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By Wigner, Weinberg and Young

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Weinberg

PRELIMINARY CALCULATIONS ON A BREEDER WITH CIRCULATING URANIUM

E. P. Wigner, A. M. Weinberg and G. Young

(S-17-45)

The following constitutes our present ideas on the breeder which, no need to say, are very preliminary and which we hope will undergo considerable changes before they will crystallize. Under ordinary conditions we would not report on work which is still as much in flux as the present one is. However, we had to shift our attention, at least temporarily, to another subject and we want to record for this reason the results obtained so far.

A. General Considerations.--The breeder is an arrangement in which 23 undergoes fission and the neutrons produced thereby are absorbed by thorium. When this thorium decays it produces again 23 via Pa. Since the η of 23 is, according to Anderson and May, 2.37 the net increase in the number of neutrons per 23 destroyed is 1.37. If all these neutrons were absorbed in thorium, we would obtain 1.37 23 atoms per 23 destroyed. It is well to remember, however, that if only as many as 15% of the 2.37 neutrons emitted are lost by escape from the system or parasitic absorption by other materials or otherwise, the efficiency of the breeder goes down to 1 and no increase in the amount of 23 results.

Another point that is important is the power to product ratio of such a breeder. Let us assume that the total losses in neutrons amount to only 7½%. In this case 1.19 atoms of 23 are produced per 23 atom destroyed. Thus the net gain in 23 is .19 atoms per 23 atoms destroyed or per .89 atoms 23 undergoing fission. As a result, the power output per net production is

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about five times greater than in the conventional piles.

During wartime it might possibly be more important to operate the systems to be described as converters of 49 to 23. In this case the above considerations do not hold necessarily and the efficiency does not have the paramount importance which it will be given in the following pages. If we disregard this fact, the main importance of the breeders is that they eventually permit the utilization of all the natural thorium. If one wants to make use of the full amount of energy one will have to construct a system in which the power generated by the breeder is utilized, i.e., in which the breeder acts not only as a producer of 23 but also as a power source. This will demand high temperature operation of the breeder. If only the energy of the 23 produced by the breeder is utilized, the energy of at least 1/1.19 = 84% of the Th is wasted if we adopt the numbers of the preceding example.

An enumeration of the losses follows:

1. Delayed neutrons which may be released outside the pile.
2. Absorption by materials present in the chain reacting part, such as the moderator and anion in case of solution, Bi and tubing in case of a second system to be described below.
3. Absorption by fission and corrosion products in pile.
4. Absorption by tank.
5. Escape in wrong direction. ends
6. Absorption by 24 formed.
7. Absorption by Pa.
8. Absorption by fission and corrosion products in reflector.

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9. Absorption by moderator and coolant in reflector.

10. Escape from reflectors, particularly of neutrons formed by fission in reflector.

11. Losses in chemical separations.

It may be well to introduce, at this point, the notion of the efficiency of the breeder. This is the net gain in the number of 23 atoms per 23 atoms destroyed. In the ideal case, it is 37%, in the example discussed above only 19%.

It will be seen in the next section that the arrangements which we think are most promising consist of essentially two parts: a chain reacting unit containing the 23 (the "pile") and a reflector containing all or most of the thorium. If all the thorium is contained in the reflector only 1/2.37 = 42% of the neutrons will be absorbed in the pile and 58% will be absorbed in the reflector. Hence a relative neutron loss ℓ inside the pile entails the loss of the fraction $.42\ell$ of all the neutrons. A relative neutron loss ℓ in the reflector gives a loss of the fraction $.58\ell$ of all the neutrons. The loss in "efficiency" caused by some impurity which absorbs in the pile .01 times as many neutrons as the 23 is 1%, the loss in efficiency caused by an impurity which absorbs in the reflector 1% of all the escape neutrons is 1.37%. A loss of 1% of all the neutrons entails a loss of 2.37% in the efficiency. All losses must be subtracted from the original 37% efficiency.

The present report assumes that the active material in the pile is 23 because only this is known to have an η considerably in excess of 2. It is worth noting, however, that 49 is an equally or even more suitable material

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for the same purpose if 40 is slow neutron fissionable. According to Miss K. Way's calculations, this should be the case. Assuming that the properties of 40 are the same as those of 49--which is perhaps a somewhat optimistic assumption--the number of fissionable atoms formed per 49 consumed is $\eta + (\alpha - 1)/\alpha = 2.05 + .33 = 2.38$, i.e., just the same as for 23. In spite of this, under the above assumption, 49 would be a somewhat more suitable material for the breeder than 23 because of its larger total cross section which would reduce the amounts needed and the concentrations necessary by the ratio of the cross sections, i.e. 630/1050 = .6. Furthermore, a loss of p percent of all the neutrons would reduce the efficiency in the case of Pu only to $\eta(1 - p) - 1 - 1/\alpha = .38 - 2.05p$ while the same loss would give, in the case of 23, an efficiency of $\eta(1 - p) - 2 = .37 - 2.37p$. This calculation assumes that 24 is not slow neutron fissionable. The possibility of using 49 is presented also because there may be a certain experimental error in the η of 23, as determined by Anderson and May.

B. Brief Description of the Arrangements Considered.--The general arrangement which at present appears most attractive consists of a relatively small pile proper which contains heavy water as moderator and all the 23 that is added to the system. The heavy water and the 23 are circulated to the outside of the system, cooled, stripped of the fission and decomposition gases and freed of the fission products outside. The chain reacting part is surrounded by the reflector. This also consists essentially of heavy water and contains most or all of the thorium. To give an idea we include a sketch at this point, although the detailed calculations will follow later on. The chain reacting unit has a volume of about 400 liters and contains of the order of 3 kg of 23. The thickness of the reflector is about 50 cm and

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contains about equal weights of thorium and heavy water. The pile is to be operated at 100,000 to 200,000 kw.

The first possibility (Fig. 1) envisages the uranium dissolved in the heavy water in the form of a salt, such as the nitrate, sulphate or fluoride. This solution is pumped into the pile through the neck which is on the bottom side and leaves the pile through the neck on the top side. The circulation rate is about 500 liters per second. The solution enters a heat exchanger and a degasser after it left the pile, whence it re-enters the pile again. The pile is surrounded by the thorium which is in the form of a slurry containing about one gram of ThO_2 per cm^3 of heavy water. If the wall of the tank separating the uranium solution from the thorium slurry is of a low absorbing material such as Be, Pb, Bi, or perhaps Cb, the neutron absorption of the tank may be sufficiently low. If, however, the tank has to be made out of a more absorbing material it may be necessary to back this up with a higher concentration of thorium, possibly in the form of small spheres ("beebees"), in order to reduce the thermal neutron intensity in that region.

The second arrangement shown (Fig. 2) contains the uranium dissolved in Bi, about 1 gm to 25 cm^3. This bismuth is circulated in suitable tubes and is cooled and freed of fission gases outside the pile. The shape of the pile is so chosen that the neutron escape at the ends shall be about 1% of the total neutron escape because this part of the neutrons will be lost. It may be possible to achieve the same purpose by another shape of the pile more similar to that in Fig. 1. This would involve, however, a more complicated inner structure of the pile. The tubes containing the bismuth are surrounded by heavy water which also has to be re-circulated, degassed and cooled outside

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the pile.

The interest in UF_6 as a pile material has been raised on this project by H. Brown and we are indebted to him for a discussion on both this and other problems. UF_6 could be used instead of metallic U in the pile of Fig. 2. The Bi would be replaced in this case by a fluorocarbon.

The first arrangement is much simpler than the second one and has a very much greater production rate, but the power which it furnishes cannot be well utilized because the temperature at which the pile can be operated is quite restricted. The greatest unsolved problem is that of the degassing of the heavy water without increasing the holdup outside the pile unduly. The second system is much more complicated and involves many serious problems among which we believe that a suitable choice of the pipes and the separation of the fission products from the Bi solution is most serious. Its production rate is much lower than that of the first pile but it furnishes the energy at a sufficiently high temperature for purposes of power production. If UF_6 is used in a fluorocarbon instead of U in Bi, the complications are much decreased but so would be the temperature of operation. The problems of chemical separation would be greatly alleviated but one would have to cope with the effects of radiation on the fluorocarbons. According to arguments presented by Burton and Franck, one may expect a somewhat more favorable situation in case of unsaturated ringlike structures, if these are sufficiently stable chemically, than in case of saturated fluorocarbons.

C. Reasons for the Arrangements Discussed.--The piles as described above assume pumping of the uranium solution by some suitable pump. Instead of this the solution can be pushed alternately into and out of the pile and

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cooled and degassed while it is outside (pulsating pile).

The most definite reason for using the 23 in solution is the need for eliminating the Xe^{135}. If we assume that altogether 5 kg of 23 are in the system and that it operates at 100,000 kw, 100 gm of 23 undergoes fission every day, the amount of Xe produced per day is 3.3 gm. If this were permitted to remain in the system as long as one day its cross section would be five times greater than that of all 23 present. In order to reduce this cross section to 1% of the cross section of the 23, one has to eliminate the Xe formed every 5 minutes. This is possible only if the Xe can be flushed out of the system at frequent intervals and eliminates the possibility of using the 23 in a solid form. If the Xe is flushed out at very frequent intervals one gains an additional substantial elimination of other fission products which might otherwise poison the pile.

The above conclusion that the 23 must be used in a non-solid form is not absolute. The maximum loss of efficiency if all the Xe^{135} is left in the pile is 5.8% and although the permissible loss must be well below 37% a loss of 5.8% could be tolerated if all the other losses can be kept at a minimum. However, circulating the 23 has many other advantages, particularly from the point of view of heat transfer and it is not evident that using it in a solid form will facilitate the elimination of the other losses. We feel, therefore, that the conclusion to use the 23 in a non-solid form is a valid one. The only other possibility to eliminate the Xe^{135} loss which we realized is to operate in such a way that practically all neutrons be absorbed at higher energies (by resonance absorption) before they reach such energies at which the Xe^{135} cross section is large.

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The Xe^{135} could be removed from the 23 inside the pile by letting a gas bubble through the liquid which contains the 23. This might be a practical possibility if the 23 is dissolved in Bi. We felt, however, that such an arrangement would introduce unnecessary complications and since it has great advantages to remove the 23 from the pile periodically for cooling and for separating the other fission products, we decided against it. This fixed one of the most important properties of the piles which were described above, that of circulating the 23 to the outside of the pile. Such a circulation of course increases the amount of 23 necessary for the pile but greatly facilitates the energy transfer from the 23 to the outside.

It will be seen that considerable difficulty is caused by the need of inserting a tank between pile proper and reflector because the tank is likely to absorb neutrons. It would be advantageous, therefore to place the thorium, or at least most of it, to the inside of the pile. This would result in the use of a synthetic "natural uranium" which would give a pile with a very small multiplication factor, an arrangement which would greatly help in decreasing the neutron losses. There is no question that there is great merit in this point of view. If we reduce the multiplication constant (which is nearly 2.37 in the pile as described above) to 1.05, only 5% of all the neutrons escape from the pile proper and only these have a chance of being absorbed by the tank. However, the diameter of the pile will be increased in this way to about 3 m

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and its volume to 14 m^3. The amount of 23 required for the pile proper would be of the order of 70 kg under these conditions. In order to keep the amount of 23 needed for the pile at a relatively low level, we have decided therefore to separate the chain reacting part from the reflector where the production of the 23 takes place. It might be advisable to add some thorium to the pile proper but this should be kept at such a level that the multiplication constant does not decrease below about 1.5. If this is done the absorption by the thorium within the pile proper will be about 60% of the absorption of the 23 and its amount in the pile will be about 60 times the amount of the 23. Such an arrangement has the advantage that the number of neutrons which have to leave the pile is reduced by a factor of almost 3 and the losses in the tank and outside will be diminished by the same factor. If the pile proper contains the 23 in a solution in heavy water we have decided against this possibility because the thorium could be present only in the form of a slurry which would aggravate the pumping problems and the problems of chemical separation.

The next question is the question of the moderator. In this respect we believe that the choice of heavy water is rather unique. If we want the number of neutrons absorbed in the moderator to be only 1% of the neutrons absorbed by the 23, the volume g of the different moderators associated with 1 gm of 23 is given in the following table:

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TABLE I

	σ_a/molecule	Density	Volume associated with 1 gm to give loss of 1% (g)	Heat capacity of this volume in joule/°C (C_s)	Age
C	.0045	1.6	44.5 cm^3	65	320 cm^2
D_2O	.004	1.1	120 cm^3	500	115 cm^2
$D_2O + \frac{1}{2}\% H_2O$	.007	1.1	70 cm^3	290	110 cm^2
H_2O	.6	1	.8 cm^3	3.3	33 cm^2
Be	.01	1.8	13.3 cm^3	42.5	200 cm^2
Bi	.017	9.8	33 cm^3	48	---
$(CF_2)_n$	.023 n	1.8	32 cm^3	~70	

Among these C is undesirable because the radius of the pile would amount to about 60 cm, assuming a multiplication constant of 2.37 and a migration area corresponding to the age alone (i.e. assuming zero diffusion length for the thermals). Such a pile would contain far too much 23. Water is undesirable because the size of the pile would become too small so that one would run into heat extraction difficulties. If the 23 is in the pile as short a time as 1/10 sec and if one wants a power output of 100,000 kw for 5 kg 23, the temperature increase is the water would amount to 20,000 x 1/10/3.3= 600° C. Be in itself is not much more suitable than graphite and Bi cannot be seriously considered as a moderator. This leaves D_2O as a rather unique choice. On the other hand, it is possible for the pile to contain substantial amounts of Bi and Be, in addition to the D_2O which has to do the bulk of the moderation.

Heavy water as a moderator gives considerable leeway for the concen-

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tration in which 23 can be used. The importance of this is the following: If we use a high concentration of 23 only a relatively small fraction of all the neutrons will reach thermal energies and many of them will be absorbed at resonance above thermal energies. There is not much reason to believe that the η is absolutely constant even in the very small energy region which extends from ordinary thermal energies to about 1 volt and if it should turn out that η increases with increasing energy it may constitute a considerable advantage to go to high concentrations and high resonance absorption. On the other hand, if it should turn out that η decreases with increasing energy of the neutrons, it would be best to keep the concentration of 23 in heavy water as low as possible. This question will be taken up again in section D.

It is well known that if the uranium is dissolved in water the fission causes a very large amount of disintegration. We estimate from data of Burton and Allen (Γ = .25) that such a pile operating at 100,000 kw would give 220 liters NTP hydrogen oxygen mixture per second. While the gases have the desirable effect that they carry with themselves the Xe and some other fission products when they leave the liquid, the disengagement of such amounts of gas causes considerable trouble even if it is done at relatively high pressures. There is considerable advantage therefore to have the 23 separated from the heavy water which can be done most simply by dissolving it in Bi and keeping the Bi separated from the heavy water by tubes of a non-absorbing material or thorium. This leads us to the two systems which we have considered most seriously and which will be described more closely in the next section.

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There is one other remark that ought to be made in this connection. At the concentrations of 23 which reduce the loss due to absorption by the moderator to the order of 1% a considerable number of neutrons will be absorbed before they reach thermal energies. If both the moderator and the 23 obeyed the 1/v law this would not change the ratio of the neutrons absorbed by the moderator. There is considerable indication, however, that the absorption of the 23 at resonance energies is considerably greater than would correspond to the tail of the 1/v absorption extrapolated from thermal energies. Mr. Dancoff estimates on the basis of experiments carried out in the Argonne Laboratory that the $\int \sigma_a dE/E$ is at least twice greater than it would be if 23 followed the 1/v law. As a result, the fraction of neutrons which are absorbed by the moderator will be about half as great for the neutrons absorbed at resonance than for the neutrons absorbed at thermal energies. Another effect which this resonance absorption has comes in in the calculation of the Laplacian which is increased by this effect. Since the ratio of the actual $\int \sigma dE/E$ to the value which is obtained by extrapolating the 1/v law is not known the calculations will be carried out under three different assumptions, viz., that that ratio is $\alpha = 1, 2, 4$.

C. Critical Size.--The critical sizes for pure fissionable material have been calculated by Christy and Wheeler (CP-499), by Weinberg, and more recently these calculations have been extended by Nordheim.

It turns out that the two group theory and the three group theory both give results which are substantially below those given by Fermi's theory. The results of the three group theory are just about midway between those of the two group theory and the Fermi theory. Plass calculated the corrections to the Fermi theory and in the case of heavy water they turned out to be quite

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negligible, as was already indicated by Weinberg's work.

Our calculation includes the effect of the resonance absorption for which the $\int \sigma dE/E$ was estimated in the following way. The value of σ_a was taken to be $(98\,\alpha)/\sqrt{E}$ which gives for $\alpha = 1$ the correct value for thermal energy. However, three values of α were used, viz., $\alpha = 1, 2, 4$, as explained before. The integration was extended from 2kT to ∞ giving 900α. Evidently these assumptions are quite arbitrary but should give the right order of magnitude. The average energy of the neutrons in the pile will no doubt be considerably higher than .05 ev, but the slowing down just above the thermal region will be considerably slower than the formula for the resonance absorption implies. These two effects give corrections of opposite sign. The following table gives the probability for escaping resonance for g = 100, 200, 300, 400 cm^3 heavy water per gram 23.

TABLE II

vol H2O/gm 23

Number of Neutrons Reaching Thermal Energy

g	$\alpha = 1$	$\alpha = 2$	$\alpha = 4$
100	.875	.769	.592
200	.936	.875	.769
300	.957	.916	.840
400	.968	.936	.875

The Laplacians L were then calculated by Mr. Wilkins on the basis of the formula developed by Messrs. Friedman and Weinberg

$$k_s \frac{\varkappa_t^2}{\varkappa_t^2 - L} + k_f = e^{-L\tau}$$

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and his calculation will be given in more detail elsewhere. In this, $k_s + k_f = k$ is the multiplication constant; k_s is the number of neutrons produced per slow neutron absorbed, multiplied with the fraction p of neutrons which are thermal when absorbed; k_f is the number of neutrons produced per fast neutron absorbed, multiplied with the probability 1 - p of resonance absorption. In our case $k_s = pf\eta_s \approx p\eta$; $k_f = (1 - p)\eta_f$. The results with Fermi's theory are given in Table III.

TABLE III

	$= 10^3 L$ cm^2			R in cm			Volume in liters			23 in kgr.		
g	$\alpha = 1$	2	4	1	2	4	1	2	4	1	2	4
100	5.63	5.83	6.19	41.8	41.1	39.9	162	152	136	1.62	1.52	1.36
200	4.53	4.66	4.95	46.6	45.9	44.6	241	228	216	1.20	1.14	1.08
300	3.79	3.90	4.12	50.9	50.3	48.9	332	317	291	1.11	1.06	.97
400	3.30	3.39	3.57	54.8	54.1	52.6	430	411	372	1.07	1.03	.93

It was assumed in the calculation that the age τ of fission neutrons in heavy water is 115 cm^2, the diffusion mean free path 2.4 cm. This gave 50, 100, 150 and 200 cm^2 for the square of the thermal diffusion length $1/\varkappa_t^2$ for the four different g. The radii R of a bare critical sphere are also given in Table III. According to calculations of Mrs. Monk and Mr. Friedman, which also will be reported elsewhere, it seems reasonable to subtract 8 cm from the radius in case of a ThO_2 slurry reflector. This gives the amounts of 23 necessary for the pile which are given in Table III in kg.

The advantage of using a high concentration lies in the smaller

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losses of neutrons to the moderator, etc. On the other hand, a low concentration of 23 would give increased bulk to the system and thus facilitates a higher production rate. If α should be large there is an increased advantage in using high concentrations because the neutron losses to the moderator are further reduced by the resonance absorption. It is seen from Table III that the holdup of 23 within the pile is quite small. There is of course a holdup outside the pile.

If it should be necessary to use the cylindrical arrangement given in Fig. 2, the height of the cylinder will have to be chosen in such a way that the escape toward the two ends of the cylinder be about 1%. This gives the following equation for the height H of the cylinder in terms of its radius R

$$\frac{\pi^2}{H^2} = \frac{1}{100}\left(\frac{2.405}{R}\right)^2$$

Calculating the volume from this equation it appears that it is 4.3 times greater than the volume of the sphere considered in Table III. This would necessitate a four times greater holdup in 23 which can be reduced, however, by reducing the diameter of the tubes containing the 23 solution toward the two ends of the pile.

The lattice spacing is not a critical quantity in the second arrangement. Of course, the total amounts of heavy water, beryllium, and solvent must be kept in such limits as can be obtained from Table I. In addition, one will not want the neutron density in the moderator to exceed substantially the neutron density in the solution. If $\mathcal{K} r_o < 1$, the neutron density at the tube will be less than 10% higher than the average neutron density in

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the tube. Since $\chi = .2$ for both the Bi and the fluorocarbon solution of Table I, this only gives $r_o < 5$ cm. The ratio of the neutron density difference between surface of cell and surface of tube, divided by the density at the tube is

$$\frac{N\sigma_{ao}r_o^2}{2D}\left(\frac{r_s^2}{r_s^2 - r_o^2}\ln\frac{r_s}{r_o} - \frac{1}{2}\right)$$

In this $N\sigma_{ao} = .048\ \mathrm{cm}^{-1}$ for the solutions of Table I, $D = .8$ cm is one third of the diffusion mean free path, r_s is the cell radius. The above ratio will be under the conditions of Table I, smaller than .1 if $r_o < 3.5$ cm.

D. Holdup Outside the Pile.--The holdup outside the pile depends to a large extent on the diameter of the tubes in which the solution passes through the heat exchanger, the temperature difference between coolant and the temperature of the 23 solution, holdup in the pumps and, particularly in case of the arrangement of Fig. 1, on the time necessary for degassing. No calculations are available on these points at present but one can estimate that the solution will spend about 2 sec outside the pile before it re-enters it. Under this assumption the power output depends solely on the holdup and the temperature difference by which the solution is cooled down. It is given by

$$P = \frac{C_s T}{t}\ \frac{\mathrm{kw}}{\mathrm{kgr\ holdup}}$$

In this C_s is the heat capacity of the solvent per gram of 23 dissolved in it (cf. Table I), t is the time of holdup outside the pile, and T the temperature difference between the solution as it leaves and re-enters the pile.

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In case of pile of Fig. 1, t = 2 sec, T = 50° C, g = 100, this gives P = 10,000 kw/kg, g = 200, P = 20,000 kw/kg.

One may consider flashing the liquid in order to achieve some cooling by evaporation as well as elimination of the fission gases. Messrs. Newel and Ginns proposed in some other connection to let the solution flow down on the inside of a cooled tube in a thin sheet and one could maintain sufficiently low pressure inside the tubes to permit evaporation and release of the bulk of the decomposition gases. These would carry the Xe and some other fission products with themselves and also a considerable amount of heat in the form of heat of vaporization. The advantage of this arrangement is that it unites the elimination of the decomposition gases with some cooling and also that the pressure drop in such a system is quite small. Considerations have not progressed sufficiently to know how large the saving in the holdup would be if this arrangement is used.

A possibility which might be applied to the arrangement of Fig. 2 is that of pulsating it. This avoids the holdup in the pump and also the need of pumping the radioactive solution. Figures for the theoretical efficiency of this cooling are given in Table IV in kw per gm holdup outside the pile. It was assumed that the temperature difference between coolant and 23 solution of the pile is 50° and that the temperature difference between the coolant and the tube of the heat exchanger is made negligible by vigorous circulation of the coolant and finning of the heat exchanger tubes. The pressure difference between the active pile and the end of the tubes alternates between .6 and -.6 atmospheres in such a way as to give a velocity of - 700 cm/sec in the tubes. The holdup in the degasser is not taken into account in the figures in Table IV.

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TABLE IV

Radius of tubes	Length of tubes	Length of cycle	kw/gr g = 100	kw/gr g = 200
1 cm	100 cm	.45 sec	37.5 (26)	75 (52)
1 cm	200 cm	.90 sec	31 (25)	62 (50)
.5 cm	50 cm	.23	82 (44)	164 (88)
.5 cm	100 cm	.45	65.5 (44)	131 (88)
.5 cm	200 cm	.90	45 (37)	90 (74)

The figures given for the power assume that the pressure adjustment at the end of the tubes can be carried out instantaneously. The bracketed figures assume an adjustment time of .1 sec. It was further assumed that the tubes can be entirely emptied on the "in" cycle. Assuming that the meniscus has to stay at least 10 cm away from the pile tank further reduces the power holdup by 20% to 5% for tube lengths of 50 cm to 200 cm. The advantage of longer tubes is, obviously, that they permit a greater total production, although with a somewhat decreasing efficiency. The advantage of wide tubes is the same and a greater mechanical simplicity.

E. Estimate of the Losses.--It will be seen that, on the whole, the losses from the different sources enumerated under A become smaller as the production rate of the pile is diminished. It appears quite natural that the losses in a pile with a high power output are greater than if all attention can be given to a greater efficiency. We proceed with the discussion of the eleven separate points given under A.

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(1) The number of delayed neutrons is about .8% in 25. Although its amount is not known for 23, Anderson found indications that it is greater for this element. As pointed out under section A, a loss of 1% of neutrons decreases the efficiency by 2.37%. Since all the delayed periods, except perhaps the first one, are long compared with the time of uninterrupted sojourn of the 23 within the pile, the number of delayed neutrons emitted outside the pile will be equal to the holdup outside the pile divided by the total amount of 23 present. From this point of view it is indeed advantageous to have a large pile and a very small holdup outside.

The flash cooling system may form an exception under this rule because it may be possible to arrange it in such a way that most of the neutrons still would be utilized. Similarly it may be possible to achieve this at least partially in the pulsating system. In spite of this there will be a loss of efficiency from delayed neutrons in every system in which the uranium is circulated at the outside. This constitutes a rather serious drawback of these systems since the loss in efficiency from this source is not very much smaller than that caused by the Xe^{135} would be.

(2) The absorption by the moderator has been discussed in detail before. It will amount to about 1% for the pile of Fig. 1 and one will be quite lucky if it is as little as 2% in the second case. However, in the first case the absorption by the anion also has to be taken into consideration. If the uranium is used in the form of a nitrate the nitrogen of the nitrate alone will give a loss of $\frac{1}{2}$%. In addition to this it appears likely that further nitric acid has to be added to the system in order to keep the uranium in solution in spite of the presence of the hydrogen peroxide. The situation would be very much better if N^{15} were available for use both in

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the nitrate and the nitric acid.

The reason that we gave the nitrate precedence over other salts is that we expect it to reduce the corrosion of the walls and also because it does not give products under radiation which may gum the system. The sulfate which has negligible absorption may precipitate sulphur although it is not clear that this cannot be avoided by the addition of peroxide. The parasitic absorption of the fluoride, the use of which was proposed by Allen, is also negligible but there is a strong opinion (Hiskey) that it will be difficult to avoid precipitation of the peroxide at such concentrations of the hydrofluoric acid which are tolerable from the point of view of corrosion. If the uranium is used in the form of a solution in Bi or fluorocarbons it may be necessary to eliminate the products of nuclear reactions of these substances along with the fission products.

(3) The estimate of the poison by the fission products is probably the most difficult at present. According to Miss Way only the Sm is a serious poison among the stable fission products. Inasmuch as the yield of this is probably 1.6% its elimination is very important and may demand an average interval of one day between purifications. In order that this be permissible, it is necessary that the purification be carried out with a loss of about .1% of the 23.

The above applies to the stable fission products. There may be, however, numerous radioactive fission products (in addition to the Xe^{135}) which have large cross sections. Their effect has been estimated by Miss Way on the basis of a statistics of the cross sections and she finds that those with an odd number of neutrons are most dangerous. All elements may give substantial contributions, the lifetime of which is an hour or more.

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Only those with a lifetime of more than a day have been surveyed to date and Miss Way estimates that they may cause a loss in efficiency of $3\frac{1}{2}\%$ if the purification is carried out once a day. If this is done it is unlikely that the corrosion products will contribute much to the poisoning.

Probably the most efficient way of eliminating the fission products is by the solvent extraction method. We are much indebted to Mr. Tepe for discussing this system with us. The solvent extraction method would permit a continuous operation and cause a holdup of only about one hour in the column. As a result it would appear that such a purification increases the amount of 23 required by only about 4%. However, it is likely that it will be impossible to use the solution of uranium in heavy water directly in the column because the deuterium will exchange with the hydrogen of the solvent. For this reason it will be necessary to separate first the uranium salt from the heavy water which may be a more time consuming operation than the operation of the column itself and consequently increase the holdup more than the fission product separation itself does.

If the uranium is used in a solution in Bi the problem of chemical separation is even graver. Although many methods were discussed no really promising one turned up so far. One must remember that after the chemical separation has been carried out the uranium must presumably be reduced again to the metallic form. It is unlikely that this operation can be carried out with as small a loss as would be necessary for the successful operation of the breeder. This is the most serious drawback of the Bi system that has turned up yet. On the other hand, if the uranium is dissolved as a hexafluoride in fluorocarbons the problem of separation may be much smaller.

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As pointed out by Brown and Anderson, distillation may lead to the goal and the extraction column also could be used because reconversion to the fluoride can probably be handled with relatively great efficiency.

The Xe^{135} occupies a particular position. Let us first consider the case in which the uranium is dissolved in heavy water. In this case there is a considerable amount of water decomposition and the hydrogen and oxygen generated must be eliminated from the system every second. Even so, the equilibrium pressure of H_2 and O_2 will amount to a dozen atmospheres. During the degassing the hydrogen and oxygen will leave the water either in form of bubbles or through the surface. In both cases the fraction of Xe eliminated will be just about as great as the fraction of hydrogen and oxygen eliminated because the solubility of Xe is only insignificantly higher than the solubility of hydrogen and oxygen and because the diffusion of Xe to the surfaces will not be much slower than the diffusion of the oxygen. As a result, the amount of Xe will be reduced by a factor of the order of $\frac{1}{2}$ every second and the amount present will be that generated during a few seconds only. Since it is necessary to eliminate the Xe only about every couple of minutes, it is evident that the need of elimination of the hydrogen and oxygen will cause in this system an about 100 times better elimination of the Xe than is necessary. Even if it should prove feasible to reduce the gas evolution to 1/10 or even 1/30 by introducing some suitable catalyzer into the system which recombines most of the hydrogen and oxygen in the water the elimination of the Xe would be quite satisfactory.

For a quantitative assessment of the problem let us assume that the mean life of the 23 in the pile is $\bar{t}$. This is the amount of 23 present,

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divided by the consumption rate and is for 100,000 kw power rate of the order of 6 kg/100 gr/day = 60 days. Assuming a fission yield of 5% for the Xe^{135}, the production rate of Xe^{135} will be .05 $N/\bar{t}$ where N is the number of 23 atoms. The total number of Xe^{135} atoms in the system will be .05 $Nt_e/\bar{t}$ where t_e is the average time after which the Xe is eliminated. The poisoning due to this amount is .05 $Nt_e \sigma_{Xe}/\bar{t}N\sigma_u$ and this will be less than 1% if

$$t_e < .2 \frac{\sigma_u}{\sigma_X} \bar{t} = 4 \times 10^{-5} \bar{t}$$

For $\bar{t}$ = 60 days this gives $t_e \approx 3$ min.

The situation is somewhat different if the uranium is dissolved in bismuth or a fluorocarbon. In the former case at least it will be necessary to sweep out the Xe by bubbling some gas through the bismuth periodically. This has to be done, as pointed out before, every five minutes at least and will cause some additional holdup, the magnitude of which will depend on the size of the bubbles sweeping through the liquid and the area over which the solution is spread out during this operation. It does not appear, however, that the increase in holdup necessitated by this operation will go beyond a few percent.

(4) The absorption by the tank is principally determined by the material of the tank. If this absorption is low it will be quite possible to have the slurry of thorium which is outside the tank in immediate contact with the tank. Mr. Friedman and Mrs. Monk have calculated this effect. In the region of interest, their results can be approximated by the formula

$$\frac{\sigma_{at}\, \ell_t}{\sigma_{as} \sqrt{\tau}}$$

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for the fraction of neutrons entering the reflector which are absorbed by the tank. In this, σ_{at} is the absorption cross section of the tank per cm^3, ℓ_t the thickness of the tank σ_{as} the absorption cross section of the slurry per cm^3, $\tau = 115\ cm^2$ is the "age" in the reflectors. We assume a slurry which contains 1 gr Th per cm^3 which gives $\sigma_{as} = .016$.

The loss of efficiency is obtained by multiplying the above expression by 1.37. It is only 1% for 1.5 mm thick Al tank ($\sigma_{at} = .014$) and correspondingly less for a tank out of lead or beryllium or for some backing of the aluminum made out of beryllium.

If the tank has to be made out of a strongly absorbing material, it will be necessary to back it up with a layer of thorium in a more highly concentrated form (cf. Fig. 3). Beebees of metallic thorium which are cooled by heavy water circulated through them may be a suitable form. This arrangement has two effects: first the thorium reduces the density of the thermal neutrons in the neighborhood of the tank to such a level that the ratio of the thermal neutrons which are absorbed by the tank to the number of thermal neutrons which leave the pile toward the reflector is given by the formula

$$\sqrt{\frac{3\sigma_b}{\sigma_{ab}}}\,\sigma_{at}\,\ell_t$$

In this σ_b is the scattering cross section in the backing per cm^3 of the backing, σ_{ab} is the absorption cross section of it, again per cm^3. One calculates that if the backing consists of 80% metallic Th and 20 volume percent of heavy water($\sigma_{ab} = .13$, $\sigma_b = .33$) the absorption of thermal neutrons by a $\frac{1}{2}$ mm steel tank ($\sigma_{at} = .22$) would give a loss in efficiency of about 1.5% if

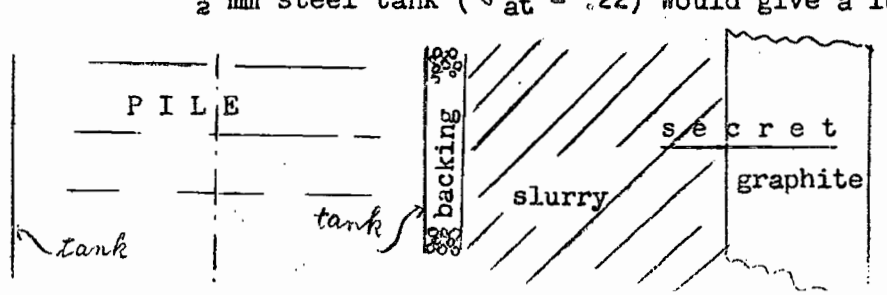

Fig. 3

one assumes that $\frac{1}{2}$ of all neutrons escape as thermal. A reinforcing of the tank by a few mm aluminum would hardly add anything to this. Behind the backing there must be of course further heavy water containing thorium to slow down and absorb the neutrons which leave the tank at higher energies. It is easy to see, along the same lines as used in the argument by Friedman and Monk, that the fraction

$$\frac{\varkappa_b \sigma_{at} \ell_t / \sqrt{\tau}}{\varkappa_b \sigma_{as} \mathrm{Ch}\, \varkappa_b \ell_b + \varkappa_s \sigma_{ab} \mathrm{Sh}\, \varkappa_b \ell_b}$$

of these neutrons will be absorbed by the tank. In this formula, as before, s and b refer to slurry and backing, σ_a, $\varkappa$ and ℓ are absorption cross section per cm^3, reciprocal diffusion length and thickness of the corresponding region. For the above backing, containing 80 volume percent Th, 20 D_2O, $\varkappa_b \sim .35$, and the above slurry, containing 1 gr/cm^3 Th $\varkappa_s \approx .14$. As a result, if the backing has a thickness of about 5 or 6 cm the absorption of these neutrons is also quite small ($< \frac{1}{2}\%$). Evidently the introduction of the backing will somewhat reduce the efficiency of the reflector. However, the 8 cm thickness which was used in the previous calculations already takes this effect into consideration. Another point that might be mentioned is that the resonance absorption given in Table II also will serve to reduce the losses due to the tank by the factors given in that Table.

(5) The shape of the tank as given in Fig. 1 or 2 is already designed to reduce the escape from the tank in the directions in which there is no reflector and it is sufficient to repeat that the extremely elongated shape given in Fig. 2 may not be necessary if the tubes toward the two ends of the

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tank are made thinner than at the center, thus reducing the multiplication constant in that region. This will, of course, cause a slight increase in the diameter of the tank but will still very substantially reduce the amount of uranium in the pile.

(6) The effect of the absorption by the 24 can be considered under the assumption that the 24 itself is not fissionable. In this case every atom 24 will eventually be transformed into 25 so that in equilibrium there appears to be a loss of 12% in efficiency due to the absorption by the 24 because in equilibrium, .12 as many neutrons are absorbed by 24 than by the 23. Of course if the cross section of the 24 is very small this equilibrium will be reached only after a very long time and until this happens the loss in efficiency is correspondingly smaller. However, even when equilibrium has been reached the loss in efficiency will be very much smaller than the 12% quoted because the absorption leads to a fissionable element 25 and the loss in efficiency is caused only by the fact that the 25 has a smaller η than the 23. When the absorption of 25 has come to an equilibrium one can consider the total effect in the following way. A 25 is formed by the absorption of two neutrons from the 24 and gives η_{25} new neutrons. If we denote, as in Section A, the fraction of neutrons which survive all losses by $1 - p$, the loss due to the absorption of 24 will be given at equilibrium by $2 \times .12 - .12\,\eta_{25}(1 - p) = .25\,p - .01$. One sees that this absorption will not cause a considerable decrease in efficiency. Strictly speaking, one should further consider the formation of 26 from the 25, but this evidently will not cause any major effect.

(7) The absorption by the Pa is a more serious effect. This element

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is, according to Miss Way, probably not fissionable and will therefore cause a loss in two ways. First of all, by the neutrons which it absorbs and second by forming an element of a relatively short half-life which decays into 24 instead of the fissionable 23. For this reason it will be necessary to keep the absorption by the Pa at a minimum. This can be done by extracting the Pa at sufficiently frequent intervals so as to reduce the absorption of neutrons by this element to about ½% of the absorption by the thorium. This will be relatively easy if the absorption cross section of Pa is of the same order as the absorption of the thorium and if all the thorium can be used in the form of a slurry, since for a power output of 100,000 kw only about 120 gms of Pa are formed per day and since the mean life of Pa is 40 days, there will be, in steady state, 4.8 kgr Pa in the reflector. Since the total amount of Th in the reflector is more than a ton, it will be sufficient to extract the Pa only n times a month if its cross section is n times that of the thorium. However, if it is necessary to use part of the thorium in form of a backing of the tank, a considerable fraction (about 25%) of all of the neutrons will be absorbed by the first few cm of this backing. As a result, the Pa accumulates in this part of the thorium much more rapidly than in the bulk of it and the purification would have to be undertaken more frequently. This is especially disconcerting because the removal and replacement of this backing is a somewhat awkward problem.

(8) Relatively frequent purification of the thorium seems also indicated by the accumulation of fission and corrosion products. The frequency of the need of this purification can be estimated in the following way: The

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absorption cross section of thorium is just about 100 times smaller than the absorption cross section of the 23. Therefore, if we divide the amount of thorium in the reflector by 100 times the amount of 23 in the pile, we obtain the number by which the cross section of the thorium in the reflector is larger than the cross section of 23 in the pile. This number is at least 4 to 5. On the other hand, the amount of fission in the reflector will be less than 1% of the amount of fission in the pile if all the thorium is distributed uniformly in the reflector. The relative poisoning of the reflector by the fission products will be about 400 times smaller than the poisoning of the pile and a correspondingly less frequent purification would be quite sufficient for the elimination of the fission products. It is seen from this that the need for purifying the thorium of the reflector will arise mainly from the absorption by the Pa, the absorption by the corrosion products, if any, and to reduce the holdup of 23 within the reflector. This last factor will demand a purification of at least once a month because after this time there will be about 3 kg of 23 in the reflector. Of course a more frequent purification may be demanded either because of the absorption of the Pa or the absorption by the corrosion products but we shall see that this does not seem to be the case.

The above discussion assumes that the thorium is uniformly distributed within the reflector but the situation is not very different if a backing of the tank is needed.

(9) As mentioned before, the absorption cross section of thorium is just about 100 times smaller than the absorption cross section of 23. Therefore, if we want to reduce the losses due to the moderator in the

reflector to 1% of all the neutrons absorbed in the reflector--this will give a loss of 1.37% in efficiency--the volumes associated with 1 gm of thorium are 100 times smaller than the figures given in the fourth column of Table I. It appears from this that only D_2O can serve as a carrier of the thorium in the reflector. Using the thorium in form of a slurry has the further advantage that a frequent mixing of the whole amount of thorium can be achieved and it will not be necessary to remove parts of the thorium at more frequent intervals.

(10) There are three causes for losses of neutrons from the reflector: The first one is the escape of neutrons toward the ends of the pile in case the arrangement of Fig. 2 is used. This, however, will be a very small quantity if the loss from the pile in the same directions is also very small. The escape of neutrons perpendicular to the main surface of the pile is governed by the tail of the slowing down density of neutrons. If one could adopt for this tail the Gaussian expression it would be sufficient to use such a thickness ℓ_s that $\ell_s^2/4\tau = 4$ which would give $\ell_s = 4\sqrt{\tau} = 45$ cm. Actually it may be safer to surround the reflector by a second reflector which may be out of graphite. In this case one can reduce the thickness of the reflector which contains the thorium to about 38 cm.

This figure is arrived at in the following way. The number of neutrons absorbed in the reflector divided by the number of neutrons entering the graphite reflector is approximately

$$\frac{4}{\sqrt{3}}\sqrt{\frac{\sigma_{ag}}{\sigma_g}}$$

In this, σ_{ag}/σ_g is the ratio of absorption and scattering cross section in C. This formula assumes that all the neutrons entering the graphite are thermal

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which is a slightly pessimistic assumption. The fraction of neutrons entering the graphite reflector is given by

$$\frac{3\sqrt{\tau}\,\sigma_s'}{2\sqrt{\pi}}\,e^{-\ell_s^2/4\tau} - \frac{3\,\ell_s\,\sigma_s'}{2\sqrt{\pi}}\int_{\ell_s/2\sqrt{\tau}}^{\infty} e^{-y^2}dy + \frac{e^{-\ell_s^2/4\tau}}{4\sqrt{\pi\tau}\;\;\sigma_{as}}$$

$\sigma_s' \approx .4\ \mathrm{cm}^{-1}$ is an average fast neutron cross section of the slurry per cm^3. The first two terms refer to the fast neutrons entering the graphite, the last term to the slow neutrons entering it. The length, 38 cm, is so chosen as to reduce the total number of neutrons escaping from the system to less than .5% of all neutrons entering the thorium reflector. The above calculation should be somewhat improved to take into account the neutrons generated by fission in thorium. However, the number of these neutrons is quite small, less than 1% of all the neutrons, and the probability of their escape remains small even if they are found somewhat to the inside of the thorium reflector.

(11) The losses in chemical separation should really be considered in conjunction with the poisoning problem. Evidently the smaller the losses the chemical separation involves, the more frequently it is worth while to carry out such a separation. The figures given under (3) were not calculated but estimated on the assumption that the loss of 23 during a chemical cycle amounts to .1%.

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The total loss in efficiency therefore can be estimated as follows:

	System I (no power)	System II (some power)
1. delayed neutrons	2	2
2. {Moderator, armor, etc. parasitic absorption in pile	1.5	2.5
3 and 11. fission and corrosion products, chemical separation	3.5	5
4. tank	1	1
5. parasitic escape Wrong direction	.5	1
6. 24 absorption	1.5	1.5
7 and 8. 13 absorption in blanket	1.5	1.5
9. parasitic absorption in reflector D_2O etc.	1	1
10. parasitic escape Reflector, etc.	1	1.5
	13.5%	17%

Quals 1/2

It is unnecessary to remark that these figures give, in the best case, an orientation and that the chances are that they are optimistic. The losses which were enumerated are probably all present and it is unlikely that they are greatly overestimated in the systems considered. However, there may be other losses which we failed to recognize. In spite of this, it appears, at present, that the efficiency losses can well be held below the 37% figure which is the margin if Anderson and May's η for 23 proves accurate.

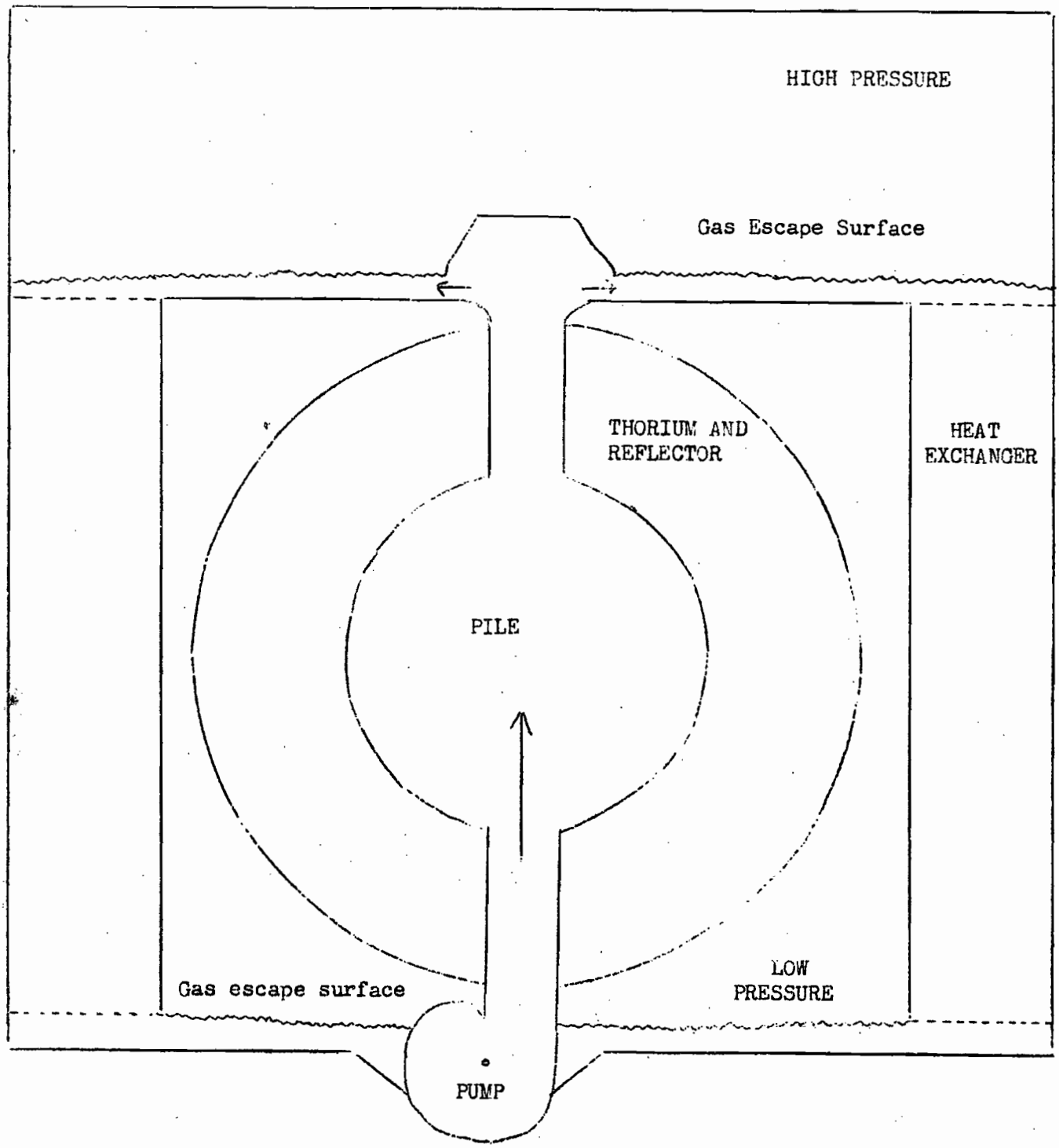

Fig. 1

The degassing method is only illustrative.

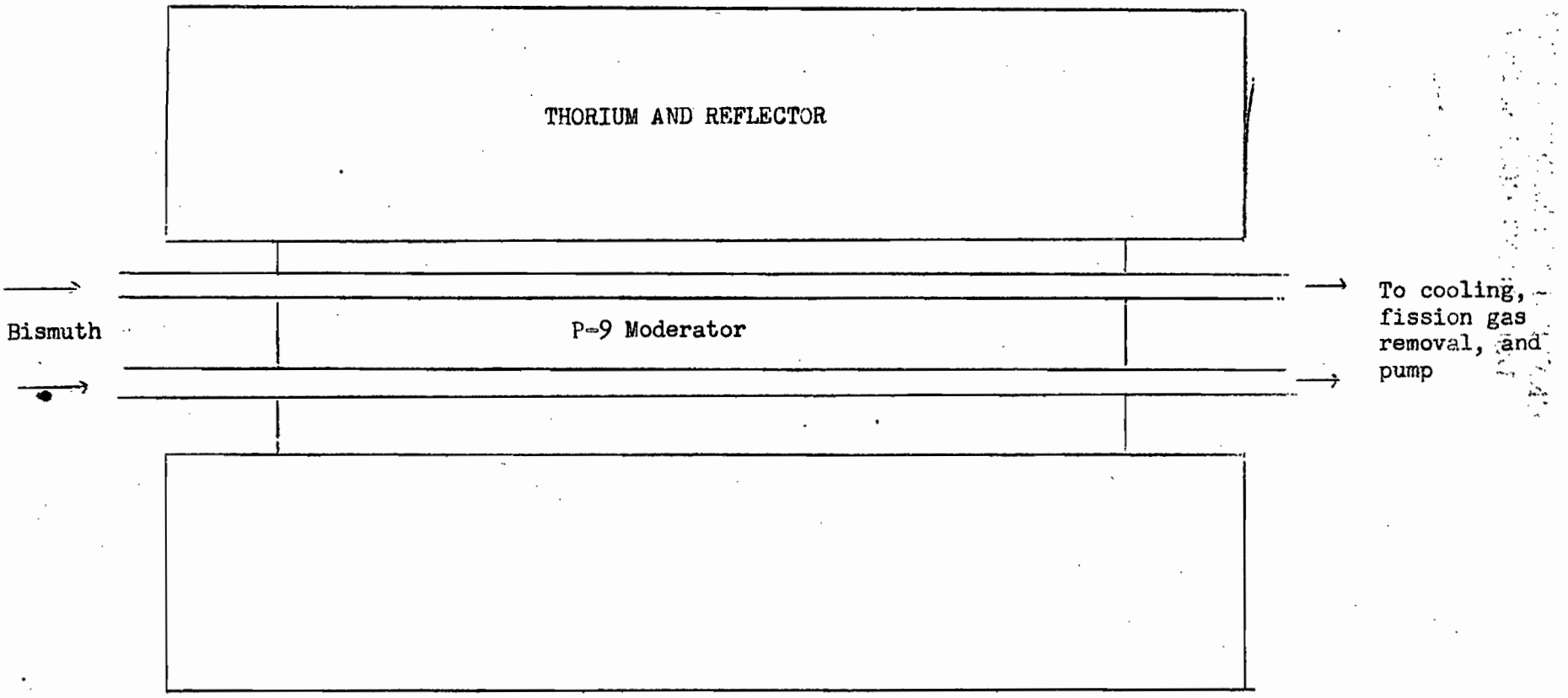
THORIUM AND REFLECTOR
Bismuth
P-9 Moderator
To cooling, fission gas removal, and pump

Fig. 2