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NEUTRON DISTRIBUTION MEASUREMENTS AT PAJARITO BY MEANS OF PHOTOGRAPHIC EMULSIONS

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ABSTRACT

A method of using photographic emulsions for the study of neutron distributions is described. The construction of a simple machine for producing experimental critical assemblies in which the method has been used is reported. Sources of error are discussed. Curves of the perturbations when small spheres of various materials are placed in the center of the assembly are shown. (auth)

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NEUTRON DISTRIBUTION MEASUREMENTS AT PAJARITO BY MEANS OF PHOTOGRAPHIC EMULSIONS

I. INTRODUCTION

The study of nuclear transformations began with the discovery by Becquerel in February of 1896 that emanations from uranium blackened a photographic emulsion. Numerous other methods of detecting radioactivity have been developed, but the blackening of a photographic emulsion remains one of the simplest. At Pajarito Site, there has been developed a method of using photographic emulsions to study relative neutron distributions. If a sheet of inert material is placed against a sheet of fissionable material, and both inserted in the flux whose distribution we wish to study, fission fragments will be caught in the inert material. If then the inert material with its imbedded fission fragments is removed from the flux and from the fissionable material, and placed against a photographic emulsion, as the fission fragments decay the resulting beta activity will make developable the grains of silver salt in the emulsion. When the emulsion is processed, the blackening at any point will be a function of the number of neutrons capable of causing fission at that point.

Alternatively, any material which can be made radioactive by neutrons can be used. In the case of such an element, a thin sheet is placed in the neutron flux, irradiated, and then placed against the photographic emulsion. The three detectors of which most use has been made are U-235, U-238, and gold. U-235 has moderate sensitivity to the whole range of neutron energies found in the assembly, with

greatly enhanced sensitivity to those of low energy, gold has moderate sensitivity to the entire range with very great sensitivity to those of about five volt energy, and U-238 is activated only by the higher energy neutrons. In this way we are able to get some information as to the distribution of neutrons of different energies. Other elements which might be used profitably are thorium, manganese, indium, and iodine.

The chief advantage of the method is the excellent resolution in space. The gamma radiation concurrent with the beta activity of fission fragment decay does not produce much blackening of the emulsion, and consequently, the limit of resolution possible is the distance the beta particles travel in the emulsion. This has been found to be a fraction of a millimeter. The aperture of the densitometer used in the present work is approximately 0.070 inch in diameter, and the foils were six inches square. This permits over 5,000 entirely separate readings from a single activation, and the number could be quadrupled by using the aperture half that diameter. Other advantages are the permanent nature of the records obtained, and the small disturbance of the assembly by the insertion of the materials used to obtain the records.

II. APPARATUS

In order to study the effect of inserting small spheres of various materials in an assembly containing essentially fast neutrons, there was constructed a simple machine for producing experimental critical assemblies. The skeleton of the machine is the base and column of an ordinary drill press. A general view is shown in Fig. 1. Occupying the position of the table of the drill press is a frame containing a portion of the active material, and a portion of the tamper. Also mounted on this frame is the "fine control" of the assembly, a 2 x 2 x 2 inch block of Tu which can be moved in or out from the active material by a small motor and a system of gearing. One of the gears is mounted on the shaft of a selsyn transmitter, the receiver of which moves an indicator on the panel in the control room, a quarter of a mile away. The total motion of the block is about an inch and a half, and this travel is indicated in thousandths of an inch. The "coarse control" is obtained by varying the relative amounts of active material and tamper. The units involved are half-inch cubes, the same as used in the Topsy machine[1]. The exterior dimensions of the Tu tamper are approximately that of a cube, eight and a half inches on a side, and the active material consists of between 22 and 23 kilograms of Oy in the center, arranged in the form of a pseudosphere. The amount of active material varies with the foreign material being studied. The full travel of the "fine control" block is equivalent in reactivity change to interchanging between three and four cubes of Oy and Tu at the surface of the pseudosphere.

Mounted in place of the spindle of the drill press is an air cylinder and on the piston of this air cylinder is a frame containing blocks of tamper. Normally, this frame is held up by springs. By admitting compressed air into the cylinder, the tamper is forced down into contact with the material on the table, and on releasing the air, the springs immediately separate the tamper from the material on the table. There is a release valve located close to the cylinder, normally open, and closed by a solenoid. In case of a power interruption, either from manual operation of a switch, the tripping of a monitor circuit, or a line failure, the valve opens. This not only acts as one of the independent safety mechanisms required, but in the "Up" position of this section of the tamper, the material on the table is accessible for making changes.

Approximately half of the active material, and of the tamper, is enclosed in a frame which is mounted on the piston of an air cylinder which stands on the base of the drill press. Admitting compressed air into this cylinder raises this material into contact with the material on the table, which completes the assembly except for the fine control. There is a system of interlocks which prevents the raising of this piston unless the fine control is in the position of least reactivity. A release valve similar to the one on the upper air cylinder is connected to this one also, and when the air is released, the material returns to the unassembled position by gravity. This provides a second independent safety mechanism, and in the unassembled position the median plane of the assembly is accessible for the insertion of the spheres of foreign material and the foils.

In operation, a hemisphere of foreign material, three-fourths inch in diameter, is placed in a cavity in the top of the ram, which is essentially the median plane of the assembly. The material of the assembly, both Oy and Tu, emits fission fragments during operation, and to catch these, and prevent them from interfering with the results, there is placed next to the material of the assembly in the median plane a 0.003 inch foil of aluminum. It has been found by test that this is sufficient to stop all significant activity due to the fission fragments from the assembly itself. Then comes the fission fragment catcher, also a 0.003 inch foil of aluminum, and then the foil of fissionable material. Our foil of U-235 is 95% enrichment, and our foil of U-238 is 5,000/1 depletion. On top of the fissionable material is placed another catcher, then another guard foil, then the other hemisphere of foreign material, and the chase containing a half-inch layer of blocks of active material and tamper. This chase is analogous to the chase used by printers to hold forms of type. It is held in place horizontally by locating cones. It can be lifted off to change foils and hemispheres. To remove foils it is necessary only to lift the chase by the handle, and the rapidity with which this can be done accounts for the fact that in several months of operation no one has received appreciable amounts of radiation. A view of the operation is shown in Fig. 2. Each row of blocks in the chase is held in position by a set screw. The adjustment of these set screws to provide uniform pressure requires care, but has been accomplished sufficiently well that there has never been a spillage of blocks.

III. PROCEDURE

After the hemispheres, foils, and chase have been put in place, a neutron source is inserted in the assembly at the interface between Oy and Tu, and the machine assembled by remote control. The power level required and the time of irradiation have been determined empirically. After an irradiation and the machine has been disassembled, there is a short wait to permit the short half-life fission fragments to decay, and to get rid of the 2.4 minute half-life of the aluminum, which is small, in any case. Then the catchers with their imbedded fission fragments are placed against the emulsion and allowed to remain twenty-four hours. When the gold activation instead of fission fragments is being studied, gold foils instead of aluminum catchers are placed between the guard foils, and the fissionable material foil is omitted. Since the half-life of the gold activity is 2.7 days, the time of exposure to the emulsion is usually lengthened, depending on the amount of irradiation given.

Almost any photographic emulsion can be used, but some are more convenient than others. Eastman Type K X-ray film is very sensitive, has good stopping power for beta particles, is coated on both sides, permitting us to use fission fragments from both sides of the fissionable material, and the graininess has not proven objectionable. It has been used in all of the measurements reported here. Developing has been done in D-19b, with constant agitation by withdrawing the film from the solution, reversing it, and sliding it back in. This assures equal development of both sides of the film, and as uniform development over the

entire film as some of the more complicated mechanical arrangements. Fixing has been done in Ansco Liquafix, with similar agitation for the first minute in the bath, and an occasional reversal for the rest of the time of fixing. Fixing is continued for twice the time required for the film to clear, and the bath is discarded as soon as the time required for clearing is perceptibly lengthened. Dry developing agents keep well, but deteriorate in solution. Also, during the development process the composition of the developer changes, and to assure uniform development, each film is developed in freshly mixed developer, which after one film has been processed is used for other purposes, or discarded.

The density readings have been made on an Ansco Densitometer[(2)]. The term density as applied to photographic films represents a complex phenomenon. Part of the light incident on the film is reflected, part is absorbed, part passes straight through, and part passes through after one or more scatterings. The proportions of these depend on the size, shape, and distribution in depth of the silver grains. No practical densitometer collects all of the scattered light, and all collect part. This densitometer has an electronic circuit designed to compensate for the loss of scattered light. However, any errors in this would be balanced out by the calibration method described later. All that is necessary is that the instrument give reproducible results, and that the same instrument be used for all readings. The stability of the instrument has given concern. It was noted that variations in the pressure of the densitometer head on the film influenced the results, and this was taken care of by using a weight to apply the pressure.

After a small change in density reading, the needle takes up the new position instantly, but after a large change there is a drift for some minutes. The instrument is also sensitive to line voltage changes, both in its sensitivity and in its zero setting. By waiting before making a reading after a large change in density, by frequent checks of the zero setting and the sensitivity, and by averaging a large number of readings, it is believed that the errors due to the densitometer are made negligible.

The characteristic curve of an emulsion, a plot of the density against exposure, is never simple. It depends upon details of development and upon the energy of the exposing agent as well as upon the characteristics of the emulsion itself. It must be measured experimentally under the working conditions as nearly as possible. A series of four exposures was made, similar except for the power level. The same time of irradiation was used, the same time of waiting before the foils were placed on the emulsions, the same times of exposure of the foils to the emulsions, and so on. Each succeeding exposure was at twice the power level of the one before. These power levels were determined in two ways, by an ion chamber made sensitive to neutrons by coating the large center electrode with B-10, connected to an electrometer tube circuit[3], and by three "long counters". These "long counters" had been checked a short time previously by the additive source method, and although there was considerable scatter in the results, there did not seem to be any significant departure from linearity. The readings of the electrometer circuit were in agreement with the readings of the "long counters".

The variations in density from the centers to the

margins of the films gave overlapping readings, and further extended the range. There were overlapping readings from densities of 0.40 to 1.80, and the total range was from 0.28 to 2.65. The readings were plotted with density as the ordinate and exposure as the abscissa on a single large sheet of paper, and the corrections for the curves across the diameter of the active material were made from this sheet. These curves are Figs. 4, 6, 7, 8, and 9. The probable accuracy of the center points relative to points near the margins is believed to be of the order of five per cent.

Not only do the characteristic curves of different types of emulsions vary, there are variations between different batches of the same type of emulsion, between different sheets from the same batch, and even between different parts of the same sheet. For the highest accuracy, it is necessary to calibrate each film, with the same energies of particles with which the measurements are to be made, and as closely as possible to the region to be studied. If we take as the permissible error one-tenth of one per cent, on examining the films it was found that no matter what foreign material was placed in the center of the assembly, the perturbations did not exceed this amount at a distance of an inch from the center. Even with polythene as the foreign material, with which the perturbations are enormous in the center, this is true, as shown by Fig. 10. This is the ratio of the densities of the film made with polythene in the center to the densities of the film made with air in a central cavity, plotted with the density of the air film as the abscissa. It can be seen that although there were different exposures for the two films, as well as there

being differences between the characteristic curves, the points lie along a smooth curve until the point corresponding to an inch from the center is reached. From the slope of this curve, we are able to correct for differences between individual films, and reduce the probable error to a few tenths of one per cent.

It was found by accident that the thickness of oxide coating on the foils of fissionable material very markedly changes the number of fission fragments received by the catchers. A strip of tape had been allowed to remain on one of the foils for some time, and when it was removed it took off most of the oxide, leaving nearly bare metal in this spot. The blackening of the film was much greater in this region than where the oxide remained. Difficulty has been experienced in getting a foil free from oxide, or with a uniform coating of oxide. A foil cleaned electrolytically was returned with streaks of oxide visible to the eye, and even more visible to the catcher foils. The best results thus far have been obtained by scouring the foils, and keeping them in a vacuum when not in use. In addition to the scatter due to differential oxide coating, to the densitometer, to the graininess of the film, and to variations from region to region of the film, there are patterns of greater and less reactivity which occurred in film after film, with all detectors. Some of this seemed to be due to neutrons reflected back from the surroundings, but the greater part is attributed to differences in the U-235 content of the Oy blocks. The result is that there is no single diameter which truly represents the assembly as a whole, and to correct for this as well as secure the repeat readings needed to reduce the statistical errors, readings

were made along 24 radii from the center to the interface between Oy and Tu, at 15° intervals. The readings for each distance from the center were averaged, and corrected for the nonlinearity of the characteristic curve of the film by means of the exposure versus density chart.

IV. RESULTS

Figure 3 is a print from a film made with U-238 as the detector, and gold hemispheres in the center of the assembly as the perturbing material. The overlay shows the boundary of the gold hemispheres, and the interface between Oy and Tu.

Figure 4 is a plot of the relative exposures at various distances from the center as derived from this same film, plotted on the same sheet as the relative exposures derived from a film made with an unperturbed assembly. The value of the exposure is taken as 100 at the center of the unperturbed assembly, and the other relative exposures are given in percentages of this. The abscissa represents distances from the center on the same scale as the assembly itself. The central point represents the average of twelve readings, and each of the other points the average of twenty-four. The right hand portion of the curve is plotted from the same data as the left hand; that is, they are mirror images of each other.

Figure 5 is a print from a film made with polythene hemispheres in the center of the assembly, and gold foils as the detector. Figure 6 is a plot of the relative exposures as derived from this film, plotted with the relative exposures of an unperturbed assembly, in the same manner as Fig. 4. The abscissae are the same, but it was necessary to use a different vertical scale in order to get the points at the center on the sheet. Figure 7 is a comparison of a solid Oy assembly with one with a 3/4 inch air cavity, with gold as the detector; Fig. 8 with U-235 as the detector; and

Fig. 9 with U-238 as the detector.

When the effect of the insertion of a sphere of foreign material is small, it seems of more interest to compare the effect with that of an air cavity, rather than with that of solid Oy. It is also desirable to make corrections for variations between individual films. Referring again to Fig. 10, it will be seen that it is possible to make a reasonable extrapolation from the curve drawn through the points from the surface of the pseudosphere to an inch from the center, and it is the vertical distance of the central point, the point 1/10 inch from the center, the point 2/10 inch from the center, and so on, to this extrapolated curve, which is a measure of the effect of polythene at these points, with this detector. It cannot be too strongly emphasized that it is not the over-all neutron flux which is observed. It is that portion of the neutron flux which activates these various detectors.

The values for the curves of Figs. 11 through 17 were derived from extrapolations made in this manner. In Figs. 11 and 12, the entire series is plotted, with the position of the assumed normal line taken as 100, in order to show the scatter of points which has been observed. In Figs. 13 through 17, only the central portion is shown. The measurement of deuterium ice with gold detector was made by moving the assembly outdoors on a cold night, and with the advent of warmer weather it has not been possible to complete the series. All of the other materials were solids at room temperature. It has been the purpose to explore the methods, as well as to get the localized effect of some typical foreign materials.

V. CONCLUSIONS

It is possible to measure with fair accuracy, and with good resolution in space, the perturbations of the neutron flux as seen by various detectors, by means of photographic emulsions. The perturbing materials studied thus far have been those which could be obtained in solid form. For powders, no binder which contains hydrogen should be used. A thin-walled container of aluminum should introduce very little error, and this sort of container would serve for both powders and liquids. Gaseous elements can be studied by means of their compounds, which can be compared with the non-gaseous element of the compound, such as polythene and carbon. The over-all accuracy, from the center to the margin of the active material, as derived from a single film, is of the order of five per cent. Since the perturbations do not extend to any perceptible degree more than an inch from the center when a 3/4 inch sphere of foreign material is used, it is possible to correct for the variations between films, and the probable error becomes about two-tenths of one per cent in the region of greatest interest. These errors can, of course, be reduced by making several films of a subject.

VI. REFERENCES AND ACKNOWLEDGMENT

REFERENCES

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(2) Electronics, 19 No. 11, Page 105, November, 1946.

(3) Electronics, Experimental Techniques, Elmore and Sands, McGraw-Hill Book Company.

ACKNOWLEDGMENT

All members of Group W-2 have assisted in this work. Especial thanks are due Mr. Kenneth Dunahugh, who assisted in the design and did most of the machine work on the assembly, and to Mrs. Thelma Thomas, who assisted in preparing the graphs.

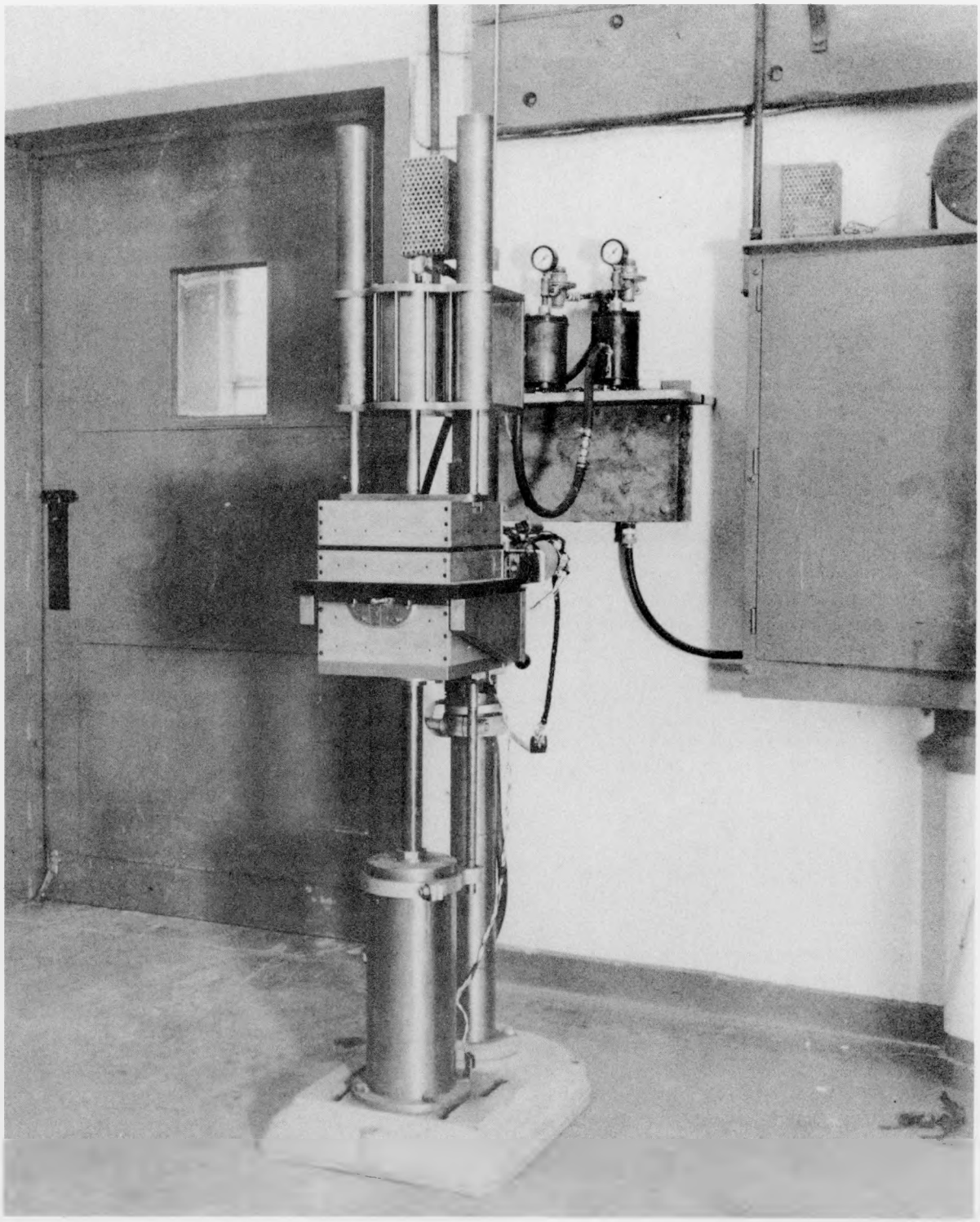


FIG. 1.

FIG. 2.

FIG. 3.

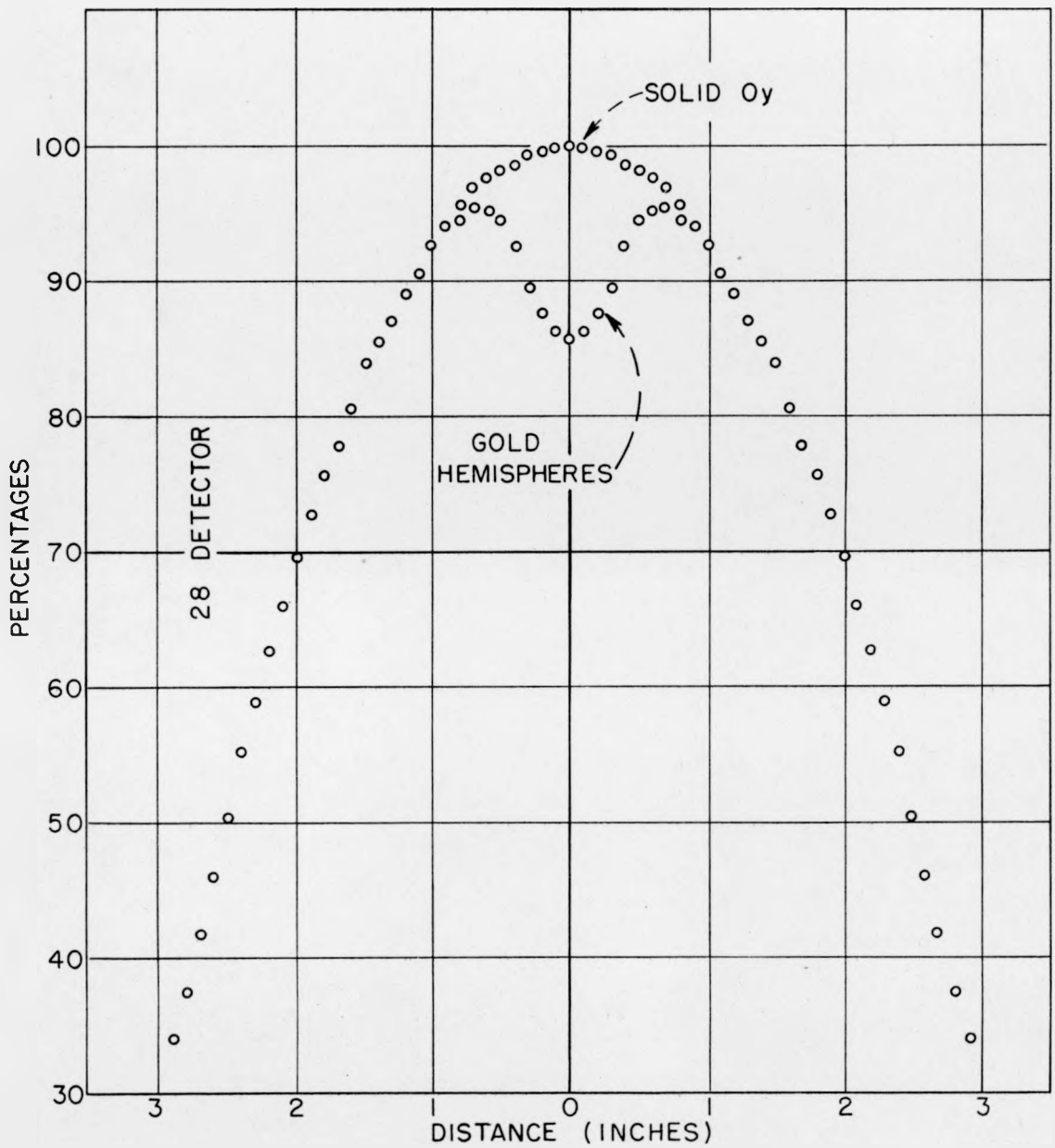


FIG. 4.

FIG. 5.

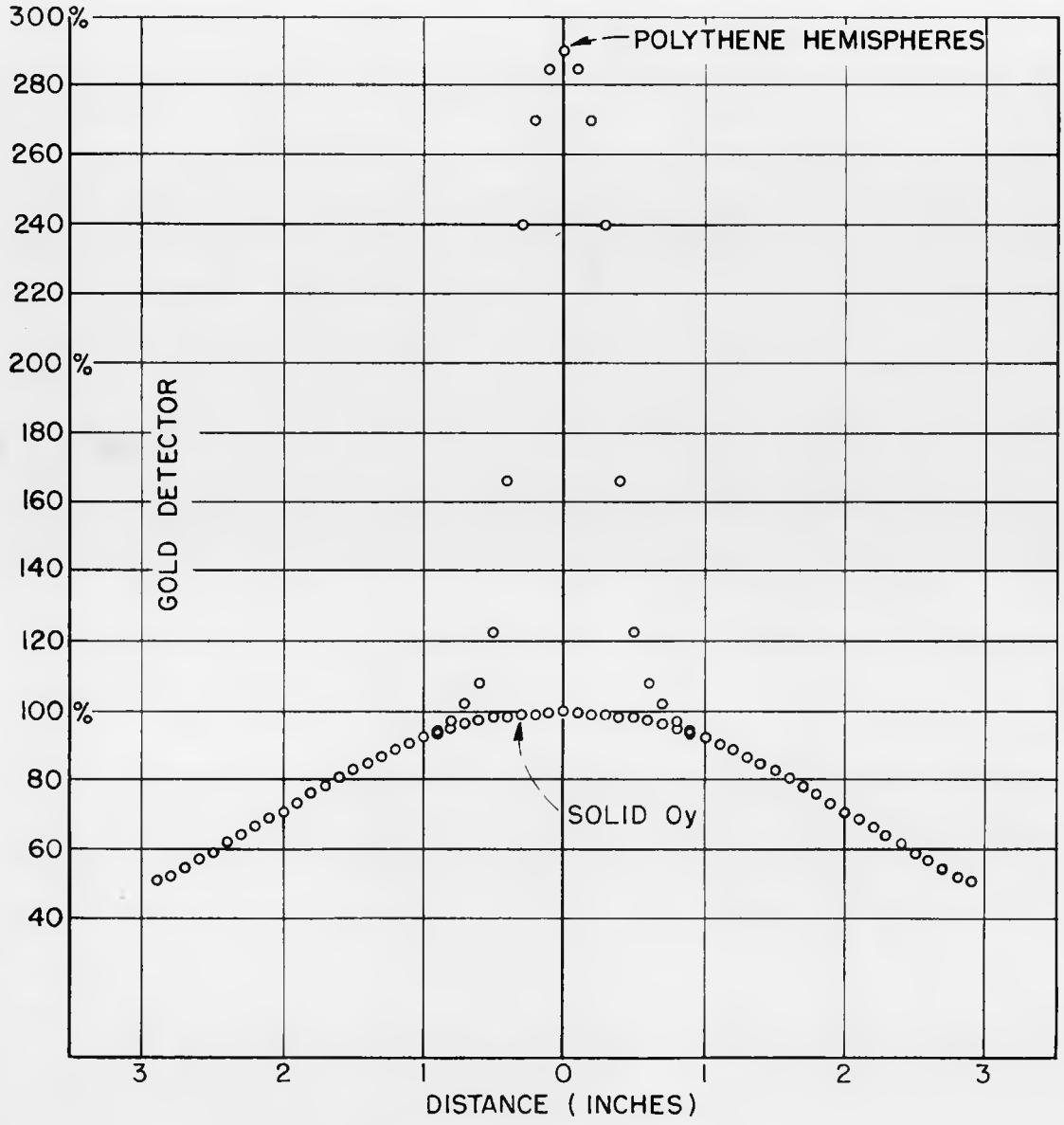


FIG. 6.

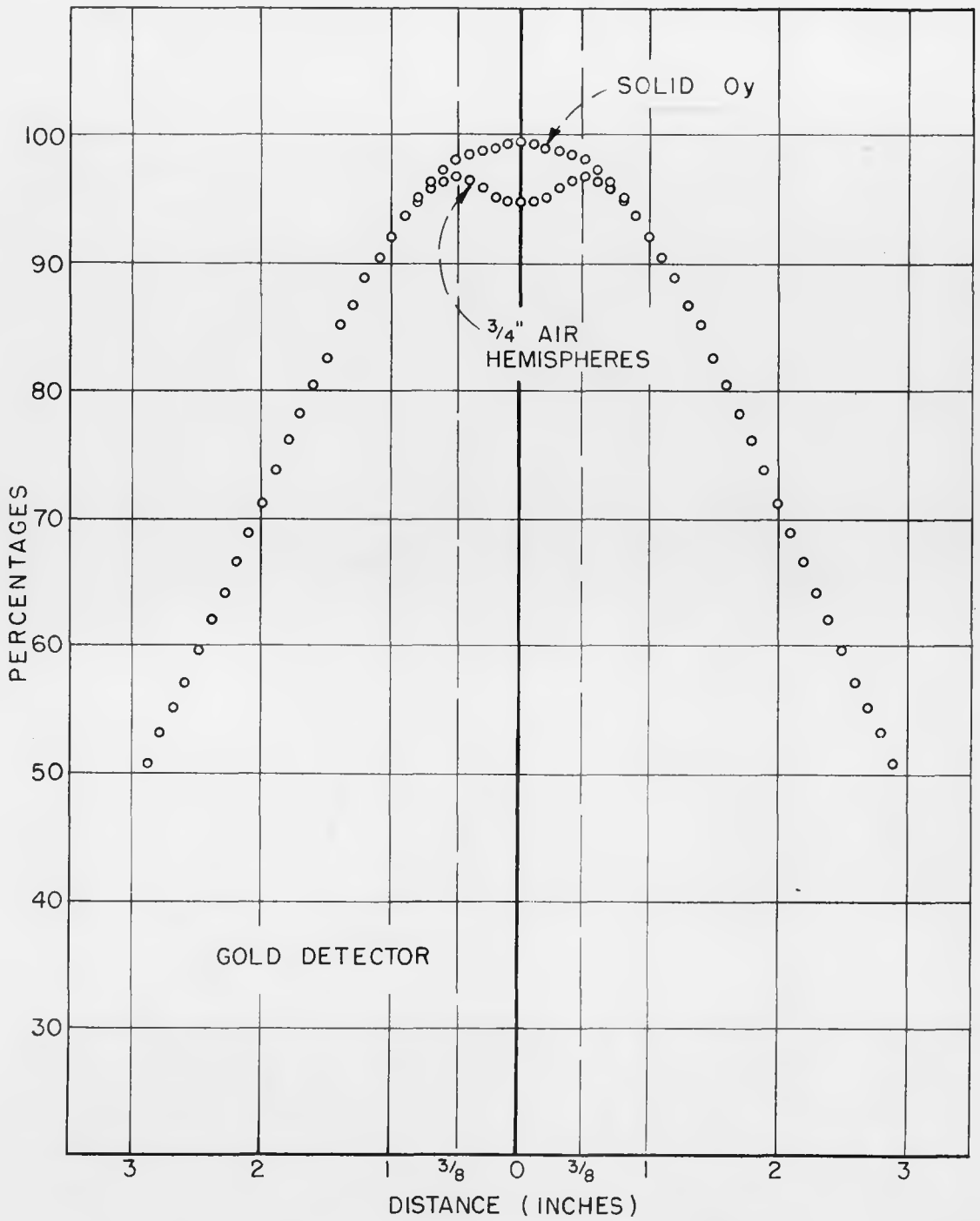


FIG. 7.

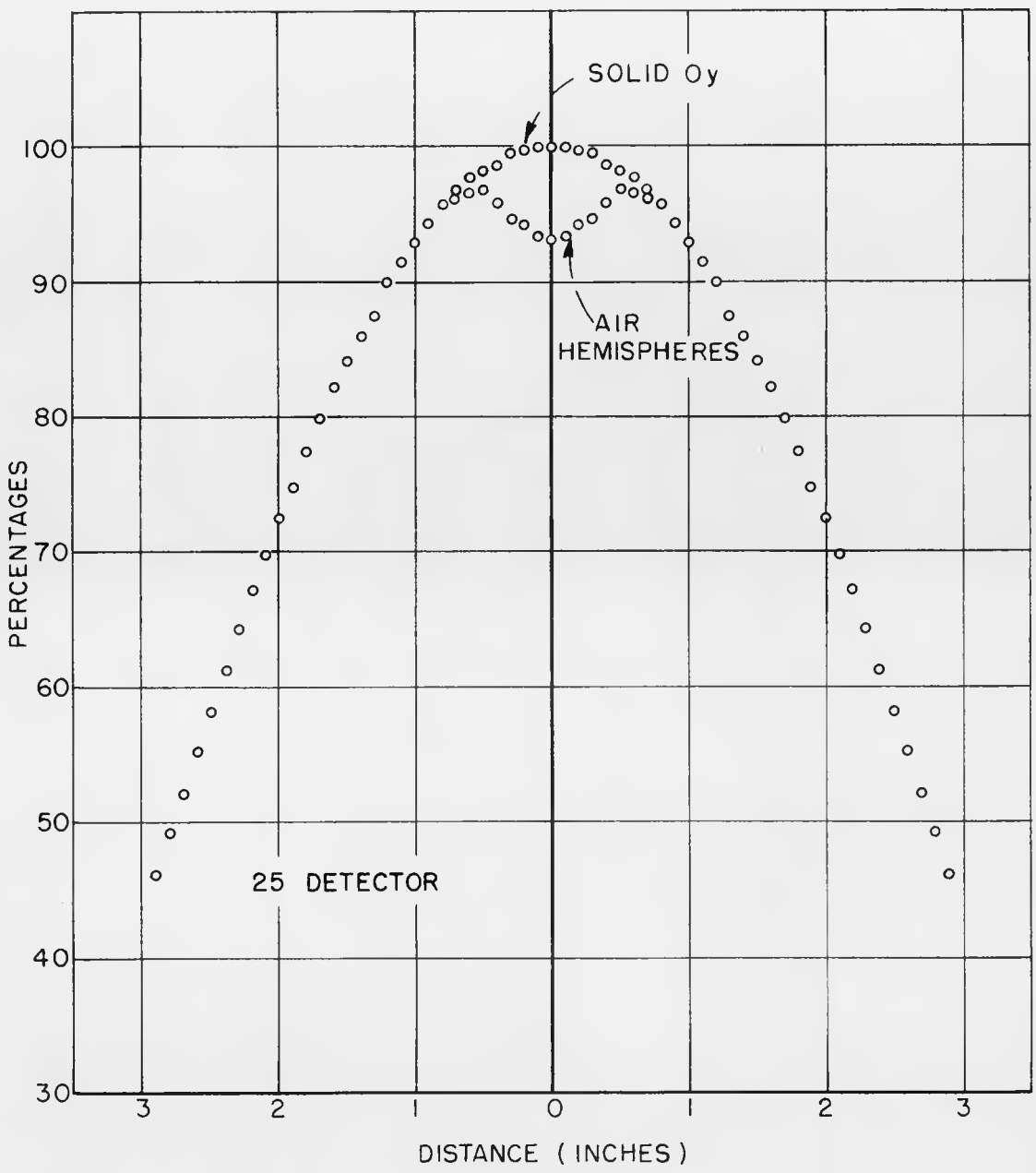


FIG. 8.

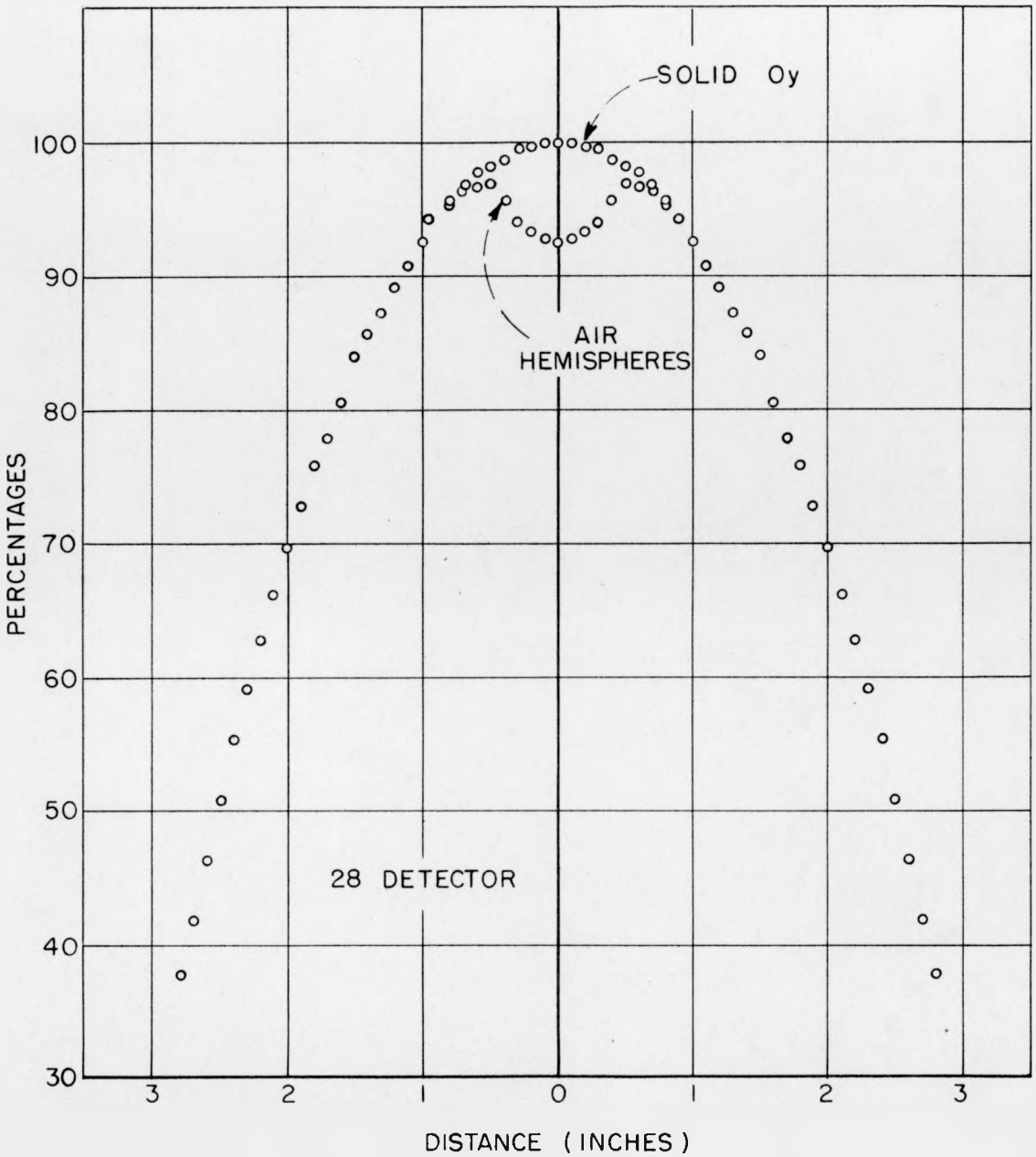


FIG. 9.

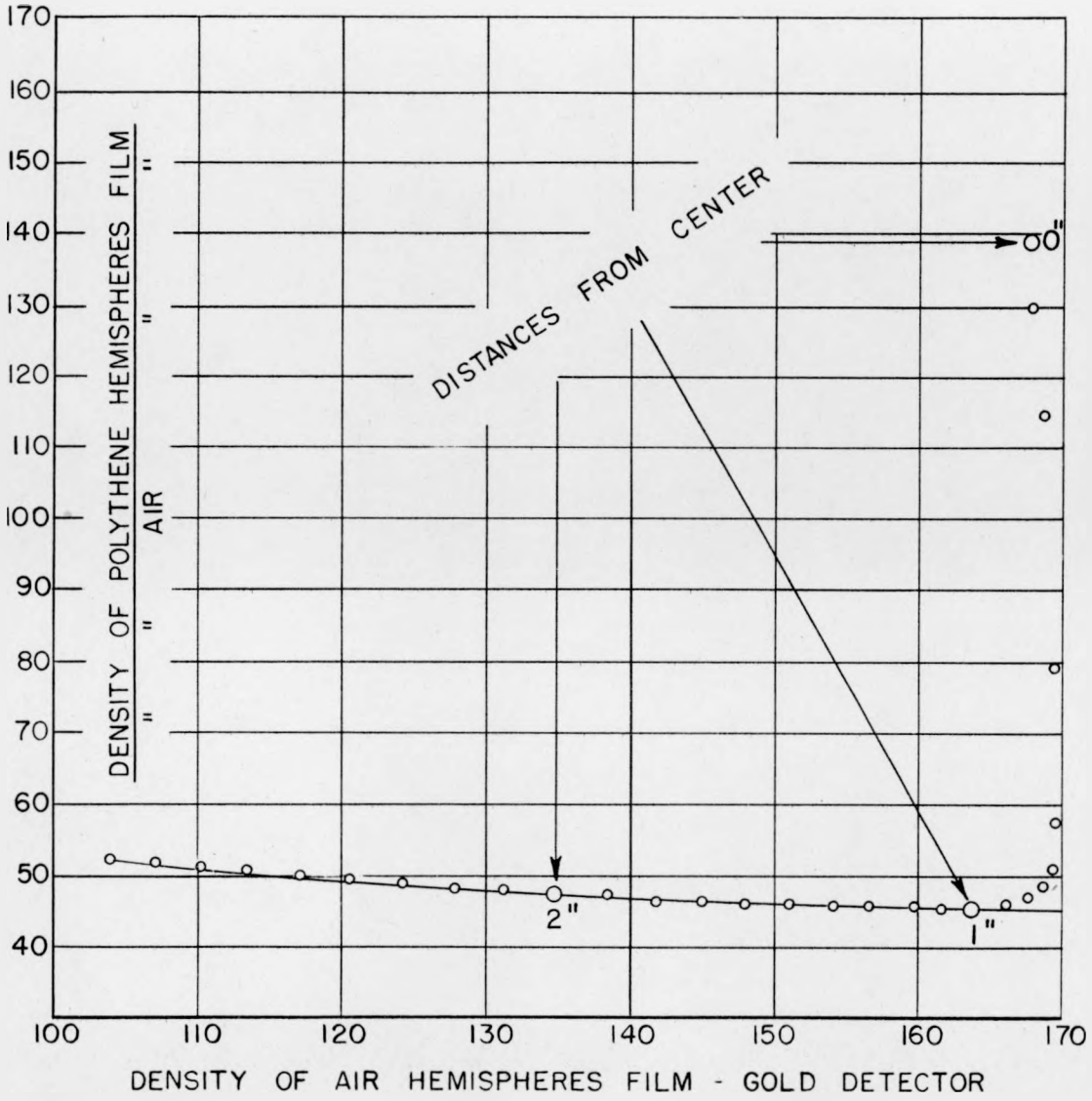


FIG. 10.

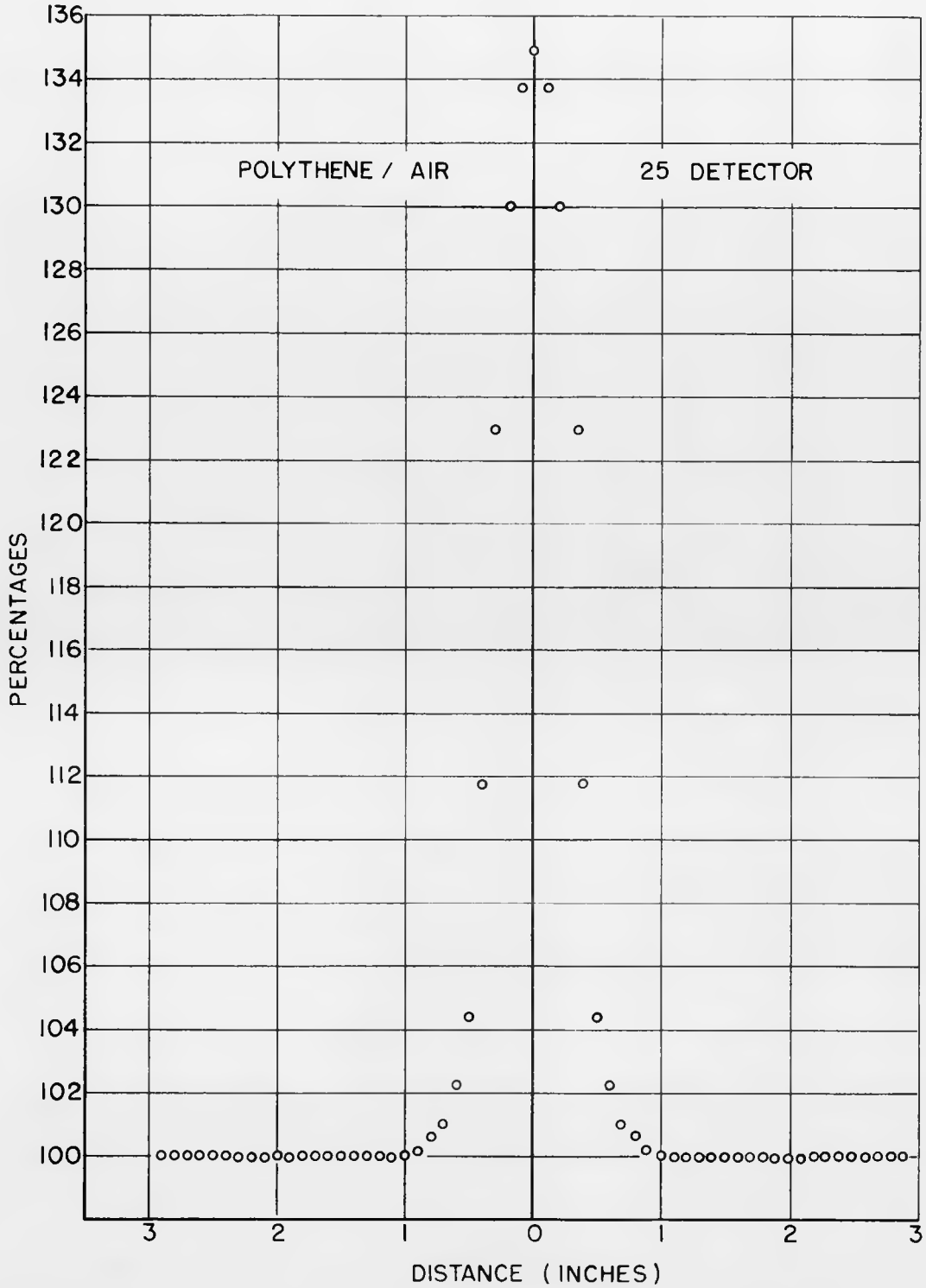
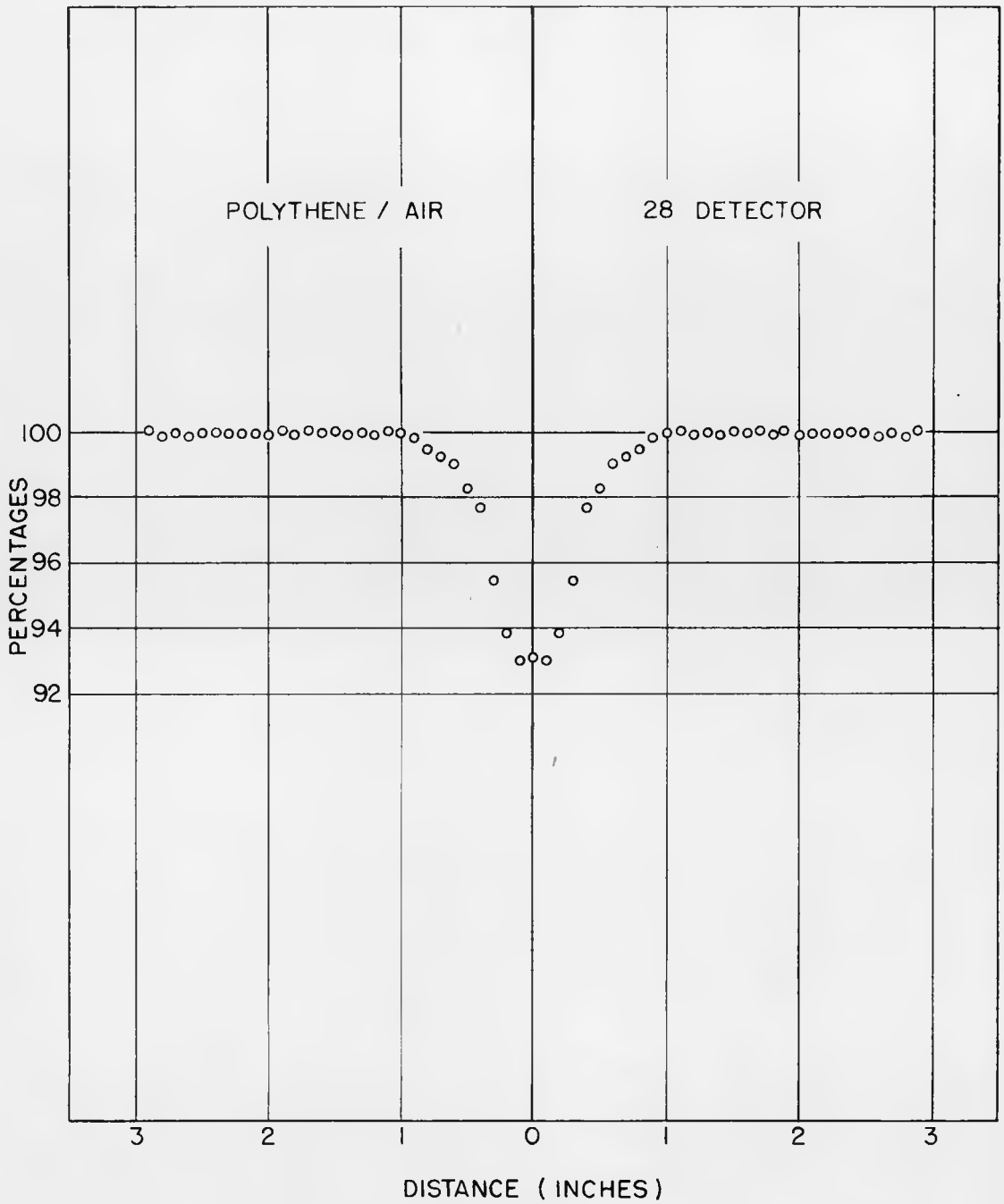


FIG. 11.



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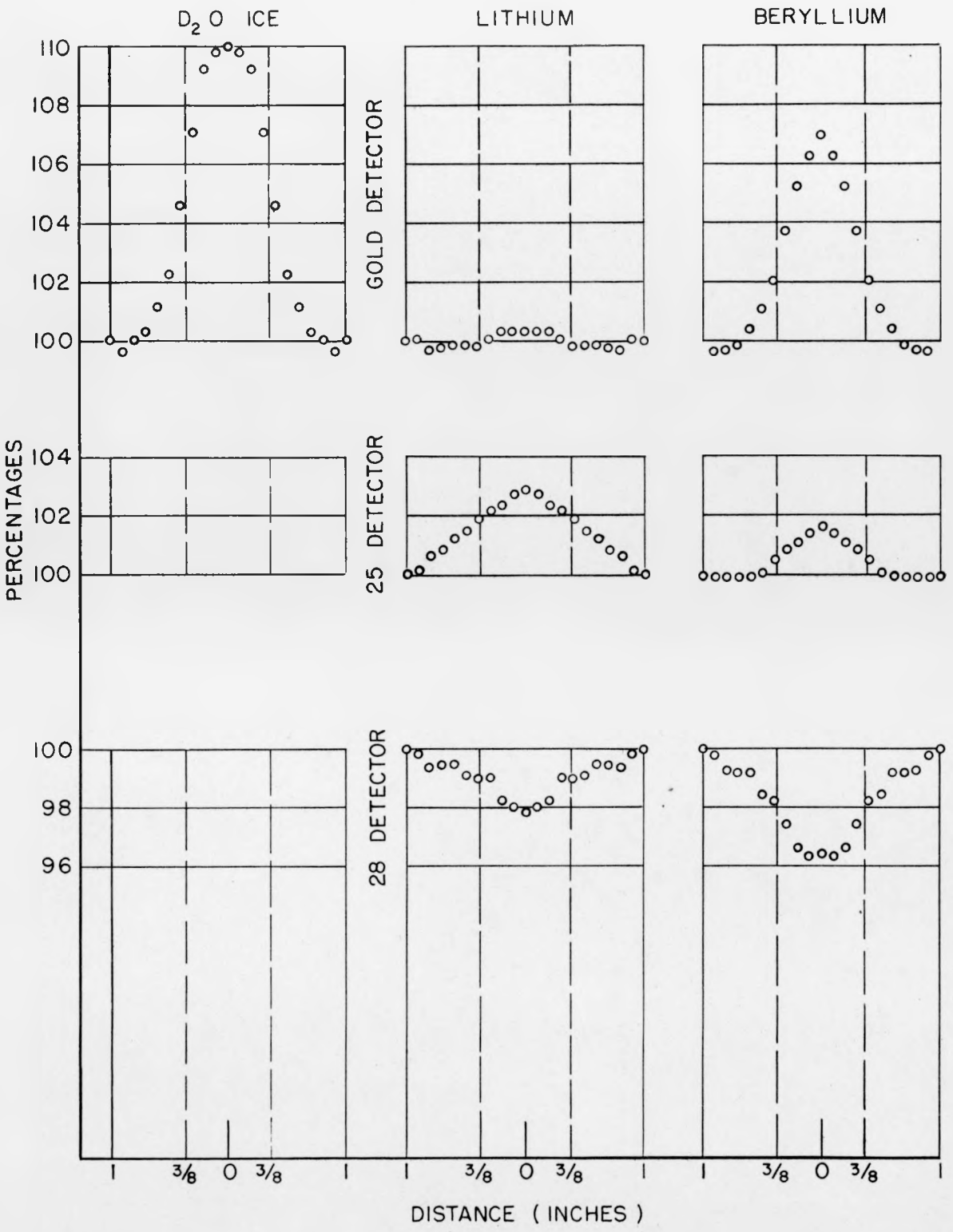

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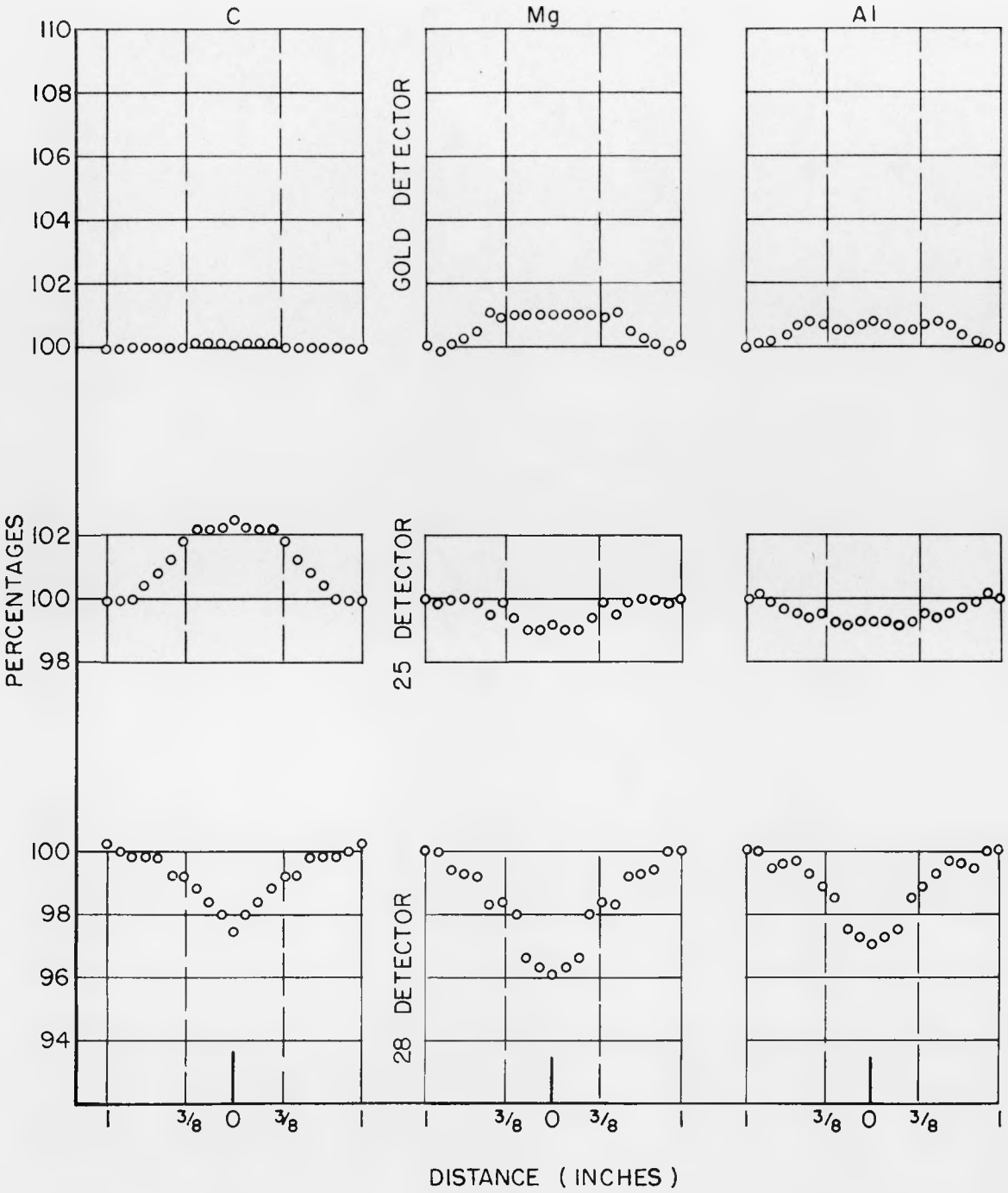

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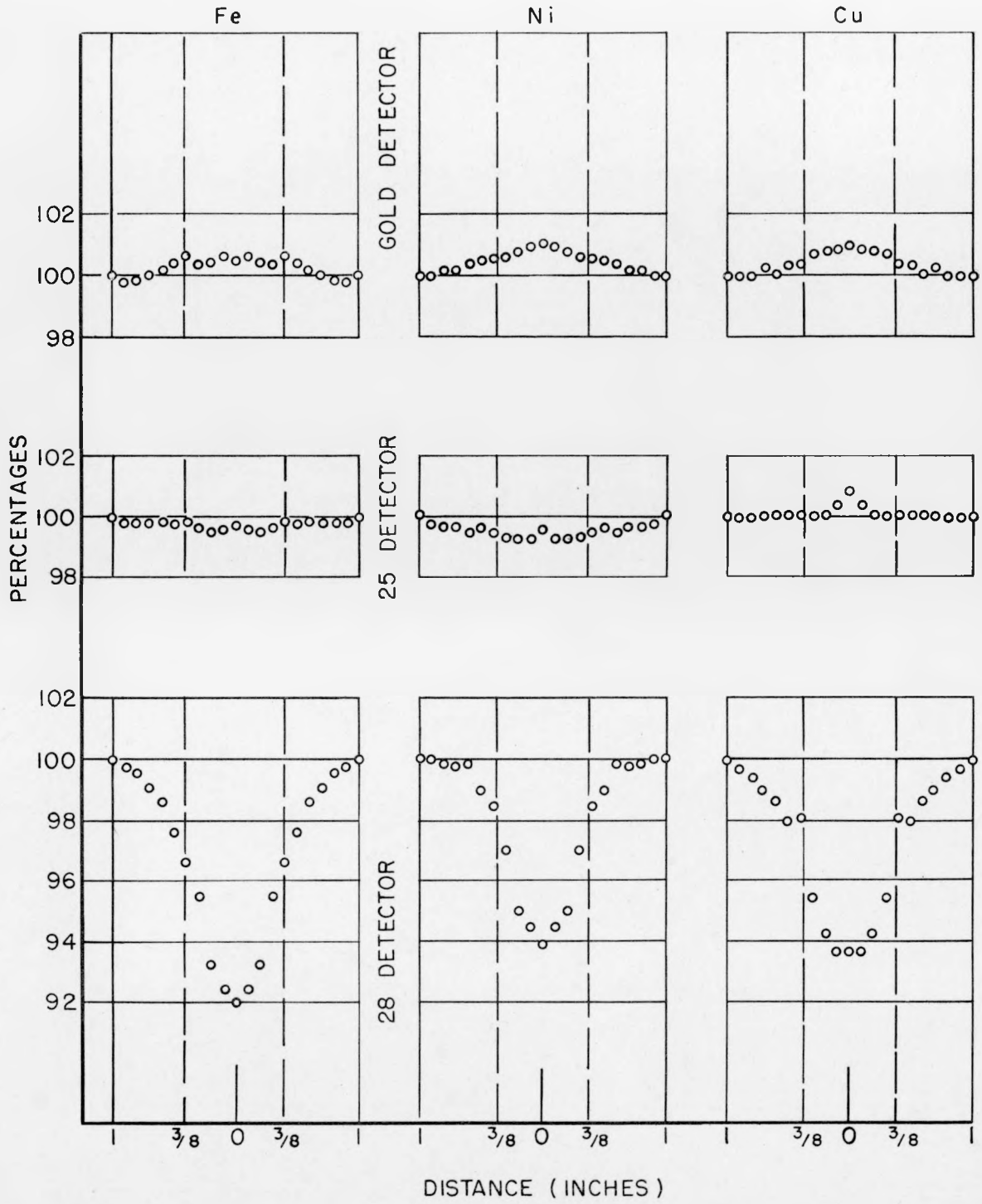

FIG. 15.

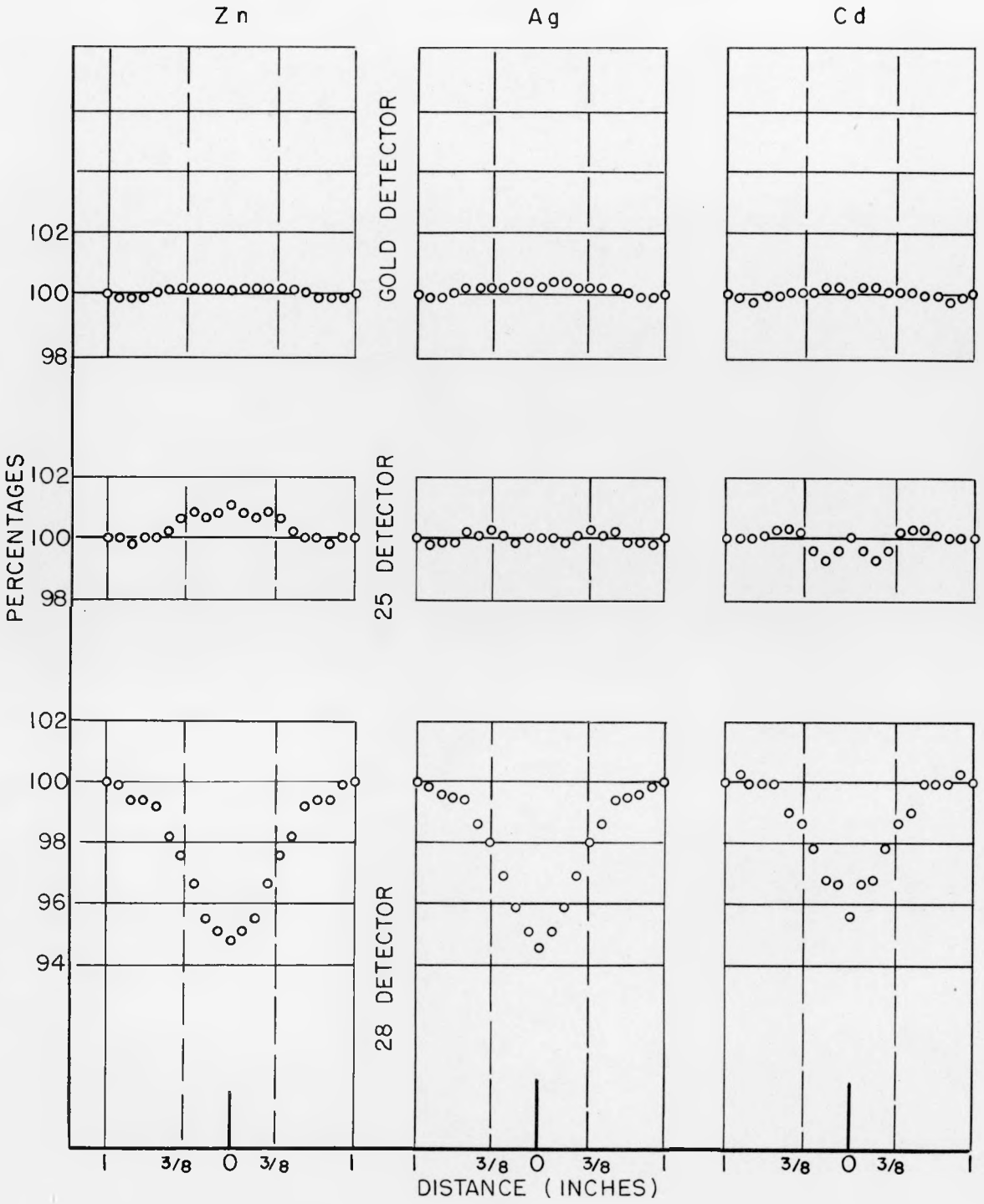

FIG. 16.

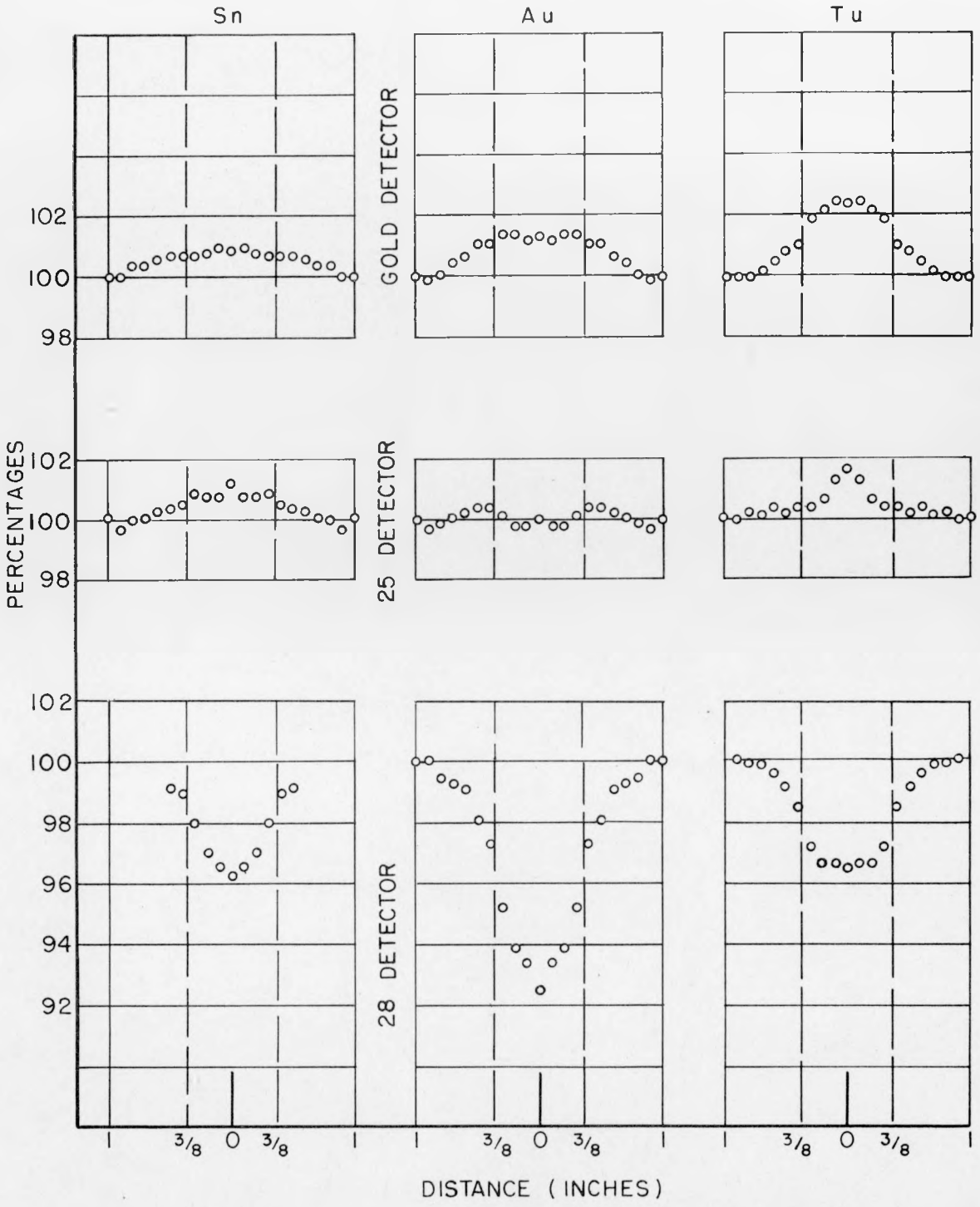

FIG. 17.