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POISONING AND PURIFICATION

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Discussion and Results

In any pile operating at a high flux, say five or ten times the W level, the growth of poisonous fission products will be very rapid. In an "enriched" pile with a high reproduction factor the poisoning may not be fatal to the chain reaction. However, if the neutrons in such a pile which are not needed for the continuance of the reaction are to be used for the production of new fissionable material, as in a converter or breeder, their loss to a poison may produce a serious lowering of the efficiency of the machine.

It may be possible to purify the reacting metal by taking out a fraction of the fissionable material from the reactor either continuously or in small batches and removing the poisons from this fraction by chemical means. However, in such a process a certain amount of the fissionable metal will also be removed unavoidably. The more rapid the cycle, the greater this latter loss will be, although at the same time the loss of neutrons to poison in the pile will be smaller. The question arises as to the most favorable purification rate for a given operating level.

In a converter or breeder, the absorption of a neutron by a poison is equivalent to the loss of a fissionable atom. The quantity

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which it is desired to keep at a minimum is thus the following fraction which will be called the loss:

$$\text{Loss} = \frac{(\text{Fissionable atoms lost in purifier}) + (\text{Neutrons absorbed by poisons in pile})}{\text{Neutrons absorbed by metal in pile}}$$

The chief difficulty at present in evaluating this quantity is that very little is known about the slow neutron capture cross sections of radioactive fission products and these may produce a large poisoning effect. However, cross sections can be assigned to these isotopes on the basis of data on stable isotopes. The results from such a procedure may be considerably in error but give the best idea of what can be reasonably expected at the present time.

The loss is a function of the fraction C of fissionable atoms removed in the purifier and the fraction $\alpha\bar{t}$ of the reacting material which is purified in the lifetime $\bar{t}$ of a metal atom in the pile. For a given C, the loss will be a minimum for a certain value of $\alpha\bar{t}$. The relation between C and $\alpha\bar{t}$ necessary to give the minimum is shown in Fig. I. Here one sees for example that if C is 0.1%, $\alpha\bar{t}$ should be about 14; that is, if the lifetime of the metal is 100 days the purification cycle should be about a week. If C is 1%, for the same metal lifetime, the optimum rate is about once every six weeks.

The actual minimum loss for any given C or any given $\alpha\bar{t}$ is shown in Fig. II. For instance, for a C of 1% the loss as defined above is 5.9% while for a C of 1% the loss is 9.7%.

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The results were found assuming the purification process to be continuous. If the whole unit is purified once in a time equal to $2/\alpha$ the average poisoning during this time interval is only about 1.14 times the poisoning with continuous purification in a cycle of length $1/\alpha$. Further discussion of batchwise purification is given below.

It is interesting to inquire how much these results will be changed if the radioactive nuclei do not follow the statistics. The information available at present is that nuclei with an odd number of neutrons have in general higher cross sections than those with an even number. However, this apparent difference may be due to the fact that so few cross sections of odd neutron isotopes are known.* According to the present statistics (see Appendix I of CP-2468) one out of eighteen odd neutron nuclei will have a cross section between 10^5 and 10^6 barns, while three will have cross sections between 10^4 and 10^5 barns. This means that of the eight radioactive odd neutron fission products with high yields, two may be expected to have cross sections greater than 10^4 barns or of the same order of magnitude as Sm^{149} (40,000 barns). The contributions of such poisons to the loss, as shown below in Table I, amount to $\sim$50% of the total for a purification cycle of 8 days, and to $\sim$37% for an 65 day cycle. ($\bar{t}$ = 100 days.)

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*Only 4 cross sections of odd neutron isotopes have been measured. The upper limits of 14 more have been found by assigning that part of the absorption cross section of an element which cannot be accounted for by activation cross sections to the odd neutron isotope with least abundance.

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If any radioactive nuclei actually have such large cross sections it seems that their effect might have been apparent at W. Analysis of reactivity changes at W (N-1771) show that at low power the data can be reasonably well accounted for by assuming a yield of Sm of only 0.35% and a total effect of all other fission products equal to one-third the Sm effect. However, it is not known at present whether any anti-poisons are created at W. If U^{236} or U^{237}, for instance, have large cross sections for fission with slow neutrons their growth will help rather than hinder the chain reaction. These nuclei will not be present in converters using Pu^{239} or breeders using U^{233} as the fissionable metals. The anti-poisoning effect seems very unlikely in the case of U^{236} but quite possible for U^{237}. Therefore, until further experiments are done, the most conservative policy is to assume that some radioactive isotopes have large cross sections.

In the next three sections the mathematical expressions for the loss and its derivatives are given, the method of taking into account the cross sections of the two hundred stable and radioactive fission products described, and the contributions to the loss of different classes of fission products tabulated for different values of $\alpha \bar{\bar{t}}$.

Mathematical Formulae

1. A Single Stable Poison.--The simple case of a single stable poison produced directly in fission is treated first.

Let n = the number of poison atoms present at any time,

σ = slow neutron absorption cross section of the poison,

σ_f = fission cross section of the metal,

σ_a = total (fission plus capture) cross section of the metal,

$\sigma_f/\sigma_a = \beta$,

$\sigma/\sigma_a = \gamma$,

$\bar{t}$ = lifetime of metal atom = 1/flux x σ_a,

τ = time/$\bar{t}$,

y = yield or branching ratio of the poison in question,

α = fraction of reacting material purified per unit time,

$\alpha\bar{t}$ = fraction purified in lifetime of metal,

N = number of metal atoms present (it is assumed that this number is kept constant),

C = fraction of metal atoms removed in the purification process.

Then

$$dn/d\tau = N\beta y - (\alpha\bar{t} + \gamma)n$$

and

$$n = \frac{N\beta y}{\alpha t + \gamma}\left[1 - e^{-(\alpha\bar{t} + \gamma)\tau}\right]. \qquad (1)$$

This quantity will approach its equilibrium value in a time equal to $1/\alpha$ if γ is smaller than $\alpha\bar{t}$ and in a shorter time if it is larger. All calculations are made assuming the equilibrium value has been reached; that is, that n has the constant value

$$n = \frac{N\beta y}{\alpha\bar{t} + \gamma} \quad . \qquad (2)$$

The number of neutrons absorbed by the poison per unit time is $n\sigma/\sigma_a\bar{t}$. The fissionable atoms lost in the same time in the purification process is $NC\alpha$. Thus the loss is

$$\text{Loss} = C\alpha\bar{t} + \frac{y\beta\gamma}{\alpha\bar{t} + \gamma} \quad . \qquad (3)$$

The loss is a minimum when

$$C = \frac{y\beta\gamma}{(\alpha\bar{t} + \gamma)^2}$$

or

$$\frac{\alpha\bar{t}}{\gamma} = \left(\frac{\beta y}{C\gamma}\right)^{1/2} - 1 \quad . \qquad (4)$$

The right hand side of (4) must be positive, which means

$$C\gamma < \beta y \, .$$

If this condition is not fulfilled, no minimum will exist and the loss will always increase with increasing α. In other words, an attempt at purification will only make things worse. For instance if Sm^{149} were the only fission product poison, C would have to be 2.68×10^{-4} to make purification worthwhile. (This results from σ = 40,000 barns, $y = 0.015$, σ_f = 715b, σ_a = 1050b.)

2. <u>A Single Radioactive Poison with Long-Lived Parent.</u>--A radioactive poison with a parent which partially shields it is now considered. This is really the most general case. Let the subscripts 0 and 1 refer to the parent and daughter respectively, let a be the decay constant times $\bar{t}$, and the other symbols have the meanings already given. Then

$$n(\tau) = N\beta y a_o \left\{ \frac{1 - e^{-(\alpha\bar{t} + \gamma + a_1)\tau}}{(\alpha\bar{t} + \gamma_o + a_o)(\alpha\bar{t} + \gamma_1 + a_1)} + \frac{e^{-(\alpha\bar{t} + \gamma_1 + a_1)\tau} - e^{-(\alpha\bar{t} + \gamma_o + a_o)\tau}}{(\alpha\bar{t} + \gamma_o + a_o)(\gamma_1 - \gamma_o + a_1 - a_o)} \right\} \quad (5)$$

and at equilibrium

$$n = \frac{N\beta y a_o}{(\alpha\bar{t} + \gamma_o + a_o)(\alpha\bar{t} + \gamma_1 + a_1)} . \quad (6)$$

The loss is then easily found to be

$$\text{Loss} = C\alpha\bar{t} + \frac{\beta\gamma_1 y a_o}{(\alpha\bar{t} + \gamma_o + a_o)(\alpha\bar{t} + \gamma_1 + a_1)} \quad (7)$$

and the condition that a minimum exist is now

$$C = \frac{\gamma_1 y \beta a_o}{[\gamma_1 - \gamma_o + a_1 - a_o]} \left\{ \frac{1}{(\alpha\bar{t} + \gamma_o + a_o)^2} - \frac{1}{(\alpha\bar{t} + \gamma_1 + a_1)^2} \right\} . \quad (8)$$

3. <u>More Than One Poison.</u>--When there are many different poisons present the loss can be found by adding poison terms for all the isotopes involved to (7) and (8).

4. Correction if Purification is not Continuous.--If the purification is not done continuously but rather in batches; that is, if a fraction of the reacting metal α' is removed at the end of each time interval T then the average amount of a poison present is

$$\frac{N\beta y}{\gamma + a}\left\{1 - \frac{\alpha'\bar{t}}{T(\gamma + a)}\left(1 - e^{-(\gamma + a)T/\bar{t}}\right)\right\}.$$

For continuous purification the amount is $N\beta y/(\alpha\bar{t} + \gamma + a)$. The ratio R of the poisoning due to this particular isotope for batchwise to continuous purification is then

$$R = \frac{1 + \zeta}{\zeta}\left\{1 - \frac{\alpha'}{\alpha T\zeta}\left(1 - e^{-\zeta\alpha T}\right)\right\}$$

where ζ has been substituted for $(\gamma + a)/\alpha\bar{t}$. If $\alpha' = 1$ and $T = 2/\alpha$

$$R = 1 + \left\{\frac{1 + e^{-2\zeta}}{2\zeta} - \frac{1 - e^{-2\zeta}}{2\zeta^2}\right\}.$$

The quantity in the brackets has a maximum value of $\sim .140$.

Fission Product Data

If the effect of many poisons is to be evaluated exactly, there should be one term for each radioactive or stable fission product, or over two hundred terms altogether. In order to reduce the labor involved the fission products have been divided into various classes and each class included as a whole. A primary subdivision into stable and radioactive isotopes was natural since, as already mentioned, the cross section of only one radioactive product has been measured so that it is impossible to include the radioactive nuclei in any way other than a statistical one. These two main groups were then still further subdivided.

Stable Fission Products.--Sixty-three stable fission products are listed in Appendix I of CP-2468 as having appreciable fission yields or branching ratios. Of these, only 31 have known cross sections. However, in most* cases the total absorption cross section of the element is known. That part of the total cross section not accounted for by known activation cross sections was assigned to that isotope for which the ratio of yield to abundance is the largest and the other isotopes then assumed to have zero cross section. Thirteen additional stable isotopes are added in this way making a total of 44. Stable isotopes having radioactive parents with half lives of a year or more were not included in the original list. Their four radioactive parents have, however, been added to the stable class and cross sections assigned to them in a statistical way. Moreover, four of the stable isotopes have ancestors with half lives of a month or more. The stable isotopes were thus divided into two very unequal classes.

Class Ia: 40 stable isotopes which are assumed to be formed directly in the fission process; i.e., to have parents with negligible half-lives.

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*For Rb, Ru, and Kr the total absorption plus scattering cross section was used for the absorption cross section.

Their cross sections, assigned as explained above, vary over a much wider range than their yields. For this reason they were divided into eight groups according to cross section and the yields in each group averaged. The first group includes all isotopes with cross section between 10^{-3} and 10^{-2} barns, the last those with cross sections between 10^4 and 10^5 barns. The average cross section in each group was taken to be the square root of the highest cross section in the group. For instance, for the first group, the cross section is taken as 3.16×10^{-3} barns. Appendix I lists all the elements in this class together with their yields and cross sections and shows how they were arranged into cross section groups.

Class Ib: The four stable isotopes in this class are listed in Appendix II together with their cross sections, yields, parents, etc. Since the members of this class are so few, their contributions to equations (7) and (8) were evaluated individually.

Radioactive Isotopes.--In Appendix II of CP-2468, there is a list of 33 radioactive isotopes with half lives greater than one day. Of these, 14 have an odd number of neutrons and 19 an even number of neutrons. Of the 14 with an odd number of neutrons, only 6 have yields less than 0.5% while 8 have yields of about 5%. The same sort of thing is true for the even numbered neutron nuclei. Here 9 have yields around 5% while the others have yields of 0.5% or less. The low yield isotopes were neglected in both cases. Cross sections were assigned to the two high yield groups on the basis of the statistics given in CP-2468 and

MUC-KW-37. It was noticed that the spread in the half lives was not very great and that these could be divided into groups pretty well. The groups that resulted are given below. The specific nuclei put into these groups are given in Appendix III and the cross section statistics used will be found in CP-2468.

Class IIa. 4 isotopes with an odd number of neutrons and long half-lives. The average half-life is taken as 45 days.

Class IIb. 4 isotopes with an odd number of neutrons and short half-lives. The average half-life = 2.8 days.

Class III. 5 isotopes with an even number of neutrons and an average half-life of 19 days. The other four nuclei with an even number of neutrons and high yields have half-lives of the order of a year or more. The statistics were applied to these nuclei and the contributions added to those from Class Ia.

<u>Contributions of Different Classes to the Loss</u>

The amount of loss due to the different classes is of considerable interest. Table I shows the losses from all the classes and from the cross section groups within the classes for three different values of α. It brings out clearly the fact that the radioactive nuclei with odd numbers of neutrons make a contribution in one case equal to half the poisoning loss. This table can be used to find the loss if experiments should show that certain groups which have a big poisoning effect on the statistical picture can actually be neglected.

TABLE I

CONTRIBUTIONS OF THE VARIOUS CLASSES TO THE POISONING LOSS*

$\alpha \bar{t} = .381$

τ	Ia	Ib	IIa	IIb	III	Total	%
1 - 10	6.05	.479	.0391	.00245	.155	6.73	1.33
10 - 10^2	57.9	6.15	2.79	.177	.685	67.7	13.4
10^2 - 10^3	168	23.7	17.1	1.23	4.04	214	42.3
10^3 - 10^4	17.9		15.0	2.15	4.90	39.9	7.89
10^4 - 10^5	1.28		68.4	30.6		100	19.8
10 - 10^6			24.0	16.1		40.1	7.93
4 x 10^4 (Sm^{149})	37.1					37.1	7.34
Total	288	30.3	127	50.2	9.78	505	
%	57.0	6.00	25.1	9.93	1.94		

$\alpha \bar{t} = 1.52$

1 - 10	.381	.0182	.00604	.000582	.0300	.436	.566
10 - 10^2	3.83	.370	.433	.0421	.133	4.81	6.24
10^2 - 10^3	15.7	1.26	2.78	.291	.793	20.8	27.0
10^3 - 10^4	3.15		3.03	.513	1.05	7.74	10.0
10^4 - 10^5	.309		16.5	7.48		24.3	31.5
10^5 - 10^6			5.97	4.00		9.97	12.9
4 x 10^4 (Sm^{149})	9.01					9.01	11.7
Total	32.4	1.65	28.7	12.3	2.01	77.1	
%	42.0	2.14	37.2	16.0	2.61		

$\alpha \bar{t} = 12.2$

1 - 10	.00595	6.17×10^{-5}	.000166	4.9×10^{-5}	.00120	.00643	.120
10 - 10^2	.0609	.00425	.0120	.00355	.00534	.0860	1.61
10^2 - 10^3	.286	.00518	.0821	.0247	.0330	.431	8.07
10^3 - 10^4	.117		.136	.0450	.0565	.354	6.63
10^4 - 10^5	.0289		1.56	.777		2.37	44.4
10^5 - 10^6			.721	.481		1.21	22.7
4 x 10^4 (Sm^{149})	.888					.888	16.6
Total	1.39	.00949	2.51	1.33	.0960	5.34	
%	26.0	.178	47.0	24.9	1.80		

*Entries in the table are the values of loss x $10^6/\alpha \bar{t} \sigma_f$

APPENDIX I

Class Ia. Unshielded Stable Isotopes

Cross section: 10^{-3} - 10^{-2} barns

Element	σ	Yield
Sr^{88}	.005	.040

Cross section: 10^{-2} - 10^{-1} barns

Element	σ	Yield
Zr^{94}	.053	.065

Cross section: 10^{-1} - 10^{0} barns

Element	σ	Yield
Rb^{85}	.69	.015
Rb^{87}	.128	.030
Zr^{92}	.33	.059
Mo^{98}	.37	.062
Mo^{100}	.23	.053
Ru^{104}	.33	.018
Te^{126}	.85	.001
Te^{128}	.148	.005
Te^{130}	.250	.018
Ba^{138}	.56	.066
	Total	.327

Cross section: 10^{0} - 10^{1} barns

Element	σ	Yield
Se^{80}	1.1	.0009
Br^{81}	1.67	.0012
Zr^{94}	1.07	.066
Cb^{93}	1.2	.062
Ru^{102}	1.2	.039
I^{127}	6.8	.0012
Ba^{137}	8.8	.065
La^{139}	8.4	.065
	Total	.300

Cross section: 10^{1} - 10^{2} barns

Element	σ	Yield
Br^{79}	12.5	.0004
Mo^{97}	28.8	.065
Ru^{101}	27.2	.043
Pd^{105}	16.3	.009
Pd^{108}	12.1	.0005
In^{113}	61.	.00002

(continued on next page)

Cross section: 10^{2} - 10^{3} barns

Element	σ	Yield
Se^{82}	128.	.0025
Kr^{86}	114.	.022
Ag^{108}	108.	.0001
Xe^{136}	279.	.062
Nd^{145}	760.	.050
Eu^{153}	768.	.0012
	Total	.138

Cross section: 10^1 - 10^2 barns (cont.)		
Element	σ	Yield
Te^{125}	48.	.00025
Cs^{133}	25.6	.043
Ce^{142}	30.	.060
	Total	.221

Cross Section: 10^4 - 10^5 barns		
Element	σ	Yield
Cd^{111}	23000	.00006
Sm^{149}	40000	.015
Gd^{155}	33300	.0004
Gd^{157}	100000	.00006
	Total	.0155

Cross section: 10^3 - 10^4 barns		
Eu^{151}	1520	.004

Radioactive nuclei added to stable class and
assigned cross sections on statistical basis

Element	Half Life	Yield
Sr^{90}	30 y	.05
43^{99}	long	.058
Cs^{137}	25 y	.043
Ce^{144}	300 d	.055

APPENDIX II

Class Ib. Partially Shielded Stable Isotopes

Stable Element	σ_1	Yield	Parent	Assumed σ of Parent	Half-Life of Parent
Y^{89}	1.1	.046	Sr^{89}	316	55 da.
Zr^{91}	24.3	.058	Y^{91}	316	57 da.
Rh^{103}	163	.047	Ru^{103}	316	42 da.
Pr^{141}	11.0	.053	Ce^{141}	316	28 da.

APPENDIX III

Radioactive Isotopes with High Yields

Class IIa. High Yield Radioactive Nuclei with Odd Neutrons and Long Half-Lives

Element	Half-Life	Yield
Sr^{89}	55	.046
$Zr^{93,95}$	65	.045
Ru^{103}	42	.047
Ce^{141}	28	.050

av. = 45 days

Class IIb. High Yield Radioactive Nuclei with Odd Neutrons and Short Half-Lives

Element	Half-Life	Yield
Mo^{99}	2.8	.058
Xe^{133}	5.3	.038
Ce^{143}	1.4	.054
La^{140}	1.7	.061

av. = 2.8 days

[redacted]

Class III. High Yield Radioactive Nuclei with
Even Neutrons and Intermediate Half-Lives

Element	Half-Life	Yield
Y^{91}	57	.059
I^{131}	8	.017
Te^{132}	3.2	.036
Ba^{140}	12.5	.061
Pr^{143}	13.5	.054

av. = 18.8 days

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FIG. I - C AS A FUNCTION OF $\alpha \bar{t}$ FOR MINIMUM LOSS

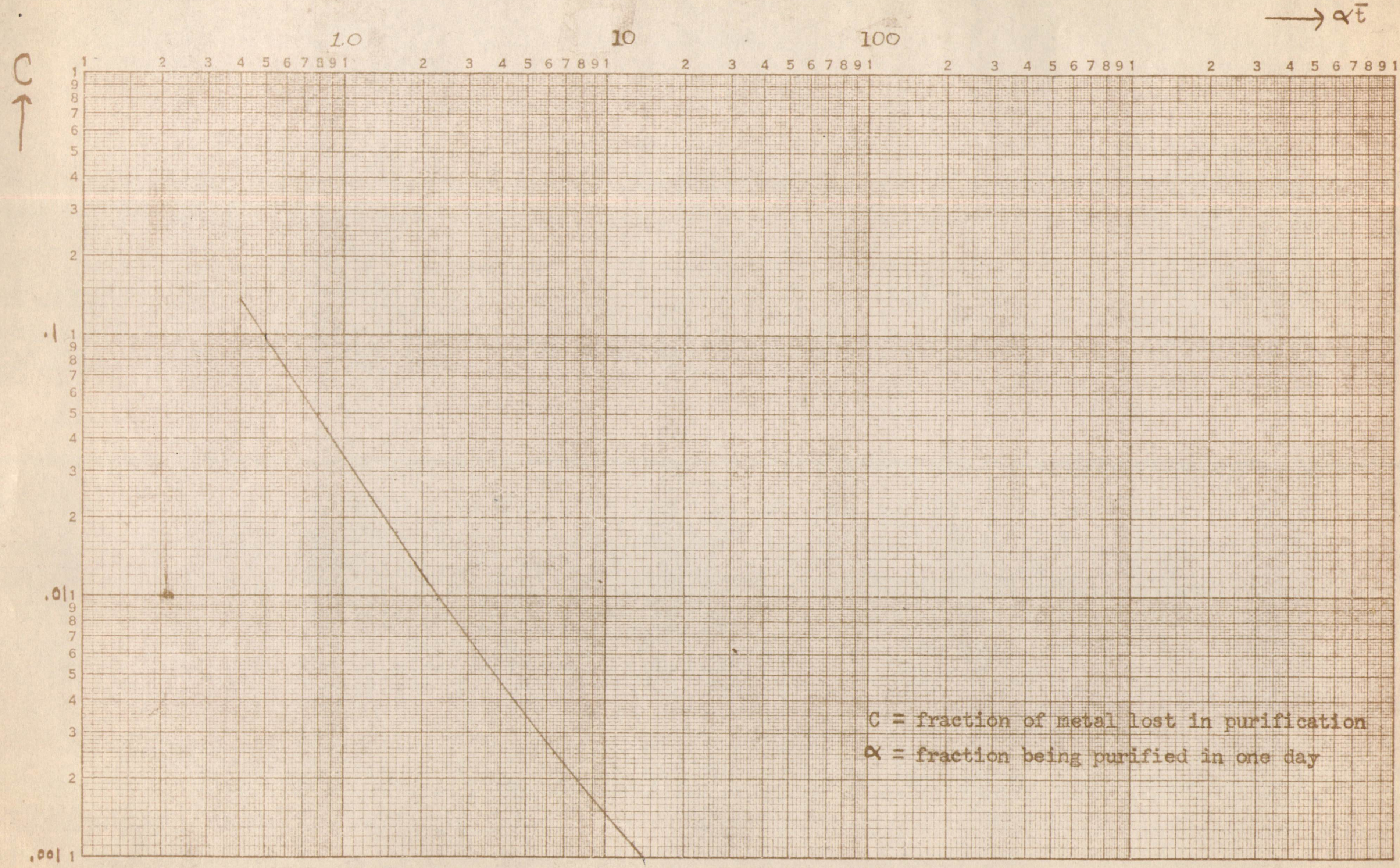
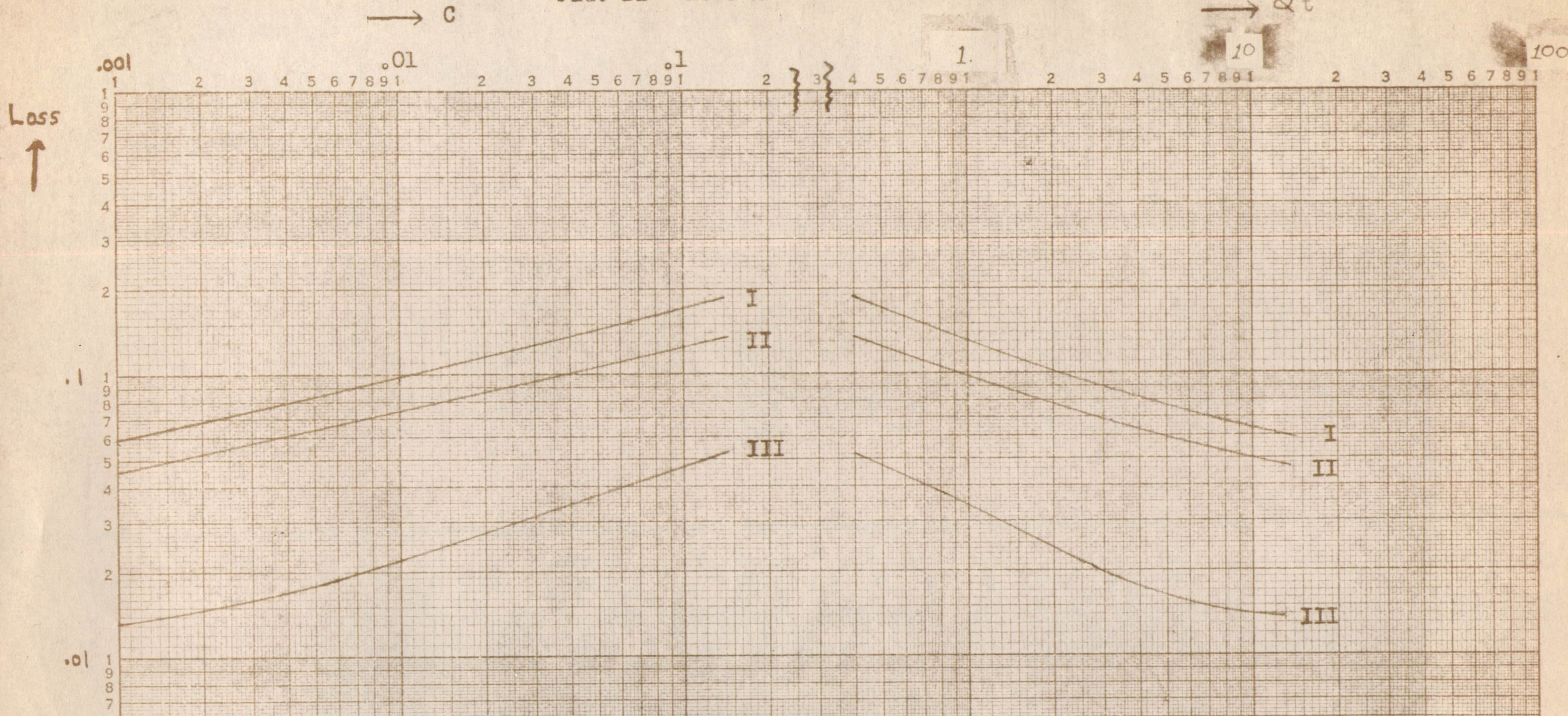


FIG. II - LOSS AS A FUNCTION OF C AND $\alpha \bar{t}$



$$\text{Loss} = \frac{\text{fissionable atoms lost in purification} + \text{neutrons absorbed by poisons}}{\text{neutrons absorbed in metal}}$$

$$= \text{purification loss} + \text{poisoning loss.}$$

C = fraction of metal lost in purification.

α = fraction being purified in one day.

I. Total loss.

II. Poisoning loss.

III. Purification loss.