cm^2/sec is equal to 10^{-6} neutrons/barn/day. Thus in a pile operating with this flux, a poison with a cross section of a megabarn will come almost to equilibrium in a day.

The following illustration shows how even the relative effect of two different poisons can be different at different times after startup in the same pile when the flux has remained constant or at the same time after startup in different piles operating with quite different fluxes.

Let one poison have a cross section of 10^6 barns and a yield of 1% and another a cross section of 10^5 barns and a yield of 2%. Then in a pile whose flux is 10^{-6} neutrons/barn/day the two poisons will grow as follows:



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The values of p_1 and p_2 shown in the sketch for 10 days will also be the values of p_1 and p_2 after 1 day of operation in a pile whose flux is 10^{-5} neutrons/barn/day. Thus one sees that when the yield of a "weak" poison is larger than that of a "strong" one, it will have for certain values of Ft the more harmful effect.

General Considerations

A more general treatment of the problem which takes into account radioactive decay of the poison and the half life of the parent of the poison leads to the following differential equations. Let n_o be the number of atoms of the parent in existence and n_1 the number of atoms of the poison. Then

$$\frac{dn_1}{dt} = n_o \lambda_o - n_1(\lambda_1 + F\sigma)^{*}$$

$$\frac{dn_o}{dt} = yF(n\sigma)_{metal} - n_o \lambda_o \quad .$$

By the usual methods for linear equations one easily finds

$$p = yF\sigma t \left\{ \frac{1 - e^{-(\lambda_1 + F\sigma)t}}{(F\sigma + \lambda_1)t} - \frac{e^{-(\lambda_1 + F\sigma)t} - e^{-\lambda_o t}}{(\lambda_o - \lambda_1 - F\sigma)t} \right\} . \qquad (1)$$

Special cases of interest are:

(1) The poison isotope is nearly stable, has a short lived parent and a small cross section; i.e., $F\sigma t \ll 1$. Then, as noted before

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* σ is the cross section of the poison. Its parent is assumed to have a negligible cross section.

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$$p = yF\sigma t \quad . \tag{2}$$

(2) The poison isotope is nearly stable, has a short lived parent but a large cross section. Then as derived above

$$p = y\left[1 - e^{-F\sigma t}\right] \quad . \tag{3}$$

(3) The poison has a short lived parent but its decay constant is of the order of $F\sigma$. Then

$$p = \frac{yF\sigma t}{(F\sigma + \lambda_1)t}\left[1 - e^{-(\lambda_1 + F\sigma)t}\right] \quad . \tag{4}$$

Figures 1, 2, and 3 show how various poisons grow with time according to this expression. The ordinate is in each case the factor which multiplies the yield to give p.

A question that comes up is what half life must a poison with large yield have to make it important after operation for a given time at a given flux. Figure 4 shows the fraction of the maximum poisoning (i.e., of the yield) contributed by poison of different half lives at the end of certain times of operation. One can see here for instance that after one day's operation any poisons with half life of the order of 1 day or greater may make a big contribution if it has high yield and fairly large cross section.

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(4) The poison isotope is nearly stable, has a large cross section but a parent whose lifetime cannot be considered short. In this case p can be written

$$p = y(1 - e^{-F\sigma t}) \left\{ 1 - \frac{F\sigma t(e^{-F\sigma t} - e^{-\lambda_0 t})}{(1 - e^{-F\sigma t})(\lambda_0 - F\sigma)t} \right\}. \qquad (5)$$

The factor in the brackets gives the reduction over the simpler case in which the lifetime of the parent can be neglected. This factor is plotted as a function of the half life of the parent for different values of $F\sigma t$ in Fig. 5. The reduction is not very sensitive to values of $F\sigma t$. A parent with half life of one day will reduce the poisoning effect about one-third.

<u>Estimation of Poisoning in Pile with Flux of $4 \times 10^{14}/cm^2$ After One Day's Operation</u>

The graphs and formulae have been applied to the special problem of estimating the poisoning in a pile with the high flux of $4 \times 10^{14}/cm^2$ at the end of one day. The contributions divide themselves into three parts:

(1) <u>Contribution from stable isotopes which are the end products of fission chains.</u>—The cross sections of many of these isotopes are known. In the cases where they are not known upper limits can be set from known total absorption cross sections. The poisoning from stable elements was estimated by Wheeler in July, 1943 (CP-889) and at his suggestion, Dempster

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investigated the absorption of samarium and found the isotope with the large cross section to be Sm^{149}. This identification removed most of the uncertainty about the contribution of the stable isotopes. Wheeler's table, revised and brought up to date, is given in Appendix I. The results are:

	Max	Min
Present $\Sigma y\sigma$ for stable isotopes whose absorption cross sections are known or for which an upper and lower limit can be estimated from the cross section for all isotopes. y given as a fraction, not as percent.	703	637
Wheeler's old values for the above	670	7

The large contributions to the total come from

Sm^{149}	$Gd^{155,157}$	$Eu^{151,153}$	Rh^{103}	Total of these four	Total of all others
600	19	7	8	634	69

As has already been pointed out, however, for large values of $F\sigma$, the poisoning is no longer given directly by $p = Fy\sigma t$ even for stable nuclei but rather by $p = y\left[1 - e^{-F\sigma t}\right]$. For t = 1 day and $F = 4 \times 10^{14}$ the maximum poisoning from stable elements is then

Element	Yield	σ	$p = y\left[1 - e^{-F\sigma t}\right]$
Sm^{149}	.015	40,000	.01125
Gd^{155}	.0004	33,000	.00027
Gd^{157}	.00006	100,000	.00006
Eu^{151}	.004	1,520	.00021
Eu^{153}	.0013	768	.00003
For all others $\Sigma y\sigma = 69$			.00273
			.01455 = 1.46%

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The yield of Sm^{149} has been taken from the Handbook yield curve for fission products. An experimental determination of the yield and some information as to the lifetime of the parent of Sm^{149} are necessary before an exact value for the samarium poisoning can be given.

(2) Contribution from Xe^{135}.—The general formula (Eq. 1) was used here with the half life of the I parent taken as 6.6 hrs, the half life of Xe^{135} as 9.4 hrs and the yield as 4.3%. The result for the poisoning at the end of a day is

$$p(Xe^{135}) = 3.9\% \ .$$

(3) Contribution from radioactive isotopes in fission chains with unknown cross sections.—In Wheeler's original memorandum long-lived radioactive isotopes were included with stable isotopes and an upper limit for their contribution to the poisoning estimated for an assumed cross section of 23,000 barns. Their probable poisoning effect is estimated here in a different way. A separate list of 37 fission products with half lives greater than one day is given in Appendix II. An analysis of the isotopes on this list that are not shielded by long lived parents shows:

	Number of these with yields greater than 1%	Number of these with yields less than 1%	Total
Isotopes with even numbers of neutrons	9	10	19
Isotopes with odd numbers of neutrons	7	7	14

A study of cross section statistics for isotopes with even and odd neutrons has already been made (MUC-KM-35).

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These statistics were used to find the fraction of isotopes with even or odd numbers of neutrons with cross sections of certain sizes with the exception that in the case of isotopes with an odd number of neutrons no isotopes were assigned cross sections greater than 10^5 barns. The reason is that according to CP-2301 there is evidence that no long lived fission product isotope with cross section greater than this exists other than Xe^{135}. The poisoning is then given by

$$p = \text{no. isotopes} \times \text{average yield} \times \sum n_i \times f_i \quad .$$

n_i is equal to the fraction of isotopes with cross sections in a certain range. f_i is found from Equation (3) for the average cross section in the range. It is assumed that all the lifetimes are sufficiently large for maximum values to be reached; i.e., that $f = 1 - e^{-F\sigma t}$.

ISOTOPES WITH ODD NUMBERS OF NEUTRONS AND HIGH YIELD

no. of isotopes = 6* average yield = 5.0%

σ_i	n_i	f_i	$n_i f_i$
3.16	.055	1.1×10^{-4}	.000
3.16×10	.390	1.1×10^{-3}	.000
3.16×10^2	.278	1.1×10^{-2}	.003
3.16×10^3	.055	.103	.006
3.16×10^4	.166	.624	.103
3.16×10^5	.055	1.00	.055
			.167

$$p = 6 \times 5\% \cdot .167 = 5.01\%$$

*The number 6 was used instead of the total number 7 given above because the contribution of two of the isotopes is reduced due to shielding by a longer lived ancestor.

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ISOTOPES WITH EVEN NUMBERS OF NEUTRONS AND HIGH YIELD

no. of isotopes = 9 . average yield = 5%

σ_i	n_i	f_i	$n_i f_i$
3.16×10^{-3}	.018	1.1×10^{-7}	.0000
3.16×10^{-2}	.125	1.1×10^{-6}	.0000
3.16×10^{-1}	.286	1.1×10^{-5}	.0000
3.16	.322	1.1×10^{-4}	.0000
3.16×10^{1}	.143	1.1×10^{-3}	.0002
3.16×10^{2}	.090	1.1×10^{-2}	.0010
3.16×10^{3}	.018	1.0×10^{-1}	.0018
			.0030

$p = 9 \times 5\% \times .003 = .145\%$

For both classes of isotopes with yields less than 1%, the average yield may be conservatively put at .2%. Using the above values of $\sum n_i f_i$, one then finds:

Isotopes with odd neutrons and low yield:

$p = 7 \times .2\% \times .167 = .234\%$

Isotopes with even neutrons and low yield:

$p = 10 \times .2\% \times .003 = .006\%$

The following table summarizes various contributions to the local poisoning in a pile operating at a flux of 4×10^{14} neutrons/cm^2/day at the end of one day:

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CONTRIBUTIONS TO POISONING AT END OF ONE DAY.

Flux = 4 x 10^{14} Neutrons/cm^2/sec

Poison		Ratio of no. of neutrons absorbed in poison to no. absorbed in metal in percent
Samarium^{149}		1.13%
All other stable isotopes		.33
Xenon^{135}		3.9
Radioactive isotopes with odd neutrons	High yields	5.01
	Low yields	.23
Radioactive isotopes with even neutrons	High yields	.15
	Low yields	.01
Total from these sources		10.76%

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APPENDIX I

Table Showing Contribution to Poisoning by Stable Fission Products

(1) All cross sections are in barns. One barn = 10^{-24} cm^2.

(2) Isotopic cross sections are used where known. When not known, the total absorption cross section which cannot be accounted for is assigned in turn to the isotopes for which the values of the yield/abundance are the greatest and smallest to get upper and lower limits for the quantity, yield x cross section.

(3) There are now only three elements in the fission product region in which only the absorption plus scattering cross sections is known: Kr, Ru, Xe. In these three cases the total cross section is used as the absorption cross section.

(4) The yield, or branching ratio, which is equal to the number of times per fission that a given fission product is formed, was taken where possible, from values given in the Handbook, III D-1, by Brady and Turkevich. When no measured values were available, the yields were estimated from the smooth curve, also in III D-1, which was revised in accordance with more recent data in CS-2135 and CC-2000.

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Element and total cross section	σ (isotopic) where known	Isotopes	Natural isotopic abundance	Yield	Yield / abundance	Yield x σ maximum	Yield x σ minimum
33 - As	4.6	75	1.00	0.00001			
34 - Se	22	74	0.009	—	—		
		76	.095	—	—		0
σ_a = 12		77	.083	0.00008	0.001		
		78	.240	.0002	.001		
	.54	80	.480	.0009	.002	0.001	.001
		82	.093	.0025	.027	.31	
35 - Br	12.0	79	.506	.0004		.005	.005
	2.25	81	.494	.0012		.002	.002
36 - Kr		78	.0035	—	—		
		80	.0201	—	—		
σ_a < 20		82	.1153	—	—		0
		83	.1153	.0030	.026		
		84	.5710	.0065	.011		
		86	.1747	.022	.126	2.5	
37 - Rb	.69	85	.723	.015		.010	.010
σ_a = 11.8	.128	87	.277	.030		.004	.004
38 - Sr		84	.0056	—	—		
	1.4	86	.0986	—	—		
σ_a = 1.2		87	.0702	—	—		
	.005	88	.8256	.040	.025	.0002	.0002
39 - Y	1.1	89	1.00	.046		.056	.056
40 - Zr		90	.48	—	.104		.29
		91	.115	.058	.505	1.41	
σ_a = 2.8	.33	92	.22	.059		.019	.019
	.053	94	.17	.065		.003	.003
	1.07	96	.015	.066		.071	.071
41 - Cb	.02 } (?)	93	1.00	.062		0.074	.009
	1.2 } (?)						
42 - Mo		92	.155	—	—		
		94	.087	—	—		
σ_a = 2.5		95	.163	.045	.276		
		96	.168	—	—		0
		97	.087	.065	.748	1.87	
	.37	98	.254	.062		.023	.023
	.23	100	.086	.053		.012	.012
44 - Ru		96	.05	—	—		
		99	.12	—	—		
σ_a = 6		100	.14	—	—		0
		101	.22	.043	.195	1.17	
	1.2	102	.30	.039		.047	.047
	.33	104	.17	.018		.006	.006
45 - Rh	163	103	1.00	.047		7.7	7.7

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Element and total cross section	σ (isotopic) where known	Isotopes	Natural isotopic abundance	Yield	Yield / abundance	Yield x σ maximum	Yield x σ minimum
46 - Pd		102	0.008	—	—		
		104	.093	—	—		0
$\sigma_a = 7$		105	.226	0.009	0.040	0.147	
		106	.272	.005	.018		
	12.1	108	.268	.0005		.006	.006
	.63	110	.135	—	—		
47 - Ag	48.3	107	.525	—			
	108	109	.475	.0001		.01	.01
48 - Cd		111	.130	.00006	.00046	1.4	
		112	.242	.00004	.0002		
$\sigma_a = 3000$		113	.123	—			0
	1.24	114	.280	.00001			
	1.4	116	.073	.000014			
49 - In	61	113	.045	.00002		.001	.001
	213	115	.955	.000008		.002	.002
50 - Sn		119	.098	.000008	.00008		
		120	.285	.00001	.00003		.000
$\sigma_a = .55$		122	.055	.00004	.00073	.000	
	.574	124	.068	.00014	.00206	.000	.000
51 - Sb	6.8	121	.560	.00002		.000	.000
	2.5	123	.440	.00007		.080	.000
52 - Te		122	.029	—	—		
		123	.016	—	—		
$\sigma_a = 3.5$		124	.045	—	—		0
		125	.060	.00025	004	.012	
	.85	126	.190	.00055		.001	.001
	.148	128	.328	.0050		.001	.001
	.250	130	.331	.018		.005	.005
54 - Xe		124	.00094	—	—		
		126	.00088	—	—		
$\sigma_a < 25$		128	.0190	—	—		
		129	.2623	—	—		
		130	.0407	—	—		0
		131	.2117	.028	.133		
		132	.2696	.038	.141		
		134	.1054	.053	.503		
		136	.0895	.062	.693	17.3	
55 - Cs	25.6	133	1.00	.043		1.1	1.1
56 - Ba		130	.00101	—	—		
		132	.00097	—	—		
$\sigma_a = 1.0$		134	.0242	—	—		
		135	.0659	—	—		
		136	.0781	—	—		0
		137	.1132	.065	.574	.57	
	.56	138	.7166	.066		.04	.04
57 - La	8.4	139	1.00	.065		.55	.55
53 - I	6.8	127	1.00	.0012		.008	.008

[redacted]

Element and total cross section	σ (isotopic) where known	Isotopes	Natural isotopic abundance	Yield	Yield / abundance	Yield x σ maximum	Yield x σ minimum
58 - Ce		138	<0.01	—			0
		140	.90	0.060	0.066		
$\sigma_a < 3$		142	.10	.060	.60	1.8	
59 - Pr	11.0	141	1.00	.053		.58	.58
60 - Nd		142	.2595	—	—		0
		143	.130	.058	.450		
$\sigma_a = 70$		144	.226	—	—		
		145	.092	.050	.54	38	
		146	.165	.040	.24		
		148	.068	.022	.32		
		150	.0595	.008	.13		
62 - Sm		144	.03	—	—		
		147	.17	.014	.082		
$\sigma_a = 6000$	280	148	.14	—	—		
	40000	149	.15	.015	.100	600	600
	96	150	.05	—	—		
		152	.26	—	—		
		154	.20	—	—		
63 - Eu	1520	151	.491	.004		6.1	6.1
	768	153	.509	.0012		.9	.9
64 - Gd	33300	155	.156	.0004		13.3	13.3
		156	.206	.00019			
$\sigma_a = 25000$	100000	157	.164	.00006		6.0	6.0
		158	.234	.00007			

Total cross section for fission, i.e., $\Sigma y\sigma$, contributed by stable elements — 703 (maximum), 637 (minimum)

Chief uncertainties in stable elements

Nd	Xe	Total
38	17	55

Biggest Contributors

Sm	Gd	Eu	Rh	Total
600	19	7	8	634

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APPENDIX II

List of Radioactive Fission Products With Half Lives Greater than One Day

(A) Elements with Odd Numbers of Neutrons

Element	Half Life	Yield in %	Parent with life time greater than one hour	Gaseous ancestor that might make it possible to remove chain
Sr^{89}_{38}	55 d	4.6		Yes
$Zr^{93,95}_{40}$	65 d	> 4.5	11 hr Y	? *
Mo^{99}_{42}	2.8 d	5.8		No
Ru^{103}_{44}	42 d	< 4.7		No
Pd^{107}_{46}	long ?	0.04	30 d Rh	No
Cd^{115}_{48}	43 d	.001		No
Cd^{115}_{48}	2.5 d	.01		No
Sn_{50} ?	11 d	.0045		No
Te^{127}_{52}	90 d	.03		No
Te^{129}_{52}	32 d	.19		No
Te^{131}_{52}	1.3 d	~ .5		No
Xe^{133}_{54}	5.3 d	3.8	22 h I	?
La^{140}_{57}	1.67 d	6.1	12.5 d Ba	Yes
Ce^{141}_{58}	28 d	5.0	3.5 h La	?
Ce^{143}_{58}	1.4 d	5.4	74 m La	?
Eu_{63}	160 d	.006		No

* ? means that a gaseous ancestor has been found for the chain but that it seems unlikely that the chain starts with the gas most of the time.

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(B) Elements with Even Numbers of Neutrons

Element	Half Life	Yield in %	Parent with life time greater than one hour	Gaseous ancestor that might make it possible to remove chain
Sr^{90}_{38}	30 y	5		Yes
Y^{91}_{39}	57 d	5.9	8.5 h, Sr	?
$Cb^{93,95}_{41}$	35 d	> 4.5	.65 d Zr	No
$Cb^{93,95}_{41}$?	3.3 d	> 4.5	65 d Zr	No
$^{99}_{43}$	long	5.8	67 h Mo	No
Ru^{106}_{44}	330 d	0.5		No
Ru^{108}_{44} ?	60 d	.006		No
Rh^{107}_{45} ?	30 d	.04	100 m Ru	No
Rh^{109}_{45} ?	4 - 5 d	0.5		No
Ag^{111}_{47}	7.5 d	.006		No
Sb^{127}_{51}	3.9 d	.13	80 m Sn ?	No
Te^{132}_{52} ?	3.2 d	3.6		No
I^{129}_{53}	long	0.24	72 m Te	No
I^{131}_{53}	8 d	1.7		No
Cs^{135}_{55} ?	~25 y	4.3		?
Ba^{140}_{56}	12.5 d	6.1		Yes
Ce_{58}	300 d	5.5		No
Pr^{143}_{59}	13.5 d	5.4	35 h Ce	No
Nd_{60} ?	300 d			No
61 ?	long			No
Eu_{63} ?	16 d	.019		No

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FIG. 1. Poisoning by Isotopes with Different Half Lives as a Function of the Time of Operation

$$\text{Ordinate} = \frac{\text{neutrons absorbed in poison}}{\text{neutrons absorbed in metal x yield}} = \frac{p}{y} = \frac{\nu\sigma}{\nu\sigma + \lambda}\left[1 - e^{-(\nu\sigma+\lambda)t}\right]$$

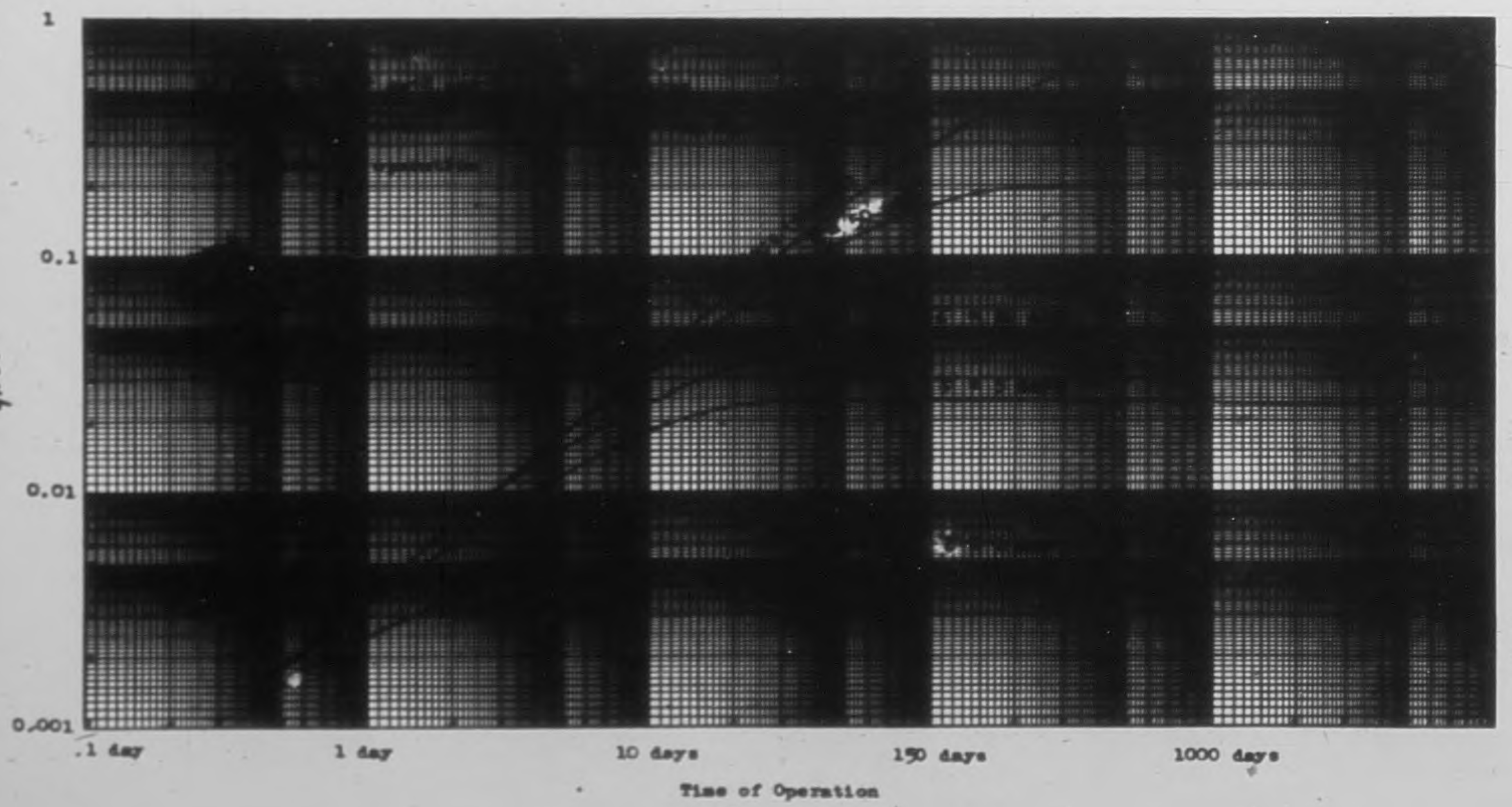

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Fig. 2. Poisoning by Isotopes with Different Half Lives as a Function of the Time of Operation

$$\text{Ordinate} = \frac{\text{neutrons absorbed in poison}}{\text{neutrons absorbed in metal x yield}} = \frac{p}{y} = \frac{F\sigma}{F\sigma + \lambda}\left[1 - e^{-(F\sigma + \lambda)t}\right]$$

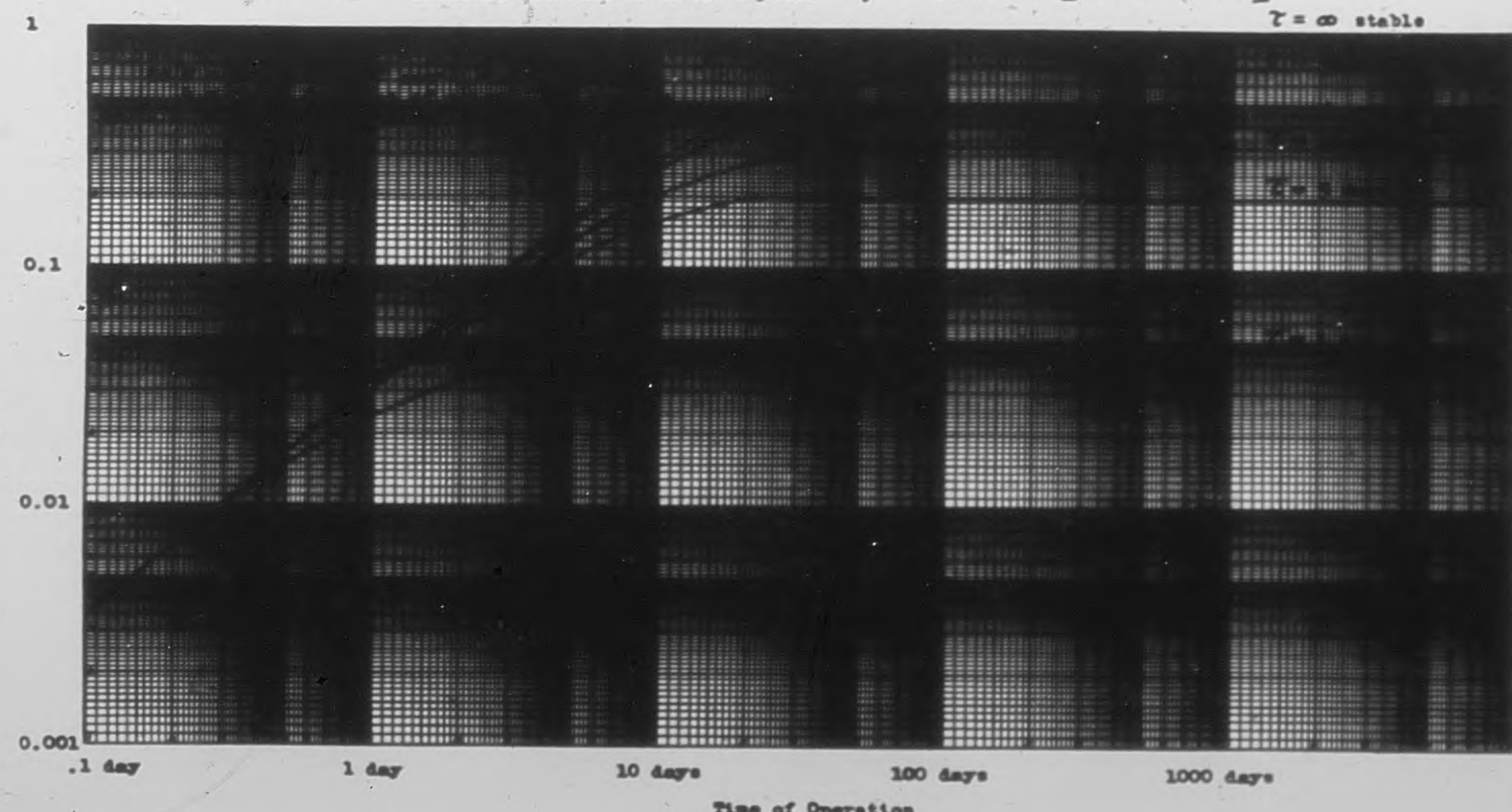

FIG. 3. Poisoning by Isotopes with Different Half Lives as a Function of the Time of Operation

$$\text{Ordinate} = \frac{\text{neutrons absorbed in poison}}{\text{neutrons absorbed in metal x yield}} = \frac{p}{y} = \frac{F\sigma}{F\sigma + \lambda}\left[1 - e^{-(F\sigma + \lambda)t}\right]$$

τ = 50 days τ = stable

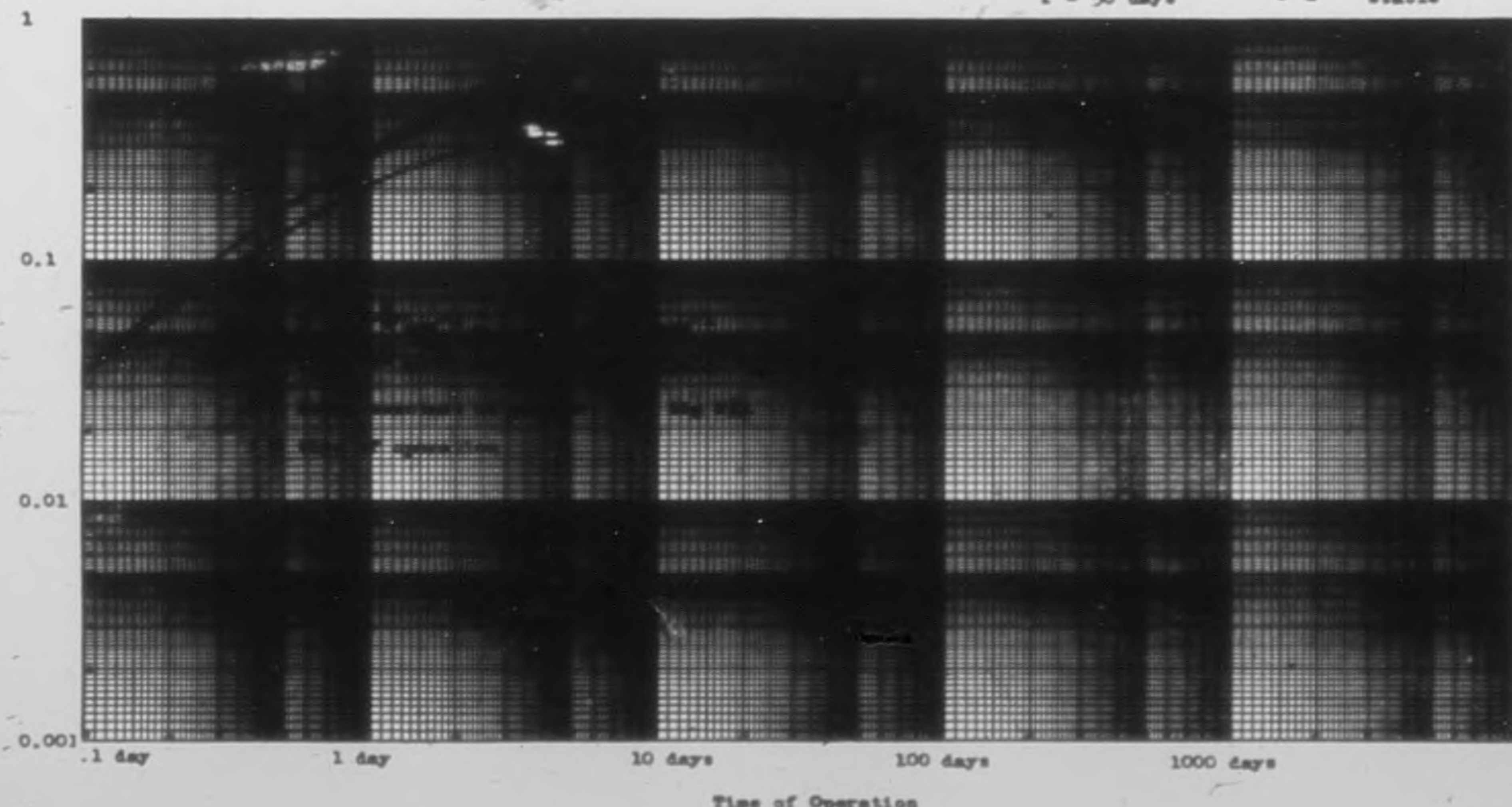

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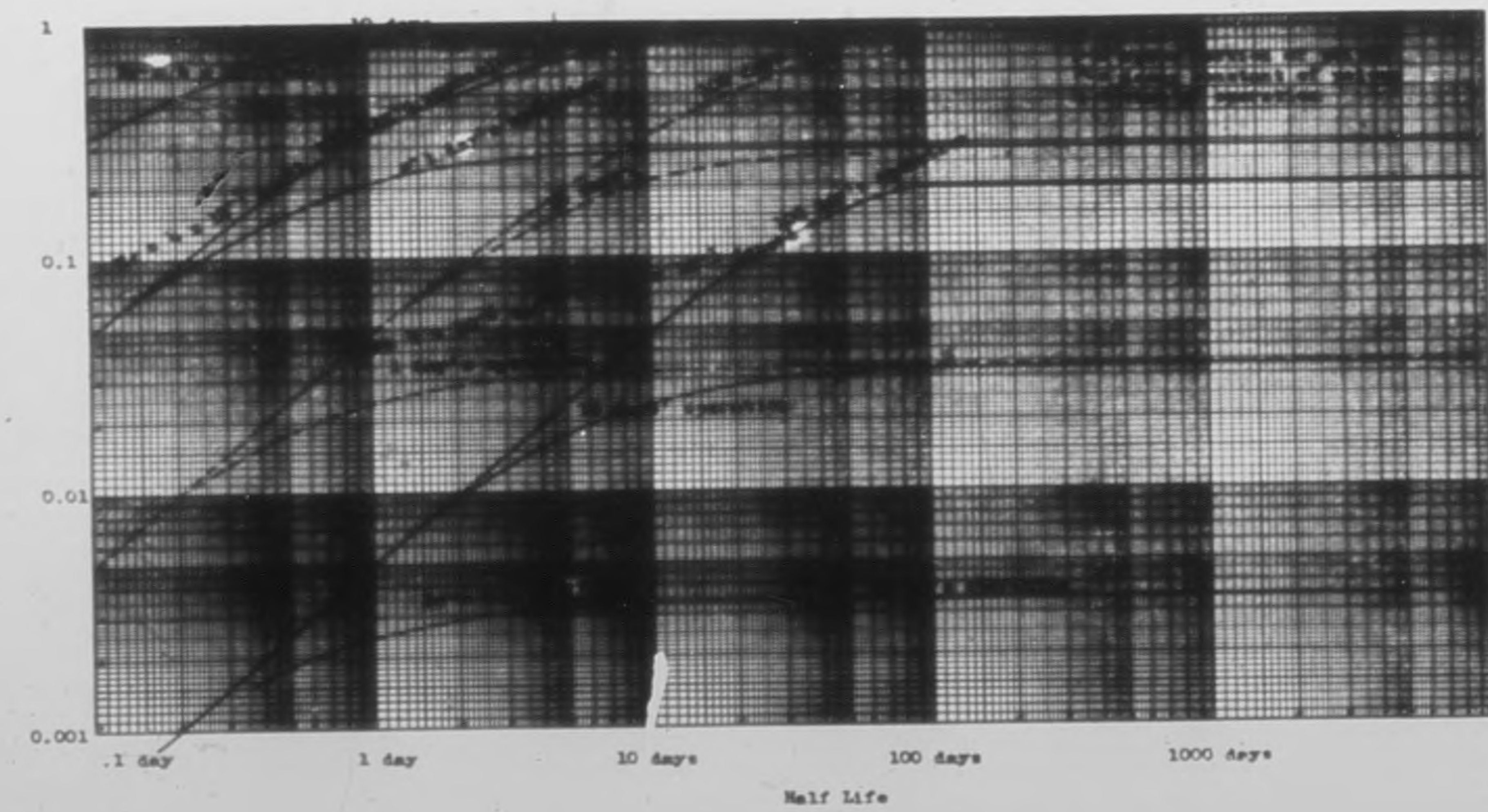
FIG. 4. Poisoning at Different Times of Operation as a Function of the Half Life of the Poison

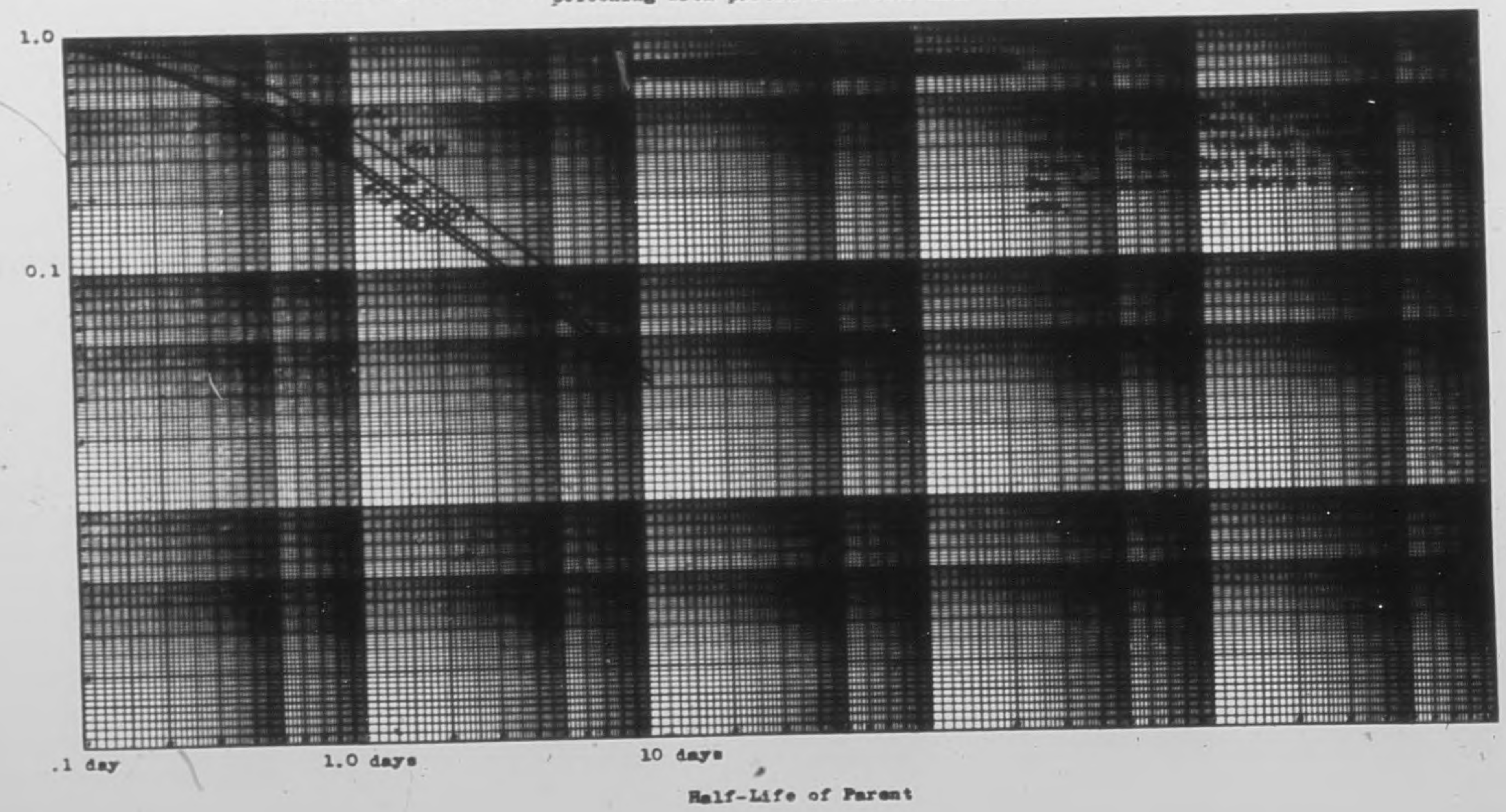
$$\text{Ordinate} = \frac{\text{neutrons absorbed in poison}}{\text{neutrons absorbed in metal}} \frac{1}{\text{yield}} = \frac{p}{y} = \frac{\Phi\sigma}{\Phi\sigma + \lambda}\left[1 - e^{-(\lambda + \Phi\sigma)t}\right]$$



CP-2462

FIG. 5. Reduction in Effect of Stable Poison due to Half Life of Parent at End of One Day

Ordinate gives ratio, $\frac{\text{poisoning with parent with given half life}}{\text{poisoning with parent with zero half life}}$. See Equation (5).



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