Dear Dr. Zinn,

Mr. Stadler informed me in a recent letter that you were surprised to find my data notebook gone and that you would like for me to return it. I am having it returned to you immediately.

I brought the notebook with me for two reasons: (1) I had insufficient time to organize the data taken on my last working day at Argonne and (2) I thought I was expected to take the notebooks which were checked out to me wherever I went.

I have now finished pondering over the data, and have, in fact, written a report which is being sent to you. A diagram of the latest and best thermopile is shown on page 162 of the notebook. A drawing should probably be inserted in the report, but I have no drawing instruments here. The data is shown on Pages 162 - 164. I plotted the thermopile and ion chamber deflections, determined the sensitivity, and estimated the time lag when the pile was warmed. The results are quite gratifying. Perhaps a graph should be included in the report also.

As you suggested I brought one of the thermopiles with me. Two other similar ones were left there for your use, in addition to the two older ones which have been in the pile about three months. Of the two new ones, one was left in the possession of Mr. Priel at Kyerson, who agreed to coat it with boron by evaporation when his new coating apparatus arrived. The other had two broken wires which I pointed out to Mr. Fowler. It can be easily repaired and coated.

The one I brought with me has the boron painted on it. For that reason I am not entirely satisfied with it, but did not have time to wait for more experimenting on evaporation.

I believe we are "over the hump" as far as solving thermopile problems are concerned. I am quite pleased with the results obtained with the last one. Here's hoping they turn out well for you.

Although the differential bimetallic strip I constructed was not a factory, the changes needed to make it practical were rather clearly indicated, namely those which would result in greater sensitivity. I discussed the problem.
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Dr. W. H. Lime
February 23, 1944

in detail with the instrument group, and Mr. Goodale planned to have one made of a design which we agreed should have ample sensitivity for even a low-intensity pile. Since his design looked very good, I left the problem entirely in his hands.

Living conditions here leave a lot to be desired, but I believe the work will be very interesting. I'm a supervisor in the 300 Area -- reporting to Mr. Dineen. For the first time in my experience with Du Pont I am in "Operations" rather than the Technical Department.

Best wishes to you and all the gang!

Very truly yours,

J. H. West

JW:1hr
At.

RECEIVED
May 18, 1943
200 Area
Classified Orders
A few weeks ago developmental work on thermopiles for measurement of neutron density had reached the stage where reduction of the temperature coefficients of the resistance seemed to be the only major problem remaining to be solved. In a report at that time it was suggested that a new design was required for overcoming this difficulty. Other improvements in design were also mentioned. The present report summarizes experimental work with an improved design which the writer believes will be quite satisfactory for monitoring piles operated at high intensities.

The advantages of the latest design over those previously described are:
(1) Its time lag under the most adverse conditions (screwing the pile) is about one second as compared with four seconds for the previous design.
(2) The size has been reduced many fold. The dimensions of the base are 1 x 1 x 1/2 inch. When placed in a graphite box the over-all dimensions are 1 x 1 x 1 inch. (3) The base is of lave instead of graphite or aluminum. This is an advantage in that lave is an excellent insulator in addition to being composed of elements having low neutron cross sections.

Like previous ones, this thermopile was made of bismuth alloy wires. One alloy contains 3% antimony; the other has 3% tin. The wires are much smaller, however. Whereas the wires in the thermopiles previously described were 0.007 to 0.010" in diameter, these have diameters of only 0.004".

Since thermopiles with both sets of junctions suspended in air followed ambient temperature changes of the pile better than those in which alternate junctions were attached to relatively large masses of aluminum or graphite and cooling fins, the present design is of this type. The wires were wire-crossed on a narrow bridge so that both sets of junctions are suspended over identical slots. The bridge is only 1/32 inch wide. Grooves were carefully machined in the bridge for positioning the wires. Each groove is 0.007" deep and makes an angle of 6° with the perpendicular to the bridge. The wires were cut to a length of 7/8" inch. The grooves were so spaced that the wires touched and exerted a slight pressure on each other at the vertices. The reduction in length of the wires made possible the use of smaller wires, without making the electrical resistance prohibitively high.

Laveite is a variety of talc, a magnesium silicate material which is soft and easily machined in its natural form but which becomes very hard when baked at high temperatures. The laveite was machined in its soft form to the required dimensions for the base. The parallel slots on either side of the bridge were filled with paraffin to support the bridge while the grooves for holding the wires were being machined.

When the machining was completed, the paraffin was melted out of the slots. The remaining film was removed with benzene. The base was then heated in a muffle furnace at 1000°C. for five hours. Upon removal the laveite was quite hard. Because of its size, the bridge was rather fragile, however, and care had to be exercised to avoid exerting much pressure on it.

After baking, lead pins were made to fit the holes for the binding posts. The pins were 1/32 inch in diameter and 1/8 inch long. These were the binding posts to which the end elements of the thermopile, and the connecting leads, were to be soldered. Small sections of brass were machined to fit exactly the parallel slots and of such a height that the top surface was about 0.005" below the bottom of the
grooves in the bridge. Thin strips of mica were cemented with Dune to the brass plugs. The plugs served as supports for the ends of the wires while the junctions were being made. The mica strips were necessary to prevent the wires from fusing to the brass. The thermal insulating properties were also advantageous in that the soldering pencil could be used at a lower temperature.

The wires were mounted individually. Tweezers were necessary in handling the wires while each end was being dipped into soldering paste. The wire was then lifted and carried by means of a small, flexible wooden stick which had been dipped in water. The surface tension of the water is sufficient to support the wire. The stick was used to place the wire in its groove and center it properly. When it was centered, a very tiny drop of clear Glyptal was placed over the wire and pushed down with a flexible probe. The Glyptal serves as a glue to hold the wire in place while the junctions are being made. Because several minutes are required for the Glyptal to dry to the desired consistency, several wires were mounted in succession before fusing the ends together. Otherwise, the surface tension of the wire as it melted was sometimes sufficient to pull it out of its centered position.

A magnifying glass is necessary for all the operations just described. Before fusing the ends, they must be examined to make certain that good contact is made. A slight pressure between the wires is desirable. A temperature just under visible red for the soldering pencil was found satisfactory for making the junctions. Some skill and patience is required in making the junctions properly, but after some practice they can be made at a rather rapid rate.

It is somewhat easier to solder the extremes of the thermopile to the binding posts if the end wires are of the Bi-5% alloy. This has a higher melting point than the Bi-2% alloy. To avoid spoiling the entire thermopile while mounting these end wires, the mounting was done in the following manner. Wires of the proper length for the end thermocouples were cut, dipped in soldering paste and placed on a sheet of mica, with their axes parallel and the ends overlapping about 1/2 mm. After making sure that the wires were in contact, the junction was made by bringing a red-hot soldering pencil near. At just the right distance the wires melt and fuse together forming a junction slightly larger than the wires themselves. The thermocouple was then placed in the end groove, with the Bi-5% wire lying in the groove in the lead binding post. A tiny bit of the bismuth-lead eutectic alloy (60% Bi, 40% Pb) was placed over the binding post and melted with the soldering pencil. The copper leads were attached in the same manner. The Glyptal was then applied on the bridge and the junction on the opposite side of the bridge made in the usual manner. In this way, if the last thermocouple should be broken, or the soldered joint at the binding post is unsatisfactory, another thermocouple can be constructed and a replacement made without ruining the entire thermopile.

When all the junctions had been completed, the thermopile was placed in an oven at 115°C for two hours. The temperature must not exceed 125°C, since that is the melting point of the bismuth-lead eutectic. A 5 Ohm bath was used to remove the excess soldering paste, after which the brass plugs were removed and the thermopile was ready for mounting. The resistance of the completed thermopile was 13 ohms.

An effort was made to coat one row of junctions with boron by evaporation. A brass box was made which shielded all parts of the thermopile except the row of junctions to be coated. Merser, Freil and Gilpatrick kindly aided the writer by operating the evaporation apparatus they had been using for coating ion chambers. It was not possible to obtain a satisfactory coating, however, because the low melting point of some of the constituents of the thermopile made it necessary to mount it at
a considerable distance from the electrode where the evaporation took place. A graphite electrode painted with a boron-carbonoid 1:1:1:1 mixture was the source. An opaque coating was finally obtained, but this was believed to contain high percentages of other materials than boron, principally carbon because several graphite electrodes burned out during the operation, and the electrodes were not outgassed in all cases before the thermopile was placed in the bell jar.

After depositing the opaque coating, the other row of junctions was coated with aquadag. The thermopile was mounted in a graphite box of dimensions 1 x 1 x 1 inches and placed at the center of the Argonne pile where its sensitivity and time response were tested. The sensitivity was extremely low—about 1 cm. per 1000 K.V., but no time lag in response was noticeable.

The sensitivity was increased by painting on the "hot" junctions small quantities of the following mixture:

2.5 gms. boron
1 gm. Carbonoid A
Acetone to make very thin.

Upon drying completely, the above mixture was rather hard. A smaller proportion of Carbonoid A would probably have been adequate for binding purposes.

After coating as described above, the thermopile was tested again. The sensitivity had increased to 0.293 cm. per K.V. This sensitivity is approximately one fifteenth that of the thermopile described in a previous report (C.P.). A smaller quantity of boron was painted on in order that the time constant might be shorter. The sensitivity is more than adequate for use in the "V" pile. The response was rapid, although a very small time lag could be observed upon scanning the pile. In the first ten seconds after scanning, the ion chamber deflection decreased by a factor of 3.4. The thermopile deflection reached the mark required to maintain the proper ratio, approximately two seconds later. This time lag was undoubtedly due to two effects.

1. The required damping resistance for the galvanometer was 48 ohms. Since the resistance of the thermopile was only 13 ohms, the galvanometer was considerably overdamped.

2. The heat capacity of the junctions was increased considerably by painting on boron. Using an insulating material such as Carbonoid A for a binder is not desirable, since it decreases markedly the rate at which heat exchanges can take place.

Perhaps one second of the time lag was due to each cause.

Data collected on thermopiles up to January 24 make possible some conclusions regarding: the practicality of using them for monitoring purposes; (1) Thermopiles made of B1-5a and B1-5b elements, with alternate junctions sealed with boron, produce an emf. under neutron bombardment which is a linear function of the intensity over the range tested; (2) The response is instantaneous except insofar as rate of heat transfer is limited by the heat capacity of the elements and the insulating properties of the coating materials; (3) No changes in calibration were detected during a period of ten weeks when a thermopile of this type was under observation; and (4) Slight changes in sensitivity can be expected if the ambient temperature changes in the region where the thermopile is located. This will be due to changes in the resistance of the thermopile and changes in the thermolectric power of the elements. A more complete discussion of this follows.
For a 1°C temperature rise, the resistance of pure bismuth wire increases by a factor of 1.00%. If it can be assumed that the bismuth alloy wire increases in the same proportion, an increase of 0.4% in resistance of the thermopile can be expected for each degree in temperature. The amount by which this reduces the sensitivity depends upon the resistance of the other elements of the circuit. If the resistance of the thermopile is small compared with that of the remainder of the galvanometer circuit, the effect of the small resistance change will be negligible. For the particular galvanometer circuit used at Argonne, the constants were:

- Resistance of galvanometer = 11.6 ohms
- Resistance of connecting leads = 5.0 ohms
- Resistance of thermopile = 15.8 ohms

Since the total resistance of the circuit is 28.6 ohms, the effect of a 1°C temperature increase on sensitivity would be \( \frac{(0.4\%)(13)}{28.6} \approx 0.16\% \)

Since the required sensitivity for monitoring a pile operating at high intensity can easily be obtained, the effect of the change in resistance due to temperature, on sensitivity, can be reduced to an insignificant quantity by using a high resistance galvanometer or by inserting a series resistance.

The change in thermoelectric power produced by temperature changes cannot be overcome so easily. Using the values given in the International Critical Tables for thermoelectric power of the alloys, a 1°C temperature rise will produce a decrease in thermoelectric power of 0.16%. Since this is the only source of E.M.F. in the circuit, the sensitivity of the instrument will be decreased by this amount. Although this change in sensitivity is not high, corrections could be applied for it by measuring the temperature at the point where the thermopile is located. It may be possible to maintain the temperature at nearly a constant value. If this is possible, the above effect can be neglected.

Another possible way to overcome the above effect, if it proves to be a serious one, is to use thermocouple elements whose thermoelectric power does not vary appreciably with temperature. Such a combination is the bismuth-silver thermocouple. A thermopile made of pure bismuth and pure silver has a number of advantages. Because of the low electrical resistance of silver, a thermopile of the same size as the one described for bismuth alloys could be constructed with a resistance of only 5 ohms. The thermoelectric power of the Bi-Ag combination is only \( 2/3 \) that of the Bi-Sb vs. Bi-3Sb alloys. Taking account of the resistance, and using the same galvanometer, the thermal sensitivity of the Bi-Ag thermopile would be \( (28.6)(2) \approx 0.52 \) times that of the one described above. The above value is only approximate since there will be some difference in the size of the junctions, the rate of conduction of heat away from the junctions, etc. The higher thermal conductivity of the silver and the bismuth should have the double effect of lowering the sensitivity and decreasing the time constant of the instrument. Since sensitivity is no longer a serious problem, the decrease in time constant would be a definite advantage.

A thermopile of bismuth and silver was actually constructed some time ago and coated with boron. The thermopile was of the type in which the uncoated junctions were attached to binding posts, however, instead of the differential type. Tin solder was
used in making the junction between the bismuth and the silver. The high neutron cross section of silver produced a residual activity which was undesirable in that the R.M.P. did not drop toward zero as rapidly as the neutron density upon standing. In the differential type, this would be overcome, since the two sets of junctions would be identical except for boron on one set.

The chief advantage of a bismuth-silver thermopile would be the fact that the thermoelectric power varies only a negligible amount with temperature. The thermoelectric power decreases by 0.03% when the temperature is increased 1%. The use of bismuth and silver instead of the bismuth alloys is recommended only in cases where the Bi-Sn or Bi-Sb thermopile cannot be used because of large ambient temperature changes in the pile.

Although a high sensitivity thermopile is not required for monitoring piles operating at high power levels, such as those at Site W, high sensitivity is a desirable feature. Calculations made on the rate at which boron impurities will disappear in the W pile led to predictions that after 100 days continuous operation at 250,000 W, approximately 1/3 of the boron in the graphite will have disappeared. If the boron on the junctions of the thermopile were destroyed at this rate, a very noticeable decrease in sensitivity would result. By making the thermopile very sensitive, it can be placed in a place where the neutron density is low, and the rate of decrease in sensitivity will be very small. A high sensitivity instrument could also be placed more easily in a region where ambient temperature changes were minimal.

It is believed that most of the important problems connected with thermopile design for monitoring piles have either been solved, or the solutions indicated. The instrument described in this report has sufficient sensitivity, and has a time lag of only one second under the most adverse conditions (scramming the pile). Its small size makes possible its use as either a portable or a permanent installation in a very small hole. There are several ways by which the time constant could be further reduced, if that is desired, namely: (1) By using smaller wires, thereby reducing the heat capacity of the junctions. Much smaller wires could be mounted under a microscope; (2) By coating the junctions with boron by evaporation rather than painting it on with a lacquer. This should be possible when water cooled supports are obtained for the evaporation chamber. The electrode should be thoroughly outgassed before the thermopile is placed in the chamber; and (3) Thin foils can be attached to the junctions to increase the ratio of surface area to mass, if this is necessary. Lead has been tried for this purpose, but tin should be better because of its greater thermal conductivity and greater ease in soldering; and (4) The ratio of surface area to mass could also be increased by depositing films of the elements instead of using wires. This might be done by evaporation or electrolyzing. Messrs. Friel and Gilpatrick have already done some preliminary work on the evaporation method. It might be possible to use ribbons instead of wires.

Lavite has never been exposed to irradiation within the pile for any appreciable length of time. Its value as a structural material is therefore unknown at present. If it does not disintegrate, it should be quite convenient and satisfactory for this purpose.

Another thermopile of the type described was made and left at Argonne to be coated with boron by evaporation, when water cooled supports are obtained for the evaporation chamber. If a good coat of boron can be applied in this manner, a considerably better instrument from the standpoint of relaxation time should result.

Although boron has proved to be better for coating the junctions than ordinary uranium, it is likely that pure U²³⁵ would be even better, because of its high fission cross section and the fact that this metal is a much better thermal conductor than boron.