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THE UNIVERSITY OF CHICAGO

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To: T. R. Hogness

From: F. Daniels

Subject: SUGGESTIONS FOR A HIGH-TEMPERATURE PEBBLE PILE

Attention is directed inthis report to the possibility of building a pile of pebbles of uranium carbide and graphite, or equivalent materials, which will operate at 1500 to 2000° C. The pile is cooled either by circulating helium or boiling bismuth. The cooling gas passes uniformly through the whole cross-section of the pile, which is kept at a steady temperature by convection and radiation. Operation at a high temperature simplifies the cooling operation, opens up the possibility of separating Pu and fission products by vaporization, and leads to greater thermodynamic efficiency in the operation of engines. Experience gained during the recent operation of a gas-heated pebble-bed furnace blowing 2000 cu. ft. of air per minute for nitrogen-fixation, warrants the expectation that there are no serious practical difficulties in the operation of such a pile at 2000° C.

Tentative Description of Pebble Pile

100 tons of U, occupying 4.9×10^6 cc is disposed throughout the pile at regular intervals as 2-inch spheres of U carbide.

Graphite spheres or pebbles, 2 inches in diameter, make up the rest of the pile. The volume of graphite spheres is 4.9×10^8 cc, which is 100 times the volume of the U.

The total volume of the spheres is 4.95×10^8 cc.

The total volume of the pile is 9.9×10^7 cc, or nearly 36,000 cu. ft., assuming 50% voids, and not allowing for close packing.

The pebble pile may be considered as a cylinder of the following dimensions:

Height	-- 36 feet
Diameter	-- 36 feet
Cross-section	-- 1018 sq. ft.
Volume	-- 36,000 cu. ft.
Weight of U	-- 100 tons
Weight of graphite	-- 860 tons

These are tentative dimensions, assumed in order to permit calculations on cooling. The exact dimensions can be determined only after careful calculations involving the influence on k of the voids and the high temperature. It is likely that the pile may have to be much larger; perhaps so large as to render the high-temperature pebble-pile impractical unless it is made with enriched U.

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Cooling with Helium at 2000° C

Helium gas is blown through the pile, which consists of a lattice of U carbide spheres and graphite spheres, every fourth or fifth sphere in each direction being U carbide. The heat from the U carbide spheres is spread throughout the whole pile by radiation, convection and conduction and is removed by the stream of helium which enters the pile at 100° and leaves at 2000° as shown in Fig. 1. The cross-section for cooling with helium is the cross-section of the whole pile, whereas in other piles containing solid rods of U in solid graphite blocks, the cooling gas must be passed only along narrow paths adjacent to the surface of the U rods. At high temperatures (2000° C) the transfer of heat by radiation is important and it tends to keep the whole pile at a uniform temperature, thus minimizing the overheating of the center of the pile. There will, however, be a vertical temperature gradient due to the flow of colder helium. The exact quantities and arrangement of U and graphite suggested here are tentative. They can be modified extensively when calculations of "k" are made.

The pile is cooled by circulating helium at the rate of 300,000 cu. ft. per minute. The helium leaves the bottom of the pile at 2000° C, is passed through a water-cooled boiler at 100° C, and returns through blowers to the top of the furnace. The heat capacity of helium is 6.3 cal/cu ft/°C. Thus, the heat removed by the circulating helium is:

$$3.0 \times 10^5 \times 6.3 \times (2000° - 100°) = 3.6 \times 10^9 \text{ cal/min} = 250{,}000 \text{ KW.}$$

The rate of flow through the 1018 sq. ft. of pile is:

$$3.0 \times 10^5 / 1018 = \text{approximately } 300 \text{ cu ft/min/sq ft of pile.}$$

The estimated pressure drop for helium blown through a 36 ft. bed of 2" spheres at 2000° C at a rate of 300 cu. ft. per minute per sq. ft. is about 3 lbs. per sq. in. In these calculations the pressure drop may be considered to be approximately the same as that of air at room temperature, because the absolute temperature is nearly eight times as much, but the gas density of helium is only one-seventh that of air. (If it is found desirable for the proper functioning of the pile to use 1" spheres, the pressure drop through the pile will be more than doubled).

The power required for blowing gas against a 3-lb. pressure drop is approximately 15 KW per 1000 cu. ft. per minute, and the power necessary to cool the pile is then

$$3 \times 10^5 / 1000 \times 15 = 4{,}500 \text{ KW.}$$

The initial cost of blowers capable of operating against a 3-lb. pressure drop is about $0.6 per cu. ft. per minute. Single blowers up to 100,000 cu. ft. per minute are available as standard equipment. The delivery time on blowers is much quicker than it was a year or two ago. The initial cost of blowers for cooling the pile would be about 300,000 x $0.6 or $180,000.

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It would be difficult to obtain U carbide in perfect spheres, but lumps or irregular chunks can be used equally well in a pebble bed for efficient cooling at high temperatures. If the neutron requirements are too unfavorable for U carbide, it would be possible to use molten U in crucibles of graphite or Be oxide packed into the pebble pile. If necessary to use solid U spheres, the pile could be operated at 900° C instead of 2000°, but the cost of cooling would be greater and the several advantages of high temperature would be lost.

Cooling with Vaporizing Bismuth at 1450° C

The heat of vaporization of a liquid provides an excellent means for cooling. Metals will withstand the high temperatures and the radioactivity without decomposition, and bismuth appears to be the best on account of its low absorption of neutrons. Dr. Leo Szilard has given careful attention to piles cooled with liquid bismuth. It is proposed to set up reflux condensers above the pebble pile, and drip the condensed liquid bismuth down into the pile where it will vaporize and keep the pile at the boiling point of bismuth, which is 1450°. A diagram is given in Fig. 2.

The liquid does not have to bathe the whole surface of all the U spheres, since the whole interior of the pile will be maintained at a fairly constant temperature by radiation and by turbulent bismuth vapor. It may be necessary to set small vertical graphite tubes in the pile below the condensers and extend them to different depths so that the liquid bismuth will be discharged more uniformly throughout the pile.

The heat of vaporization of bismuth is 47,800 cal/mol. The atomic weight is 209. To remove heat equivalent to 250,000 KW or 3.6×10^9 cal/min requires the evaporation of

$$209 \times 3.6 \times 10^9 / 4.78 \times 10^4 = 1.57 \times 10^7 \text{ g Bi/min} = 17.3 \text{ tons Bi/min.}$$

The bismuth vapor contained in 18,000 cu. ft. of void spaces at 1450° C weighs 0.8 ton. In addition there will be a considerable amount of liquid bismuth dripping down over the spheres or in the channels.

There are 216 spheres per cu. ft., each with an area of 0.09 sq. ft. and in the total pile there are 7,770,000 spheres, with a total surface of 699,000 sq. ft. The necessary "hold up" of liquid bismuth in the pebble bed pile is very uncertain. Assuming a complete turn-over of the evaporated metal every 10 seconds, there will be 3 tons of Bi in the pile at all times. The cost of 3 tons of bismuth is about $8000. No blowers would be required, except for a small pump to circulate tothe top of the pile any liquid bismuth which falls below the bottom of the pile.

Because the heat transfer from condensing bismuth to the condenser is very efficient, the pile could probably b e operated at much higher heat levels, 500,000 or 1 million KW. - limited only by the quantity of bismuth which can be allowed in the pile and the effectiveness of the condensers.

If the pile is operated without attempting to recover the power, air condensers could be substituted for water condensers. The top of the pile would then be occupied by many tall chimney-condensers of iron pipe lined with graphite tubes, around which a strong current of air is blown.

Control Rods

Water-cooled iron pipes containing cadmium rods as cores can be lowered into graphite cylinders (not shown in Figs. 1 and 2) which are packed vertically in the piles. In the helium-cooled pile the graphite tubes are closed at the bottom to prevent short-circuiting of the gas; in the bismuth-cooled piles the graphite tubes are open at the bottom to permit drainage of the liquid bismuth. Water-cooled iron pipes have been used in the magnesia pebble bed furnace which has been operated for many months at the University of Wisconsin. There was no difficulty in having a water-cooled iron pipe surrounded by a mass of pebbles at 2100° C.

For simpler control rods it may be possible to use vertical iron tubes of large diameter filled with boiling water and provided with water-cooled reflux condensers at the top of the furnace.

Metals, such as sodium, could be dropped into the pile to reduce k in case of an emergency. In an extreme emergency, ordinary water could be sprayed into the furnace from safety sprinklers at the top. The neutron concentration and the temperature would be reduced immediately. The hot uranium might suffer damage, but experiments on the magnesia pebble beds at 2100° showed that there is no serious explosion hazard from the steam.

Power

The high temperature of the circulating helium or the condensing bismuth permit excellent heat transfer and efficient operation of a boiler for power purposes. The negligible distance between pile and boiler minimizes heat loss and construction cost. The fact that the boiler is entirely outside the pile makes possible the use of ordinary water, iron, and other standard materials.

The heat of the pile can be liberated just as well at 2000° as at 200° and the possible thermodynamic efficiency is then very much greater. In order to utilize this high efficiency (1900/2273) if the lowest temperature is 100°) it would be necessary to use gas engines or turbines. The development of high-temperature turbines has been hampered by the fact that all high-temperature gases obtained by the combustion of fuel contain oxygen which reacts chemically with the materials of construction which are capable of withstanding high temperatures. However, helium or bismuth permit the use of a turbine made of tantalum, tungsten or [illegible] material which retains its mechanical strength at high temperatures in the absence of nitrogen and oxygen.

Removal of Fission Products

At temperatures above two-thirds of the melting point, the diffusion of material in a crystal lattice is reasonably rapid. It seems likely that fission product elements produced in U carbide at 2000° would diffuse to the surface of the spheres in a few hours. Some of these fission product elements such as I, Br, Xe, Kr, Te, would volatilize out and be condensed in the boiler tubes where they could be dissolved daily. In this way at least part of the chain poisoning by fission product elements would be eliminated, thus preventing the decrease of k. Moreover, the intense radioactivity would not be accumulated at the end of the 3-month period which is planned for the W pile, because the radioactivity would be removed to a safe distance at frequent intervals, thereby simplifying shielding problems during recovery. With the enormous volume of helium sweeping through the pile, a considerable amount of material would be evaporated, even if the vapor pressure is very low. For example, if the vapor pressure of a substance in the piles is only one-millionth of a millimeter at 2000° C, the volume of gas swept by is so great that a gram mole will be vaporized every day.

To assist in the removal of fission products, Dr. James Franck has suggested the use of electrostatic precipitation. There is intense ionization in the gas but helium with its high ionization potential will contribute but few ions and the fission product elements and other impurities in the helium will carry the current and be deposited on the charged water-cooled plates from which they can be dissolved at intervals and collected.

Removal of Product

After operation of the pile for a sufficient length of time, the furnace is cooled down by inserting the control rods and the pile is dumped through water-cooled doors into a dissolving pit at a lower level. By making the U carbide lumps slightly smaller than the graphite spheres, they can be sifted out from the graphite as the material passes along an inclined trough. The U carbide is dissolved in acid and the Pu is separated by the standard chemical methods now in use. The graphite spheres can be carried to the top of the furnace by a conveyer operated by remote control and dumped into a new pile, the fresh lumps of U carbide being dropped in by remote control at the proper positions and at suitable depths while the pile is being filled.

It may be possible to distil the product out of the pile during the operation of the pile. This procedure would make it unnecessary to dismantle the pile every three months, particularly if some of the fission products could be removed also by distillation fromthe pile. Experiments here indicate that U carbide does not distil appreciably out of a graphite crucible at high temperatures. Reports from Site Y suggest that pure Pu metal can be vaporized from Pu carbide at high temperatures. Above certain temperatures then, Pu, which is formed in the crystal lattice of U carbide, might diffuse to the surface, be evaporated in the stream of helium and condensed out on the water-cooled surfaces or in an electrostatic precipitator. It could be dissolved from these surfaces when necessary by injecting a suitable solvent through water-cooled pipes without shutting down the pile. If the dissociation of the carbides and the volatility are not sufficiently different in the case of U and Pu to effect a complete separation, they may be sufficient, nevertheless, to give a more concentrated mixture of Pu in U which can be used for "enriched piles"; or they can be separated chemically by the standard method, using pounds of material instead of tons.

It may be possible to separate the volatile radioactive fission products from the Pu by distilling out the former at a lower temperature and then raising the temperature to distil out the Pu, or Pu mixed with U.

One of the present difficulties with the ultimate use of the product lies in the presence of about three percent of 40-10 produced by subsequent absorption of neutrons by Pu. If it is possible to distil Pu out of the pile within an hour or so of the time of its formation, pure 49 could then be obtained, free from 40-10. If necessary to accelerate the removal of Pu from the pile, the U carbide may be used in the form of thin discs instead of spheres, so that the diffusion distance to the surface is reduced. The production of pure 49 in this way seems quite possible.

Alternative Piles

Beryllium oxide is a suitable moderator and it is a solid up to 2400° C. If a pile is made with pebbles of U oxide mixed with pebbles of beryllium oxide it is then possible to cool with air. Recirculation and cooling will not be necessary then if the radioactive fission products are removed.

It should be emphasized that in a pebble pile it is not necessary to have perfect spheres; lumps or irregular chunks are entirely satisfactory. No machine work is necessary. Lumps of U oxide and beryllium oxide can be used directly without reduction to the metal.

As a cooling gas hydrogen would be much better than helium, but its probable reaction with graphite renders it impractical. Carbon monoxide might be used, but its higher molecular weight would increase pumping costs.

Possible Difficulties

The high temperature of 1450 to 2000° C may seriously affect k. However, there may be advantages as well as disadvantages in these high temperatures.

The void spaces may require a larger increase in size than has been allowed for in the calculations given here in order to give a large value of k, and the void structure may require an excessive width of reflecting material. It should be emphasized that there are no structural materials within the pile, no iron, aluminum tubes nor water. The elimination of these features will partly offset a possible decrease in k due to high temperature and voids.

The heavier U carbide spheres may settle among the lighter graphite spheres, thus spoiling the lattice arrangement. Experience with magnesia pebbles of different sizes at 2100° C suggests that this is unlikely. If necessary, graphite rods can be used to preserve the lattice; or the U carbide can be in the form of large disks with protruding rods of U carbide or graphite to act as anchors.

The blowers may become too radioactive for repair and upkeep. Standard blowers are available which run for years without repair. Most of the volatilized radioactive material will be deposited on the water-cooled surfaces, but if necessary a large chamber packed with pebbles to give a large surface could be interposed between the boiler and the blower. The electrostatic precipitator may prevent the circulation of radioactive fission products.

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The water boiler may not be adequate to lower the temperature of the circulating helium to a point where it can be safely fed into the blowers. Supplementary water coolers can be introduced beyond the boiler. If necessary, the warm gas can be cooled in pebble beds which have a high heat capacity and very effective heat exchange with a gas flowing through them. A pair of pebble beds is used; one is cooled by blowing through air while the other is in use in the stream of warm gas.

The U carbide in 2" spheres may attain a temperature which is too far above that of the surrounding graphite spheres. This situation is unlikely at these high temperatures where the transfer of heat by radiation is large; but if necessary the U carbide lumps may be made smaller or the shape may be changed to give a flatter thinner lump with quicker heat conduction from the center.

Leakage of air might lead to oxidation or nitridation of the U carbide, but the welded iron enclosure will be tight. The helium will be kept at a pressure slightly above atmospheric. The large excess of graphite will react with any oxygen which might get in.

In the boiling bismuth pile the whole pile is enclosed in iron and the air originally present is swept out with helium or carbon dioxide.

The hold-up of liquid bismuth on the surface of the spheres may be so great as to cause a large absorption of neutrons. This question can be answered by simple laboratory experiments.

Production of 23

Surrounding the pile is an enclosure of graphite bricks and then about two feet of insulating material such as powdered graphite, granulated coke or magnesium oxide of size 8 mesh to dust. By using thorium oxide for the insulating material surrounding the pile, it would be possible to obtain production of 23 in quantity. The 23 can be extracted from the thorium oxide when the pile is shut down, or the thorium oxide can be drawn off at the bottom at frequent intervals during operation of the pile and replaced by fresh thorium oxide added at the top.

Thorium or thorium carbide could be placed within the pile itself at frequent intervals if the k is sufficiently large. These pieces of thorium can be recovered when the furnace is dumped, if they are made somewhat smaller than the U carbide spheres and screened out in the chute.

The present piles at X and W, already constructed, can not now be used for the production of 23 from thoria. It would be easy, however, to produce 23 around a new pebble pile such as the one described here. There are no complications in the form of pipes protruding from the sides, so that it will be a simple matter to pack thoria and graphite around the pile. There are seve al reasons why it would be advantageous to build a pile which can be used for the production of 23.

The following research problems are suggested;

I. Calculations of k.
II. Experiments at high temperatures using an induction furnace.
1. Relative volatility of Pu and U from a synthetic sample of U carbide containing small amounts of Pu.
2. Volatility of Pu from U carbide exposed in a pile
3. Volatility of fission product elements from U carbide exposed in a pile.
4. Electrostatic precipitation of Pu and fission product elements evaporated in a stream of helium from U carbide exposed in a pile.
5. Distribution of temperature (by radiation) in a lattice of spheres in which some are heated hotter than others by induction.
6. Possible interaction between U carbide and graphite.

III. Experiments with a pilot plant furnace (1 ft. by 6 ft. high) in which U carbide and graphite spheres are heated to 2000° C with circulating helium passing through an electric arc between graphite electrodes.
1. Measurement of pressure drop through the hot furnace
2. Efficiency of heat transfer to a water boiler at one end of the furnace
3. Data on the cooling efficiency of liquid bismuth, the hold-up of liquid bismuth on the spheres and the efficiency of heat transfer to the boiler.
4. Possible settling of U carbide spheres.

Summary

I. Experience gained in the operation of gas-heated pebble beds of magnesia give confidence that there are no serious engineering difficulties involved in the operation of a pebble pile at 2000° C.

II. The advantages of high-temperature pebble piles are:

1. Simplification and reduction in the cost of cooling.
2. Uniformity of temperature through increased importance of thermal radiation at high temperatures.
3. Increased thermodynamic efficiency for power purposes if a turbine is used; increased boiler efficiency if a steam engine is used.
4. Elimination of water cooling, canning, and corrosion.
5. Elimination of the Wigner effect in which energy is stored in a rigid graphite lattice.
6. Elimination of machining and precision work in the construction of the pile.
7. Elimination of complicated pipe structure at the sides of the pile, thereby permitting simple water shielding against neutrons and simplifying the production of 23 from thoria.
8. Elimination of cooling pipes and foreign material from the interior of the pile, thus avoiding a decrease in k.

III. There may be additional advantages from the continuous diffusion to the surface and evaporation of some of the fission product elements, including the following:

1. Removal of some chain poisons, thus tending to maintain the full value of k.
2. Reduction in radioactive radiation from the pile.
3. Reduction in radioactive radiation from the product during the chemical operations of recovery.

IV. There is a possibility that Pu may be distilled out of the pile continuously and collected, with the following advantages.

1. Long operation of the pile before shutting down for the recovery of Pu in the U carbide spheres.
2. Recovery of the Pu in small batches of distilled product instead of one large batch. The handling of pounds instead of tons reduces the size of equipment and the shielding.
3. Removal of the Pu from neutron bombardment as soon as it is formed before there is an opportunity to build up the objectionable 40-10 isotope.
4. If some U is distilled out with Pu, the mixed product can be used for "enriched piles" without further purification if the fission products can be distilled out at a lower temperature or precipitated electrostatically.

V. By using thorium oxide for an insulating lining around the pile, it is possible to obtain considerable quantities of 23.

VI. A high-temperature pebble pile permits the production of Pu and the production of power at the same time.

VII. Laboratory tests could be made on:

1. Temperature distribution in a bed of U pebbles surrounded by pebbles of nonconductors heated in an induction furnace.
2. Cooling by helium of hot pebbles of U carbide and graphite.
3. Effectiveness of heat transfer of helium and bismuth vapor to water-cooled boiler tubes.
4. Diffusion and volatility of fission products and Pu in U carbide at 1450° and at 2000°.
5. Electrostatic precipitation of fission products volatilized at high temperatures
6. Cooling efficiency and hold-up with boiling bismuth.

VIII. If calculations should show that the value of k is satisfactory, it would be possible to build a pile in a short period of time, because no special machinery and no precision work on the construction of the pile would be needed.

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IX. Operating data and heat losses from the magnesia pebble bed furnace operated at 2100° C will be made available to the project if there is an interest in them.

X. If the high temperature and the void spaces should lead to the requirement of impractically large quantities of U and graphite, the high-temperature pebble pile described here may still find post-war application in enriched piles for the generation of power.

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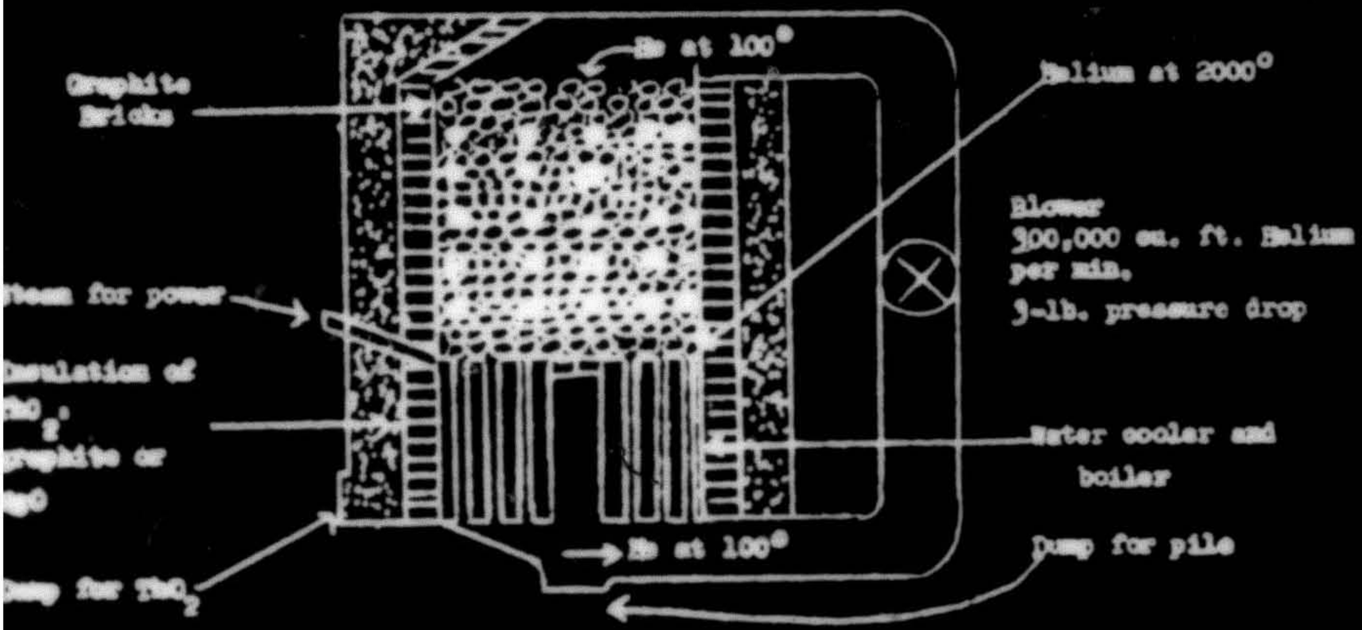

Fig. 1 Helium-Cooled Pile Operating at 2000° C

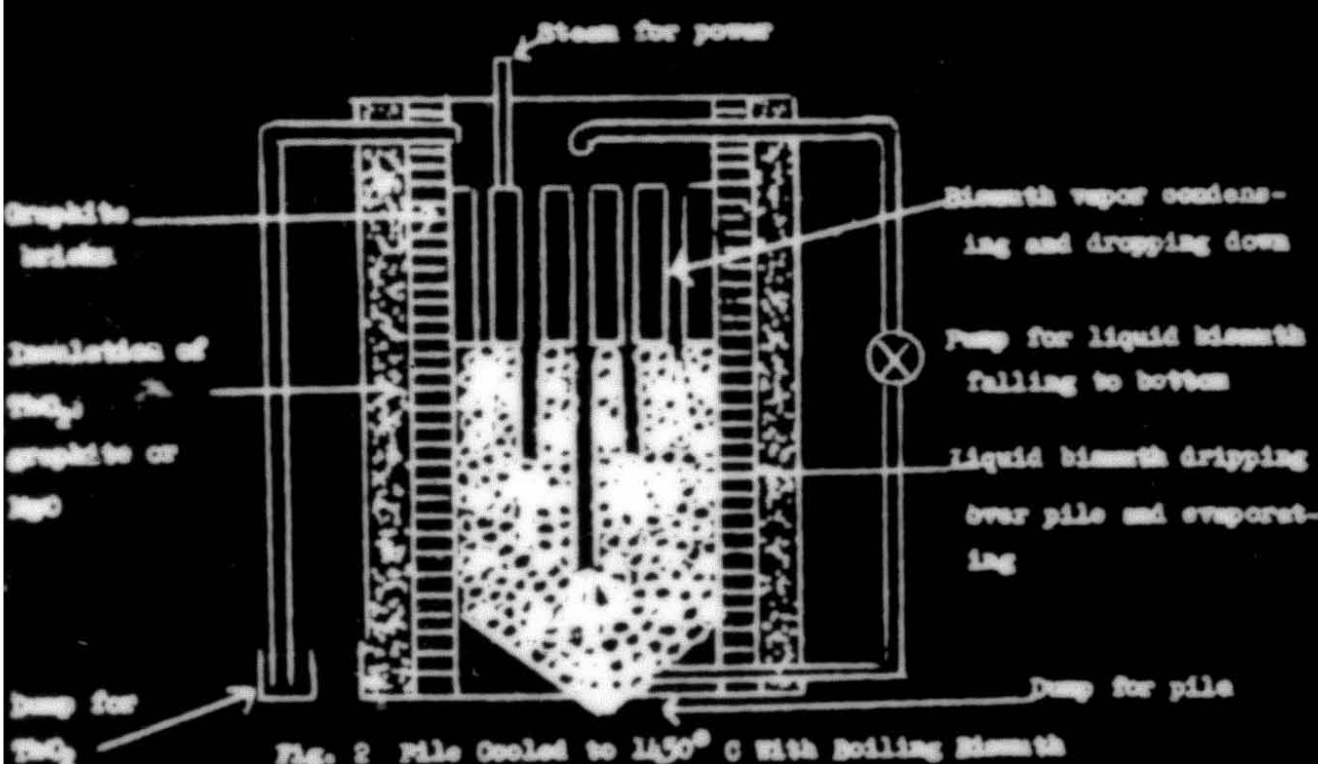

Fig. 2 Pile Cooled to 1450° C With Boiling Bismuth