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MINUTES OF LECTURE ON APRIL 14, 1945

HIGH TEMPERATURE PEBBLE FILE

Farrington Daniels

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(Minutes written by L. A. Ohlinger)

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MINUTES OF LECTURE ON APRIL 14, 1945

Farrington Daniels

(Minutes written by L. A. Ohlinger)

HIGH TEMPERATURE PEBBLE PILE

In practice, most machines and engines cool off and our major problem is trying to keep them hot for efficient operation. With piles, the reverse is true. However, the question arises whether our attempts to keep the pile operating temperatures all the way down to a comfortable level may not be over-done. Accordingly, it is suggested that we look into piles which operate at a rather high temperature for a change. The use of high temperature operation adds the potentiality of better realizing the power available from the pile.

Of course, when one thinks of taking out heat, one usually thinks of water because it is inexpensive, easy to handle, and its properties are well known. However, as soon as water comes into the picture, corrosion problems enter with it, which means protecting a vulnerable material like heavy metal. Since the materials available for corrosion protection are practically limited to aluminum or beryllium, the maximum operating temperature of the unit is limited because these materials are more subject to corrosion at higher temperatures. Obviously, one limitation leads to another and so one reasons that dropping these limitations and starting over on a new approach might have much to be said for it.

Breeders are important for the future since, ultimately, we must design atomic power units with a view toward conservation of the critical fissionable material and breeders satisfy this need by producing new fertile isotopes. Let us consider the removal of heat from a unit which produces, say, 100,000 kw from a large pile such as a graphite thermal pile. The mechanical problem is not too difficult, if we are working with a cube some twenty feet or more on the side, but, if we must remove the same amount of heat from a six-foot cube, as is the case with enriched material, the problem is much more complicated.

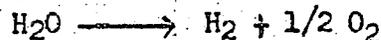
Let us consider the possible methods for removing heat from a pile:

- (1) by flowing a liquid coolant such as water, diphenyl, liquid bismuth, liquid sodium, fluorocarbons, etc., through the pile.

- (2) by flowing a gas such as helium, carbon monoxide, steam, etc., through the pile. Steam as a coolant is inexpensive and quite within reason since there will not be much steam in the pile at any one time and, therefore, the neutron absorption by the steam will be small compared to that of water (about 1/1000).
- (3) by utilizing the latent heat of vaporization of a liquid such as water or bismuth. About three tons of bismuth vaporized every ten seconds will remove 100,000 kw. This represents about three and one-half billion calories per minute.
- (4) by utilizing the heat of fusion of certain materials.
- (5) by promoting a chemical reaction; e.g., by decomposing nitrogen tetroxide to the dioxide. Other highly endothermic reactions are



and



but these two reactions require temperatures over 2500°C which are beyond the practical range of the present refractories. Phosphorus pentachloride decomposition might be utilized but one of the products will be a halide so that the problem of corrosion might become severe. The heat which can be taken out by a chemical reaction varies from 10,000 to 100,000 calories per mole and is not greater than that which can be achieved with other methods of cooling at high temperatures.

Any coolant used for removing the heat from a pile must be a material which absorbs heat readily and has a good thermal transfer. It must be chemically inert and easy to pump. It must have a low neutron absorption coefficient.

With these ideas in view, let us start making a general pile design before compiling a specific picture. First, let us put all structural materials which are absorbers outside of the pile and the enclosing blanket and reflector. Second, let us get rid of all walls and put them completely outside the pile. Third, let us get rid of the poisons as rapidly as they are formed rather than eliminate them by intermittent chemical purification which causes an appreciable loss in fertility.

Let us digress for a moment to consider the argument for high temperature operation as it affects poisons in the pile. If we were to

plot the total cross section for  $Xe^{135}$  against energy we would obtain a curve like that shown in Fig. 1. The Maxwell-Boltzman distribution of neutrons is indicated in Fig. 2 for 0.05 electron volt, corresponding

FIG. I

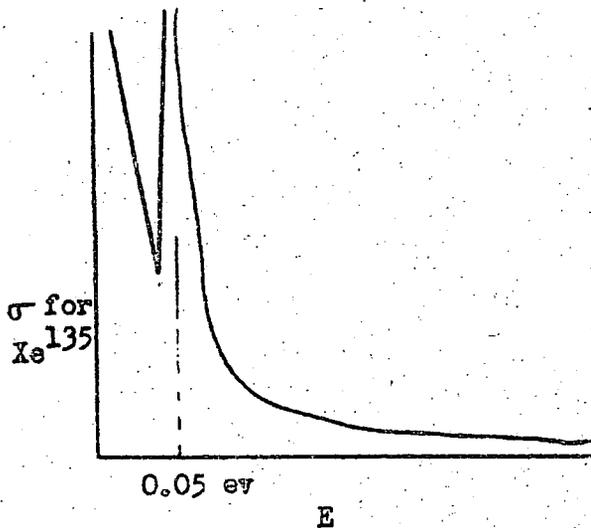
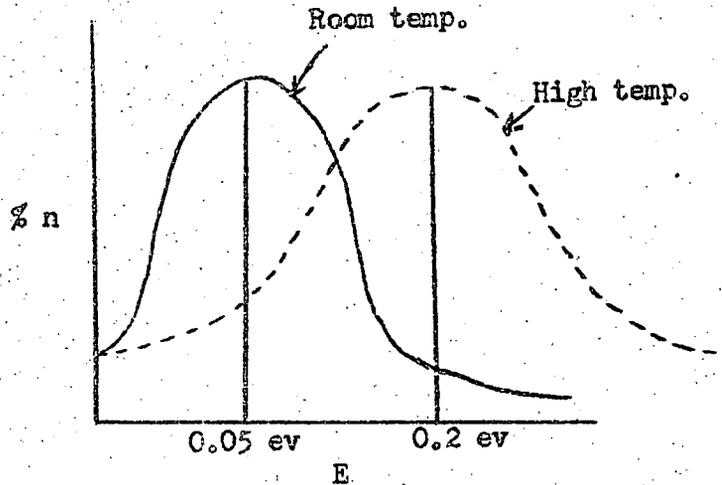


FIG. II



to room temperature, and for  $0.2 \text{ ev}$  corresponding to a high temperature pile at about  $1500^\circ\text{C}$ . It is seen that at room temperature most of the neutrons are absorbed in  $Xe^{135}$  by resonance but that at  $1500^\circ$  the majority of the neutrons will have energies so great that they will miss this resonance peak and escape absorption by the  $Xe$ . It is likely that the other fission products such as  $Gd$  and  $Sm$ , which act as serious poisons for present piles, will absorb neutrons at room temperature and will fail to absorb strongly in a high temperature pile. It must be emphasized that although the poisoning by some of the fission product elements will be eliminated by operating at high temperature, other fission product elements which are not now regarded as poisons may have resonance absorption at  $0.2 \text{ electron volts}$  which would then cause them to be poisons for high temperature piles. At least one would avoid some of the known poisons.

It has been the practise at W and X to can all heavy metal for corrosion protection and then dissolve the can away in order to process the metal, but at  $2000^\circ$  one has to discard canning and substitute some other form of protection. This has an obvious advantage since canning has been a troublesome operation at best. By using oxides of the metal, one no longer is confronted with a corrosion problem.

If bismuth were used as the coolant on the high temperature pile, the Health Division would be much more concerned about the health

hazards than they are for the present piles because the concentration of neutrons within the pile is greater, and dangerously large quantities of polonium are produced.

Now, let us develop a rough picture of a high temperature pebble pile as we now conceive it. The furnace or reactor would be about 6-feet in diameter and about 6-feet high composed of pebbles or spheres of active material and moderator interspersed in predetermined arrangement. The most promising form of the moderator would probably be BeO which would be formed into pebbles by compounding a paste of powdered BeO and  $\text{HNO}_3$ , molding the paste into pellets of the desired size and shape, and then burning out the binder. Similarly, pellets of  $\text{U}^{233}\text{O}_2$  or  $\text{PuO}_2$  could be formed and the desired lattice arranged by interspersing the active material pellets with the moderator pellets. However, for continued and practical operation, it will probably be necessary to use a homogeneous arrangement with no fixed placement of the active material pellets in the pile.

The pellets can be made of mixed oxides of BeO containing a little  $\text{UO}_2$  by precipitating from a solution containing both Be and U salts. In this way the  $\text{U}^{233}$  will be dispersed uniformly through the whole pile thus reducing the localization of heat. In order to retain the pile proper in its original shape without the use of a structural tank, it is proposed to surround the six-foot cylinder with another layer of BeO pebbles of graded sizes compacted and, if necessary, bonded together to form a pebble tank. It is hoped, that this pebble tank would retain its general shape without disintegration when the adjacent active pebbles were removed for processing and replaced by new pebbles. Immediately surrounding this pseudo-tank would be another blanket of pebbles of ThO interspersed with pebbles of BeO for the production of 23. A final layer of small BeO pebbles would surround the Th-Be blanket for heat insulation as well as for nuclear purposes. Outside this would be the enclosing steel tank to which the coolant piping would be connected.

The coolant is pictured as water in the form of steam in either one of two fashions—either as saturated steam at very little above the temperature of boiling water or as water near the boiling point which would then be vaporized in the pile and pick up additional heat as superheat in the steam. The latter arrangement adds the advantage of utilizing the latent heat of vaporization of the water for carrying away the heat of reaction from the pile but complicates the problems by the presence of boiling water in the pebbles in the lower section of the pile and steam in the upper section. Fundamentally then, we have a system such as sketched in Fig. 2.

Water at, say,  $180^\circ\text{C}$  would be pumped into the pile at the bottom and split into cooling streams which would enter the central section of the pile proper and the thorium blanket surrounding the pile at about

10 atm. This water would be vaporized in the lower section of the pile and pass upward through the pebbles as steam picking up superheat en route and emerging from the pile at temperatures near 2000°C at about 1 atm pressure. This superheated steam could then pass through equipment in which a portion of the heat would be transferred to another medium (such as in a waste heat boiler) which could utilize the heat at temperatures consistent with the present top operating temperatures for mechanical equipment. The partially cooled steam could then be utilized to drive turbines or other mechanical equipment in which the steam would ultimately be condensed back to water or else the remaining heat could be dissipated by condensing in a heat exchanger. The resulting condensate should pass through a filter of some material, say, amberlite, which would remove any suspended fission or corrosion products. From this point a pump would pick up the water and deliver it back to the pile at around 10 atm. If necessary, the water entering the pile could be superheated to nearly vaporizing temperature by counter-current exchange with a portion of the existent steam at any point in its travel from the pile to the ultimate condenser.

A few actual figures might be of interest at this point. Such an arrangement of pebbles in a pile would contain about 40% voids. To remove approximately 100,000 kw from such a unit would require pumping about 100 cubic feet of liquid water per minute as steam passing through the bed of pebbles. To force this much water and steam through a pebble pile would require about 9 or 10 atmospheres pressure drop through the system. Line sizes for the steam piping would not be excessive. The steam lines from the pile would undoubtedly require a refractory lining of MgO for such operating temperatures. The pressure drop through the pile and turbulent activity are factors which would tend to lift the pebbles right out of the bed so that a net or screen or a heavy weight would be required to hold the pile shape. This pebble disturbance raises the question of dust production from the abrasion of pebbles rubbing and shuffling against each other in their turbulent state in the pile.

The controls for such a pile would probably consist of a liquid cooled iron pipe inserted in a thimble-shaped crucible built into the bed of pebbles. Experience on high temperature pebble units for nitrogen fixation have demonstrated that such shapes can successfully be operated in temperatures such as we are considering with this unit.

It must be pointed out that the design described above is in its most preliminary stages as the subsequent discussion indicated. For example, no blanket is shown at the top and bottom ends of the pile in order to facilitate the removal and replacement of the pebbles in the pile and blanket. This means that there will be a loss in neutron utilization at the ends of the pile with a subsequent loss in fertility. Obviously, a blanket should be added at these ends which constitute such

a large percentage of the total surrounding surface. Pebbles and larger spheres of BeO at the top and bottom would provide a satisfactory reflector. This complicates the removal and replacement problem which is already a difficult one at best and some further study must be instituted to develop a practical means of handling the pebbles in the pile and blanket. The feasibility of removing only the pile and blanket pebbles without disturbing the pseudo-tank pebbles and outside layer of heat insulation does not bring to mind any immediate solution for the mechanical problems involved.

It has been suggested above that the steam emerging from the pile be utilized in a waste heat boiler which would convert the resultant steam in the secondary system into usable power by means of a turbine. This means an overall loss in mechanical efficiency and brings out the possibility of utilizing such steam directly in a turbine instead of going through a secondary medium. The major difficulty involved in this solution is that, at this time, no materials are known which would be suitable for turbine parts for a machine operating in the neighborhood of 1500-2000°C. The best operating temperature for turbines which has been achieved to date is 1500°F (815°C). Undoubtedly, the erosion problems for such high temperature steam would be serious.

It was hoped that piles operating at temperatures in the neighborhood of 2000°C would have the advantage of boiling out the undesirable fission products thereby avoiding the necessity for chemical processing and purification which causes a loss of the fissionable material. The first experiments have shown already that there is considerable volatilization of the fission products from UO<sub>2</sub> at 2000° and further work is in progress.

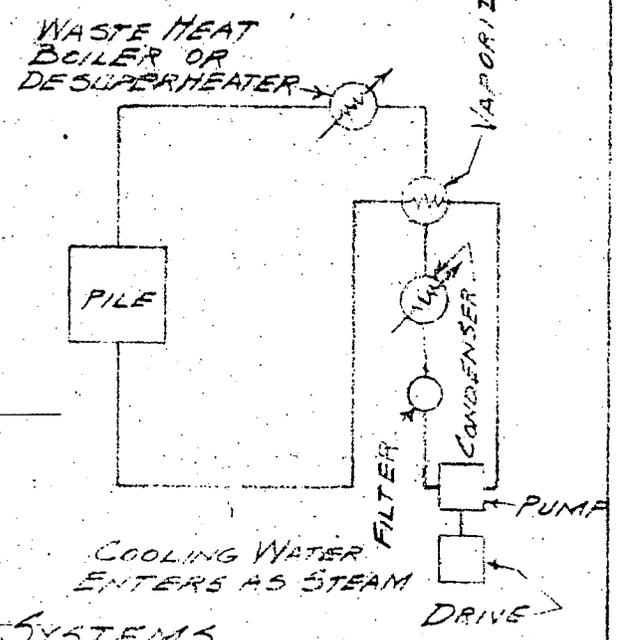
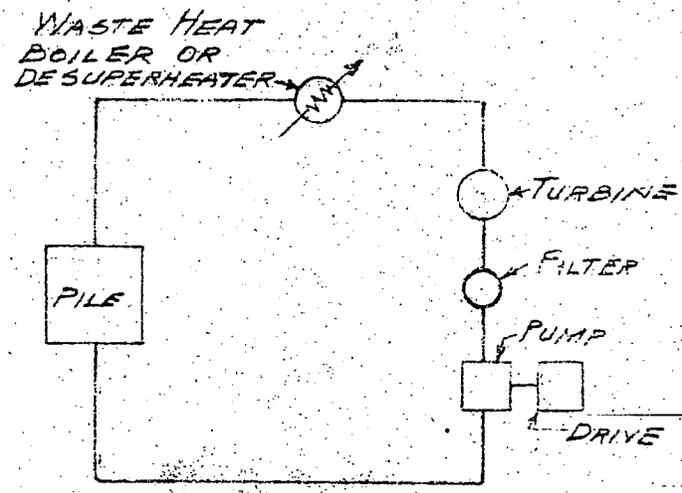
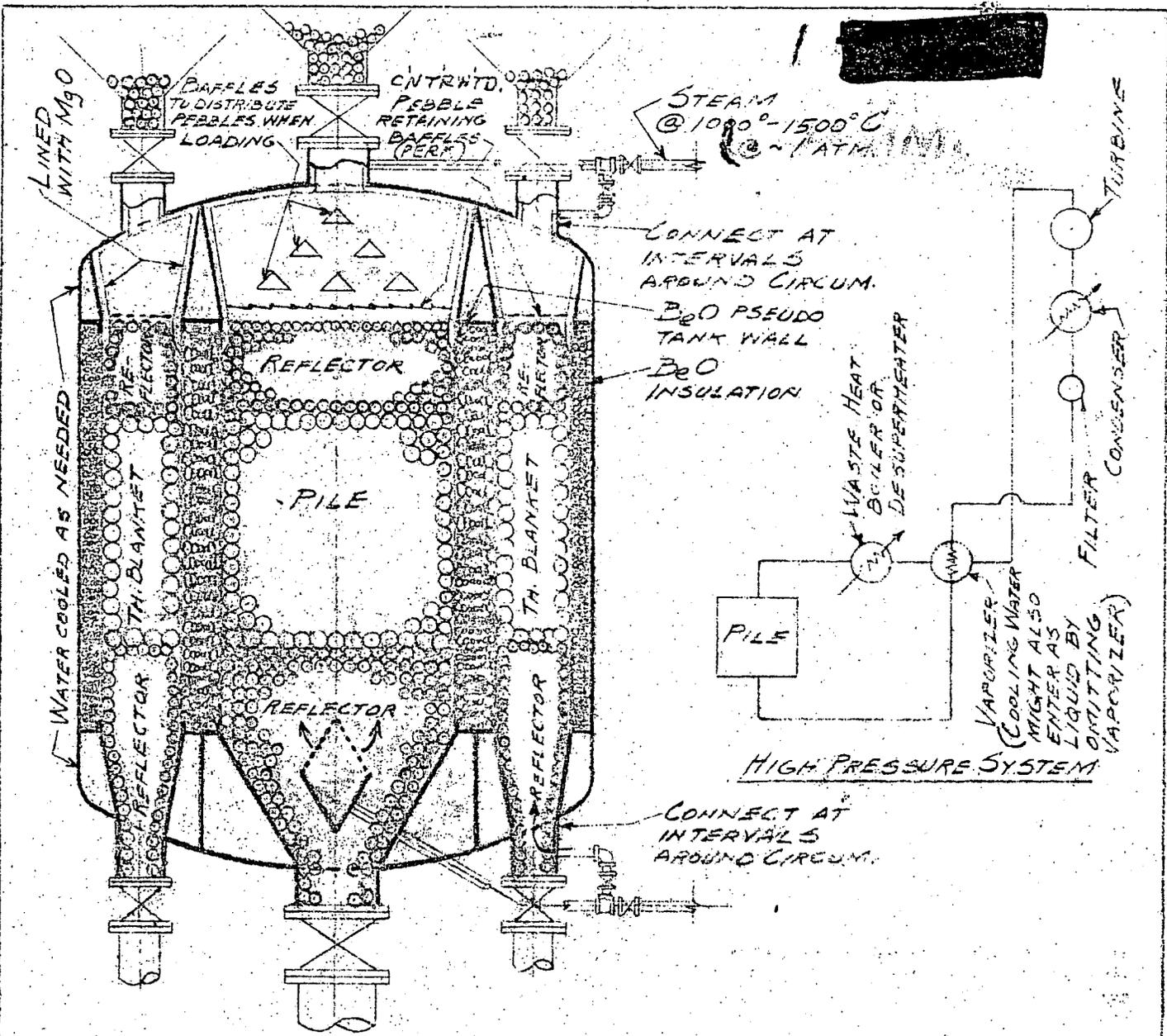
The high operating temperature proposed for the pile brings up some other questions. Whatever the emergent steam temperature might be, it is obvious that the temperature at the center of any pellet will be appreciably higher and that there will be a marked gradient from the center to the surface of any such pellet because of the poor transfer coefficient for tuballoy oxide. This would introduce thermal expansion problems and might actually cause disintegration of the pebbles by thermal cracking. The high operating temperature at the center of the metal pebbles in localized areas might actually exceed the melting point of the material so that it is not inconceivable that some pebbles might actually disintegrate and drip down through the balance of the pebble pile in such a way as to block off a portion of the upper area for gas passage. In the mixed pellets of BeO containing less than 1% UO<sub>2</sub> this excessive heating will be less serious.

By operating at rather high pressures, say, 50 atmospheres, it is found that the drop through the pile may be only around 1 atmosphere

which would indicate that the hot water pump might be omitted in favor of a stand pipe which would provide a water leg of condensed liquid sufficient to insure a suitable flow of coolant through the pile without the use of mechanical equipment.

The manufacture of pebbles for such piles does not appear too difficult even where residual activity necessitates the use of remote control in manufacture. The active dust raised in manufacturing or in the pile might be removed by cyclone separators or electrical precipitators. Tubes or rods of the BeO and UO<sub>2</sub> might be substituted for pebbles in order to eliminate some of the objection raised above by the question of heat transfer and thermal expansion for such shapes. This means that these would have to be more carefully investigated before arriving at any decision. Condensing the steam to water which can be readily pumped appears to be the simplest and least expensive way of recirculating a coolant. If helium were used instead of steam a great deal more helium would be required than steam and one fears a mass of helium equipment comparable in size to that proposed for a graphite pile.

Obviously, there are many physics, chemistry and engineering problems which must be solved before such a pile can approach reality. Many objections arise, not the least of which is the operating temperature of 2000°C which will probably have to be lowered somewhere to 1000°-1500°C but experience is available in the continued operation of nitrogen fixation furnaces at 2100°C. The arrangement for a high temperature pebble pile and the use of the stable, refractory oxides have been presented not as a proposed ultimate design but as a foundation or starting point for preliminary thoughts on a unit which will produce power while breeding fertile isotopes.



DESIGN BY L.A.O.

LOW PRESEURE SYSTEMS

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