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NEUTRON RADIOGRAPHY - A SECOND PROGRESS REPORT\*<sup>1</sup>

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# NEUTRON RADIOGRAPHY - A SECOND PROGRESS REPORT<sup>1</sup>

By

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## INTRODUCTION

Because relatively little has been published previously on the subject of neutron radiography,<sup>2,3,4,5</sup> it seems appropriate to begin this discussion by restating the reasons for the interest in this potentially useful method for non-destructive testing. This *raison d'etre* for neutron radiography comes about because the relative neutron absorption of materials is different from the relative absorption of these materials for X-rays. Mass absorption coefficients of the elements for thermal neutrons present a random picture if they are examined in terms of regularly increasing atomic number, whereas the mass absorption coefficients for X-rays increase with some regularity if they are similarly examined.<sup>4,5</sup> If there is any pattern at all to this examination of thermal neutron mass absorption coefficients versus atomic number, it is that light elements have high absorption and heavy elements have low absorption for thermal neutrons. This reversal of absorption characteristics of materials for thermal neutrons as compared to X-rays, and the overall random pattern of thermal neutron absorption combine to make neutron radiography a potentially useful technique for nondestructive testing. Because of these absorption differences, neutron radiography should be useful in many inspection problems which present difficulties by X-ray techniques.

These previous statements have specified that neutrons in the thermal energy region have the absorption characteristics which make neutron radiography attractive. Actually many of these absorption differences continue, for increasing neutron energy, through the epithermal and slow neutron energy ranges. In the fast neutron energy region, however, most of these differences in neutron absorption have appreciably diminished, making the use of fast neutrons relatively un-

attractive for general radiographic applications. This present discussion, with only a few exceptions, will be limited to the use of thermal neutrons.

One other limitation to this report is that only photographic methods of detecting the neutron image will be discussed. The reader desiring further information on other methods is referred to brief reviews of nonphotographic neutron image detection methods in the literature.<sup>1,6</sup> Since photographic emulsions alone are influenced very little by neutrons,<sup>7</sup> intermediate materials which emit some photographically detectable radiation upon neutron bombardment are frequently used to intensify the photographic response of film to neutrons.

Two different exposure techniques can be used. In one, the direct exposure method, the photographic film and converter material are exposed to the neutron beam together. In the other, the transfer method, only a screen is exposed to the neutron beam. This radioactive, image-carrying screen is then placed next to photographic film after the neutron exposure is completed. The film is exposed by the radioactive decay radiation of the screen.

This discussion will be concerned with a comparison<sup>8</sup> between many of the useful converter materials primarily in regard to photographic speed and image sharpness qualities. Explanations for some of the observations made concerning these imaging methods will be offered.

#### EXPERIMENTAL PROCEDURES

##### A. The Neutron Source

A beam of neutrons which contains a very small gamma intensity is one which would be most useful for neutron radiography. This follows from the fact that neutron radiography is of interest because the absorption of neutrons in materials is different from that of X-radiation. Therefore a gamma radiographic image superimposed on a neutron radiographic image would be expected to lessen the usefulness of the neutron radiograph. It follows, then, that a neutron source having

a high yield of thermal neutrons and a low yield of gamma radiation is desirable for neutron radiography.

A source of thermal neutrons having this feature has been available for the study reported here, and has been used for most of the data which will be given here. This neutron beam is taken from the neutron spectrometer used for neutron diffraction work<sup>9</sup> at Argonne's CP-5 reactor. A schematic diagram is given in Figure 1.

The monochromatic beam from the monochromatizing crystal emerges through the defining slits and the beam monitor toward the diffraction sample. The portion of the neutron beam which goes through, or around the diffraction sample enters a beam catcher some distance away. It is this beam, normally unused, which has been employed for this study.

By the time the beam reaches the beam catcher, the relatively low contrast image of the diffraction sample is usually not detectable. The beam is somewhat oval shaped and has the approximate dimensions of 2 1/2" x 3 1/2". A central portion about 1/2" x 1" contains the maximum beam intensity. At the beam catcher location, this maximum intensity has been determined to be  $3 \times 10^5$  n/cm<sup>2</sup>-sec. by means of a gold foil activation method. The neutron wavelength used for the majority of these tests was 1.05 Å.

The gamma intensity in the beam is very low as far as its photographic effect is concerned. This has been confirmed by the facts that direct neutron exposures of very light objects could easily be obtained and that exposures of KK X-ray film to the beam filtered by 1/4" boral sheet yielded no detectable film darkening with exposures as long as 20 minutes.<sup>8,10</sup> By comparison, direct neutron exposures using KK film were normally obtained in the order of 4 minutes or less.

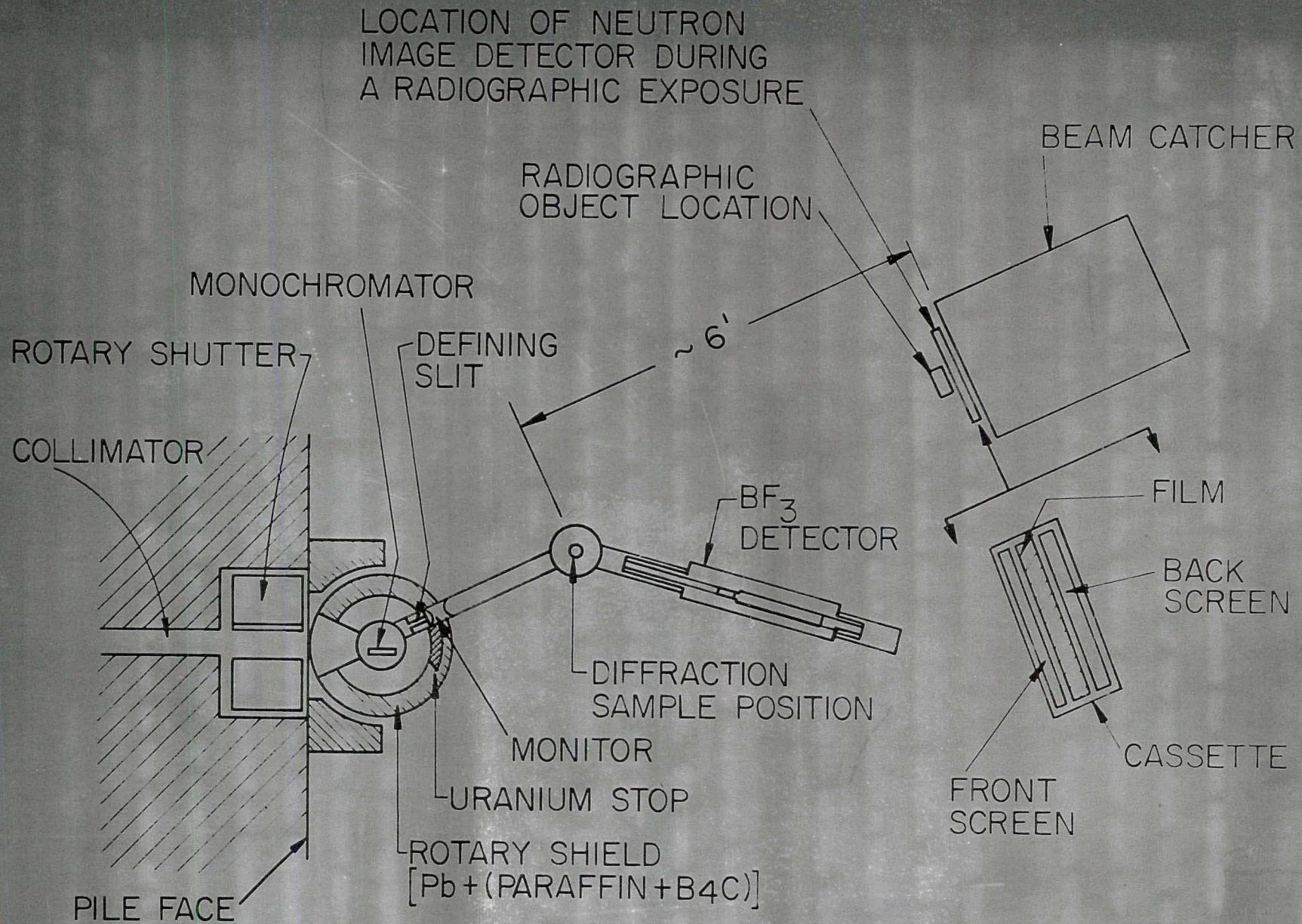


FIGURE 1

SCHEMATIC DRAWING OF NEUTRON SPECTROMETER SHOWING ARRANGEMENT USED FOR NEUTRON RADIOGRAPHY

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### B. Photographic Detection Methods

Although standard photographic film alone can be used to detect neutrons,<sup>7</sup> much faster, and in many ways more satisfactory detection of a neutron image can be accomplished by using various converter materials in conjunction with photographic emulsions.<sup>8</sup> The converter materials, upon neutron bombardment, emit some photographically effective radiation. Typical reactions include  $B^{10}(n,\alpha)Li^{7}$ ,  $Cd^{113}(n,\gamma) Cd^{114}$  and many reactions which result in radioactive materials (materials such as rhodium, silver, indium and gold are commonly used). A more complete description of many of the useful neutron image detection reactions has been given elsewhere.<sup>1,8,10</sup>

For the metal screens used in the direct exposure method, it should be pointed out that, whether the screens become radioactive or not, a considerable portion of the film exposure appears to be caused by prompt  $(n,\gamma)$  radiation from the screen material.<sup>8</sup> This, along with the high cross sections for many of the very short half-life activities,<sup>1</sup> helps account for the fact that most of the film exposure occurs during and very shortly after the neutron exposure, at least for the exposure conditions used for these tests.

One other comment concerning direct exposure materials is that it has been found much more effective to combine those materials (such as B-10 and Li-6) which emit alpha radiation upon neutron bombardment, with a phosphor so that light rather than alpha radiation blackens the film.<sup>2</sup> Three such scintillators have been studied in this investigation. One is a B-10 enriched (92%) boron polyester type containing  $ZnS(Ag)^{11}$  such as that described by Sun.<sup>12</sup> The other two are  $LiF-ZnS(Ag)$  mixtures such as the one described by Stedman<sup>13</sup> and the similar one employing the modifications made by Shull.<sup>14</sup>

In the direct exposure procedure normally used in this investigation, the converter screen and the photographic film were placed in an aluminum front,

spring-loaded X-ray cassette. The object being radiographed was attached to the cassette front and the combination was then placed in the exposure position<sup>15</sup> shown in Figure 1. Exposure times were controlled using a manually operated boral sheet shutter.

In the transfer method procedure used, the metal screen which would eventually carry the radioactive image, was normally supported and exposed within the aluminum front cassette. After the neutron exposure was completed, the foil was transferred to a film loaded cassette in order to complete the photographic exposure. The transfer of the radioactive screen to film could usually be accomplished within 30 seconds of the end of the neutron exposure of the screen.

For the most part, commercially available, double emulsion X-ray films were used. Development was done in Kodak Liquid X-Ray Developer ( $68^{\circ}\text{F} \pm 1/2^{\circ}\text{F}$ ) for 5 minutes, without agitation. Density measurements were made using a MacBeth Ansco Densitometer (Model 12A) having a density range of 0 to 6.0.

#### EXPERIMENTAL RESULTS

##### A. Direct Exposure Method

A study has previously been made to determine the thicknesses of metal screens which yield the best neutron photographic speed for each of several metals in a double screen technique.<sup>1,8</sup> In this method, the film is sandwiched between two metal screens during the neutron exposure. The metals studied in this manner, and the resultant thicknesses of metal screens which yielded the best neutron photographic speed for a given neutron exposure are given below. For cadmium the best screen combination was 10-20 where the numbers are the screen thicknesses in mils, of the screen closest to the neutron source and the back screen respectively. Using similar notation, best speed results were obtained for rhodium using 10-10 screens, indium with 20-30 screens and silver with 18-18 screens. In each of these cases, the screen thicknesses were chosen at the point where further increases in screen thickness produced little or no speed improvement.

This work has now been expanded to determine the thickness of a single screen, and the photographic film orientation to use for best neutron photographic results. In most cases the use of a single metal screen rather than a double screen does involve a sacrifice of about a factor of two in speed but, for many applications, the improvement in image sharpness justifies this sacrifice. An overall comparison of photographic speed for the materials studied is given in Table I. The double screen method using cadmium screens and type KK film has arbitrarily been rated 1.0.<sup>16</sup> It should be emphasized that this speed information is strictly true only for the exposure conditions used here (2 minutes exposure for double screens and 4 minutes for single screens at a neutron intensity of  $3 \times 10^5$  thermal n/cm<sup>2</sup>-sec). For other neutron intensities and exposure times, speed variations would occur as a result of reciprocity law failures with the scintillator and because of the different times involved for saturation activity with the radioactive screens. These speed values may be used as guides, however over a rather broad range of exposure conditions.<sup>17</sup>

A few comments about each of these screen materials, including the reasons for the choice of the single screen thickness and film placement shown in Table I, are given below. The image sharpness investigations are still in progress so that such information given in regard to these materials should be considered as preliminary.

#### Scintillators

The B-10 loaded scintillator<sup>11,12</sup> has yielded very fast detection results with type F X-ray film. The sharpness of the radiographs obtained using this material has been very good and in fact, among the direct exposure methods it is second only to that obtained with gadolinium screens. The major drawback to a definite recommendation for this detection method is that the radiographs obtained with the scintillator have a mottled background which appears to be caused by

TABLE I

RELATIVE PHOTOGRAPHIC SPEED FOR SEVERAL DIRECT EXPOSURE NEUTRON IMAGE DETECTION METHODS

Converter Material and Screen Configuration <sup>b</sup>	Film Type	Relative Speed <sup>a</sup>
B-10 Loaded Scintillator as back screen	F	26
Double Rhodium <sup>c</sup> screens (10-10)	KK	1.4
Double Gadolinium <sup>d</sup> screens (0.5-2)	KK	1.1
Double Indium <sup>c</sup> screens (20-30)	KK	1.1
Double Cadmium screens (10-20)	KK	1.0
Double Silver <sup>c</sup> screens (18-18)	KK	0.8
Single Gadolinium <sup>d</sup> as back screen (2)	KK	0.75
Single Cadmium as back screen (10)	KK	0.67
Single Rhodium <sup>c</sup> as back screen (10)	KK	0.62
Single Indium <sup>c</sup> as front screen (20)	KK	0.5
Single Silver <sup>c</sup> as front screen (15)	KK	0.35

- (a) The relative photographic speed was obtained by comparing film densities and relative exposures for each detector subjected to a similar neutron exposure. The neutron intensity used was  $3 \times 10^5$  n/cm<sup>2</sup>-sec.
- (b) The numbers given with the metal screens refer to the thickness in thousandths of an inch, of the front and back screens, respectively.
- (c) Radioactive screen materials were permitted to decay on the film for three half-lives or more after the neutron exposure was completed. For most of these materials (except indium) this extra transfer time produced little change in film exposure (see reference 1).
- (d) The gadolinium screens available for this study have been very small (see Figure 10) until recently, when 5" x 7" screens were received. These speed numbers should be regarded as tentative until further data can be obtained with these larger screens. The screen thicknesses given appear about optimum for speed, based on information from these initial tests.

non-uniform distribution of the ZnS(Ag) phosphor. A picture demonstrating this is shown in Figure 2.

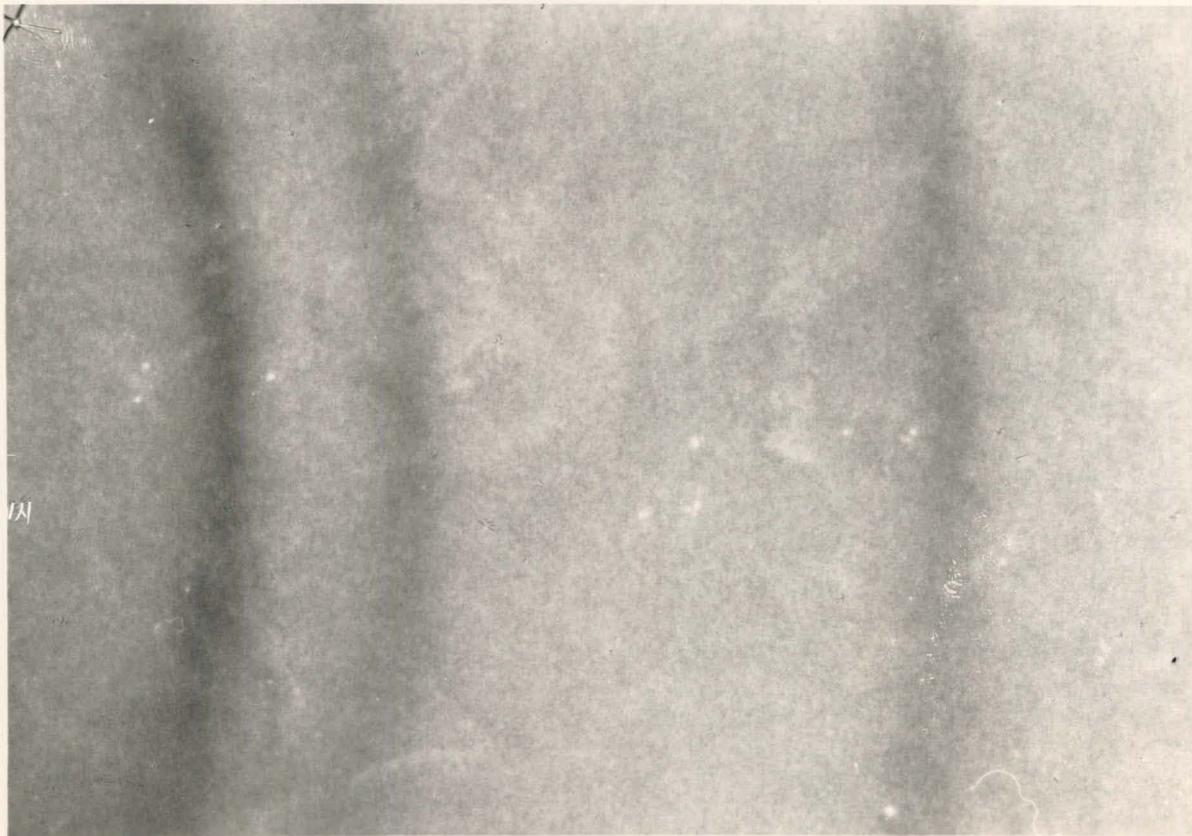
The other scintillators tested have been combinations of LiF and ZnS(Ag). Tests have been made using the scintillators both in the form described by Stedman<sup>13</sup> (LiF, ZnS(Ag) and lucite powder moulded into buttons) and in the form used by Shull<sup>14</sup> (LiF and ZnS(Ag) pressed into a metal form). Using natural LiF and type F film in both cases a relative speed rating of about 8 (on the same basis as the values of Table I) was found in these tests for the Stedman type buttons and about 17 for the 4:1 ZnS(Ag) - LiF mixtures.<sup>18</sup> Comparable speed values were found using Polaroid type 57 film (3000 speed). Pictures produced by these scintillators were relatively grainy. In all cases the scintillators have been used as back screens during the neutron exposures. On the basis of these preliminary data on the lithium scintillators, it seems reasonable to point out that, with Li-6 enriched material, scintillators of the type described by Shull and either type F X-ray film or Polaroid type 57 film would yield the fastest response to thermal neutrons, of any of the methods tested in this study thus far.

#### Rhodium

Rhodium screens have yielded radiographs having the third best sharpness (behind gadolinium and the scintillators) of the direct exposure methods. For a single screen the best film density for a given neutron exposure occurs for a screen thickness of about 0.010", as shown in Figure 3. As with most of the single metal screen methods the best photographic film density is obtained for back films (films used on the side of the screen opposite to the neutron source) while best sharpness qualities are found for front films (photographic film on the same side as the neutron source). In the preliminary image sharpness studies made thus far, little difference in image sharpness has been observed for radiographs made with rhodium screens varied from 0.003" to 0.010". Therefore, for

FIGURE 2

A reproduction of a neutron diffraction pattern obtained from a powdered nickel sample using a B-10 loaded scintillator and Type F X-ray film in a flat cassette. Typical exposure for such a picture is about 1 hour. The cassette was approximately 15" from a relatively thick sample (7/16" dia.). The mottled picture background is due to the scintillator. The lines visible on this picture are the 111, 200 and 220 lines of nickel respectively, starting from the left.

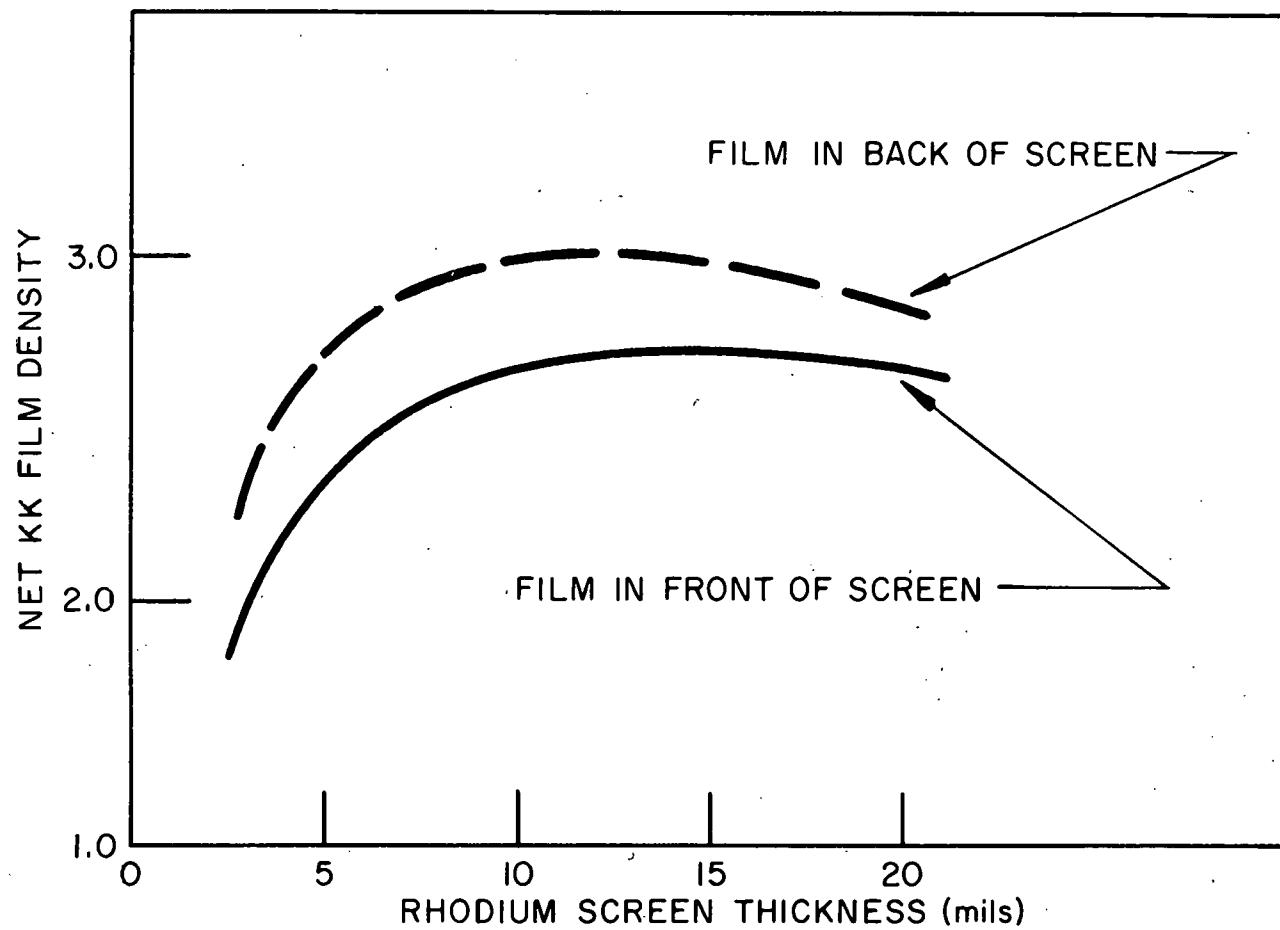


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FIGURE 3



Determination of single rhodium converter screen thickness and film location for best neutron photographic speed by the direct exposure method. The information shown was obtained by exposing different screen thicknesses and film to the neutron beam for a constant neutron exposure, and comparing the resultant film densities produced. In the case of the radioactive screen-materials (shown in this figure and in figures 4 and 6) a three half life decay period after the neutron exposure was given before the film and screen were separated. Films exposed on the neutron source side of the converter screens are called front films, those on the other side are called back films.

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rhodium, a 0.010" screen and a front film would be recommended because of the improved speed over the thinner screens and because of the improved sharpness for the front film.

Silver

The thickness of a single silver converter screen which will yield the best film density for a given neutron exposure is approximately 0.015" as shown in Figure 4. Little difference in image sharpness qualities versus screen thickness has been observed for silver over the range of 0.005" to 0.020". As in the case for rhodium, best film exposure was found for back films. For silver screens however, the relatively low neutron photographic speed, particularly for front films, may help account for the fact that little difference in sharpness was observed between front and back films. It is thought that the increased influence of secondary and scattered radiation on the film during the long exposures needed for front films may have had some affect on this result. This was definitely true for the case of the prompt  $(n, \gamma)$  radiation from the cadmium test object (see Figure 10) used for the sharpness studies in that more film exposure from this radiation was found for front film exposures than for back film exposures. The choice for silver was therefore a 0.015" thickness screen and a back film.

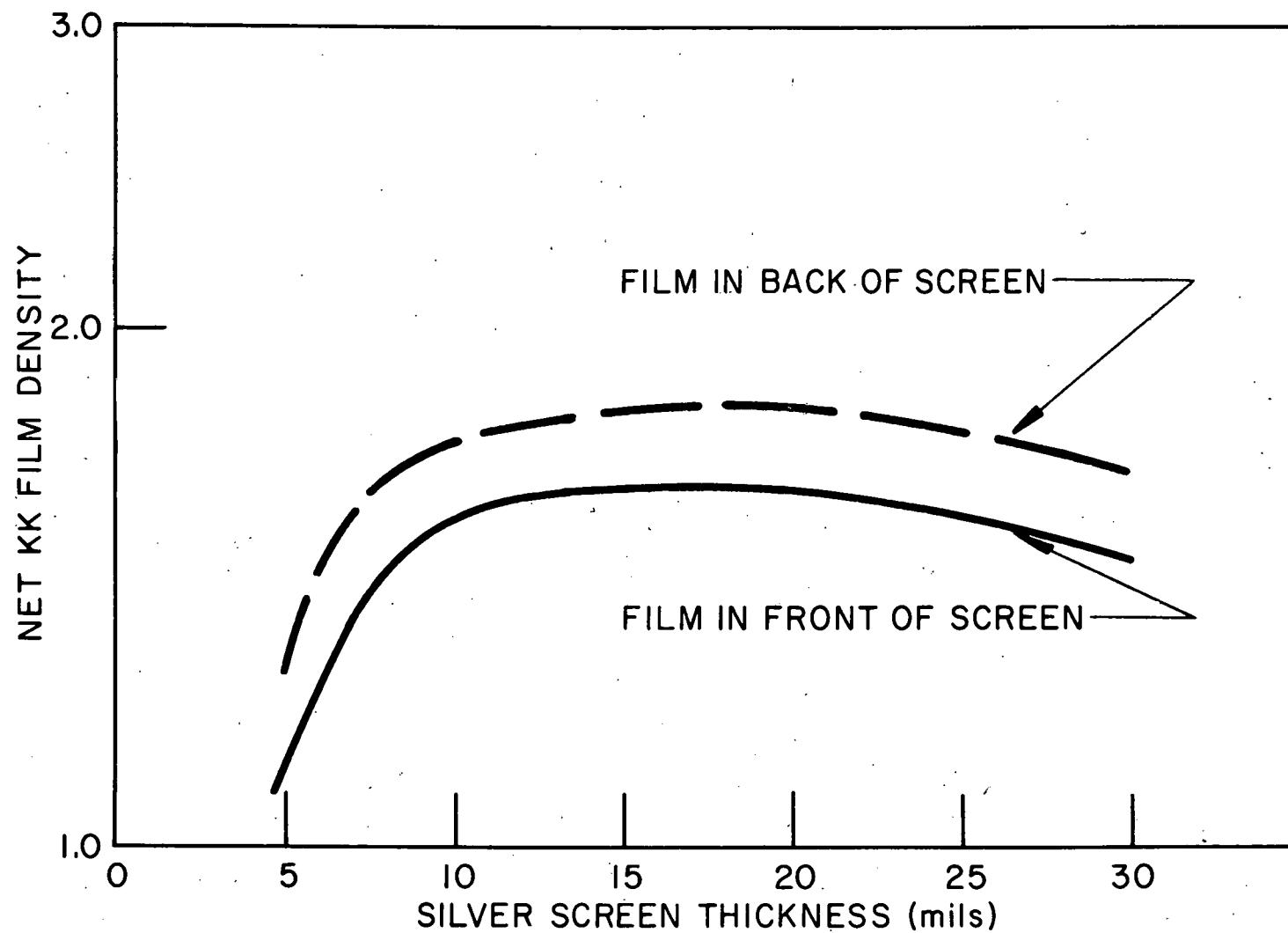
Cadmium

The choice of the most useful cadmium screen and film placement was dictated by the facts that front films yielded much better sharpness than did back films and that this sharpness was relatively independent of cadmium thickness over the range 0.001" to 0.010". The combination of good density and sharpness yielded by the 0.010" screen (see Figure 5) and a front film was the choice for cadmium.

Indium

The sharpness found with indium screens appeared to be best for back films (perhaps for the reasons noted for silver exposures) and was relatively independent of screen thicknesses from 0.005 to 0.020". Since a 0.020" indium screen

FIGURE 4



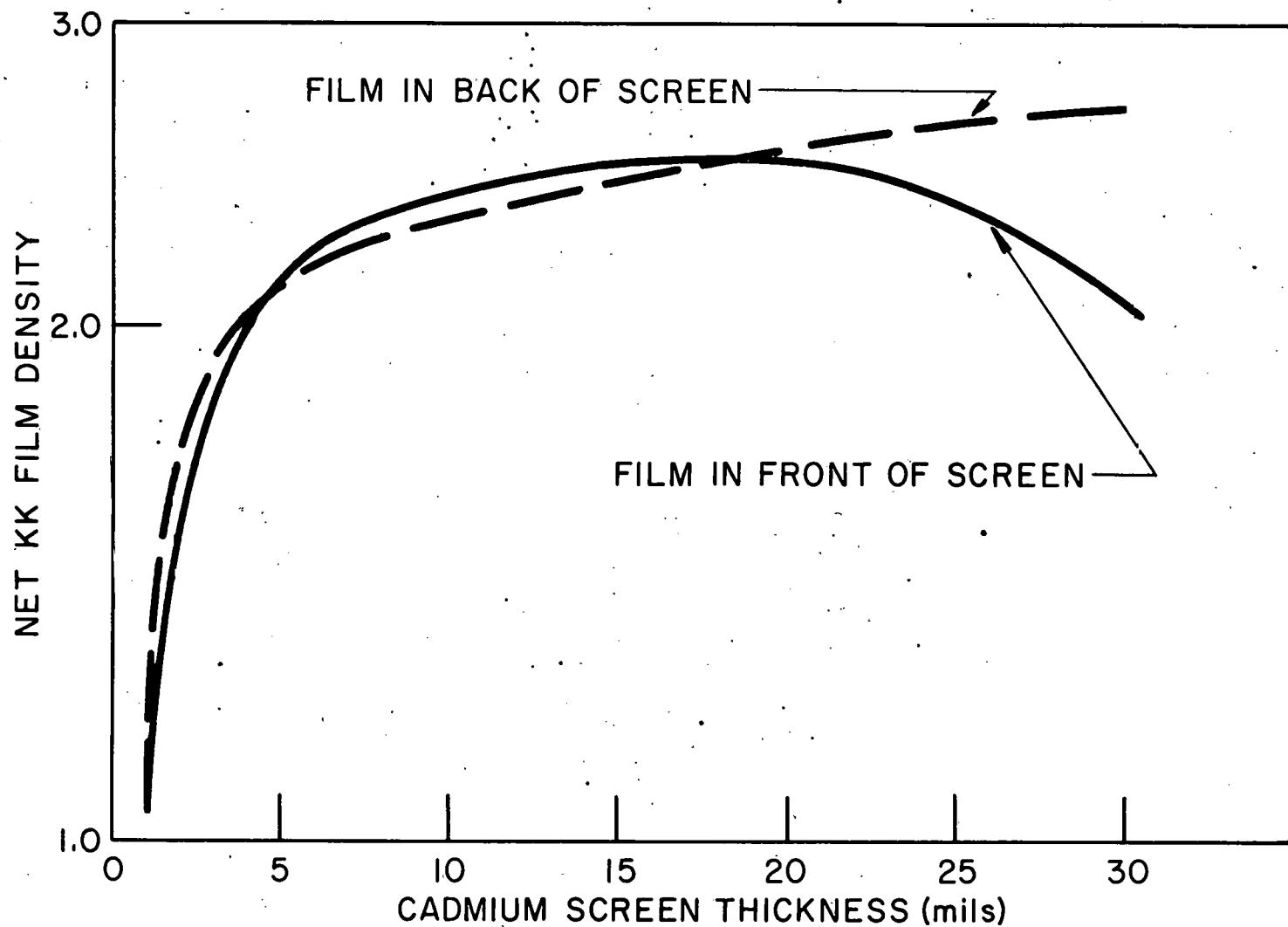
Determination of single silver converter screen thickness and film location for best neutron photographic speed by the direct exposure method. See caption on Figure 3 for further details on this plot.

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FIGURE 5



Determination of single cadmium converter screen thickness and film location for best neutron photographic speed by the direct exposure method. See caption on Figure 3 for further details on this plot.

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and a back film also produced about the best film density as shown by Figure 6, this was the choice for indium direct exposures.

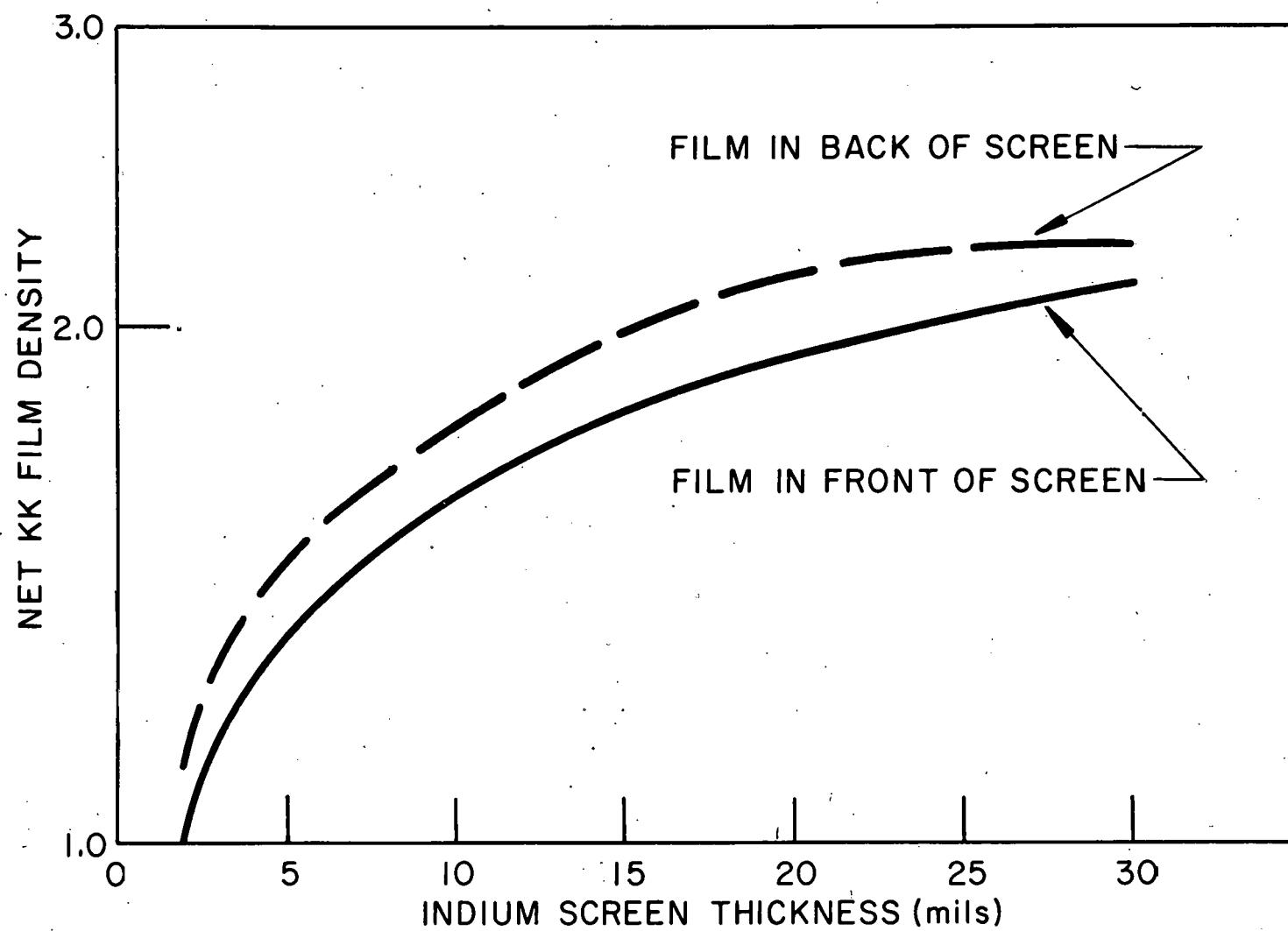
Gadolinium

Since useful size gadolinium screens have just recently been available for these tests, the gadolinium data must be regarded as preliminary. Without question however, it can be stated that direct exposure neutron radiographs taken using gadolinium screens have the best image sharpness of any of the other methods tested to date, including the transfer methods. Although a detailed study of image sharpness versus screen thickness has not yet been made, there is no obvious indication of sharpness change over the thickness range of 0.00025" to 0.002", for front films. In general, front films do yield appreciably better sharpness than back films. The thickness of gadolinium to use for best speed is in the order of 0.002" from the data shown in Figure 7. This thickness screen, with a front film would be a highly recommended method for direct exposure neutron radiography. It has good speed and excellent sharpness qualities.

Good image sharpness results have also been obtained for the double gadolinium screen method. Almost equal speed results have been found for this method for both the 0.25-2 and 0.5-2 gadolinium double screen combinations. As the front screen increases in thickness beyond 0.0005", the speed of the double screen method begins to decrease. For the data shown in Table I, the speed of the 0.5-2 gadolinium combination was given.

Our present work with gadolinium involves a more detailed study of the image sharpness and contrast which can be obtained for the use of single gadolinium screens of different thicknesses, for double gadolinium screens, and for a single gadolinium screen used with a lead intensifier screen. (This last technique is further discussed in a following section of this report.) All these methods yield excellent image sharpness and it is hoped that, as a result of this further study, a definite recommendation can be made for the best of these methods.

FIGURE 6



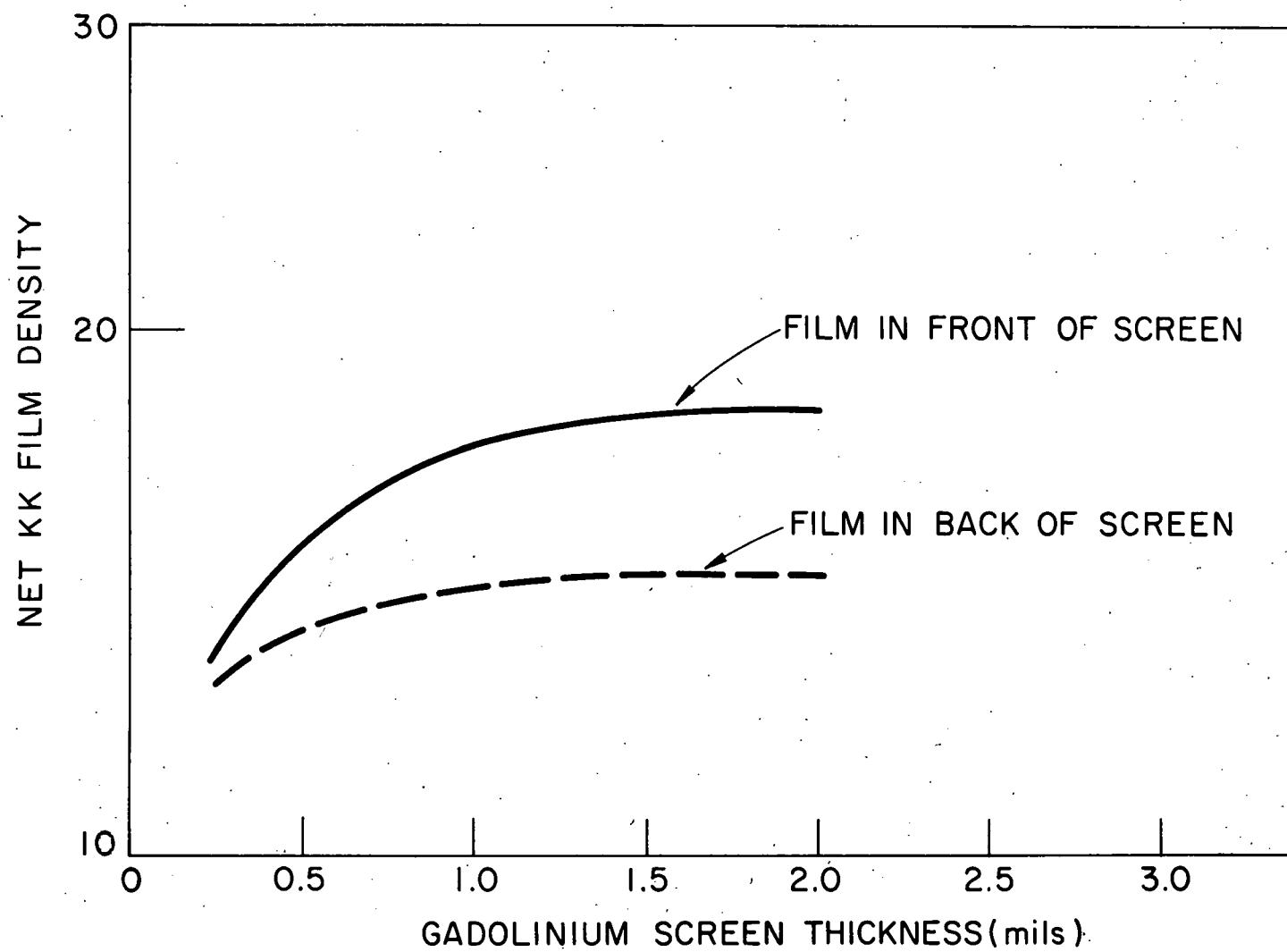
Determination of single indium converter screen thickness and film location for best neutron photographic speed by the direct exposure method. See caption on Figure 3 for further details on this plot.

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FIGURE 7



Determination of single gadolinium converter screen thickness and film location for best neutron photographic speed by the direct exposure method. See caption on Figure 3 for further details on this plot.

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Gold

Because of the low speed of gold direct exposure methods with double screen techniques,<sup>1</sup> this method has not been fully tested for single screen use. Gold, along with cadmium produces direct exposure neutron radiographs which have relatively poor image sharpness. This fact, along with the poor speed, would seem to render gold direct exposures of limited use for neutron radiography, at least with neutron beam intensities of the same order as those used in these tests.

Other Direct Exposure Materials

Other materials which have been tested for direct exposure neutron radiography include loaded emulsions and X-ray film itself. The speed results with both of these methods for neutron image detection were very low.<sup>8</sup> Speed numbers, on the same basis as those of Table I, were 0.03 for type KK X-ray film used alone<sup>10</sup> and less than 0.1 for both lithium and boron loaded emulsions<sup>19</sup> if density was taken as the basis of measurement.

The pictures obtained, particularly with X-ray film were normally of poor quality. The apparent reason for this is that, in the long exposures required, the film also recorded much other radiation including that emitted from the objects under study and from the cassette, as well as recording gamma radiation in the imaging beam.

B. Transfer Exposure Method

Of the converter materials mentioned thus far in this report only indium and gold have yielded useful film blackening for the transfer method in the neutron intensity available. Rhodium and silver, which have reaction cross section values in the same order as those of indium and gold, have not been useful for transfer radiography in this neutron intensity because the short half-lives of the radioactive isotopes of these materials preclude their use with neutron exposures long enough to build up the required radioactivity. In addition to

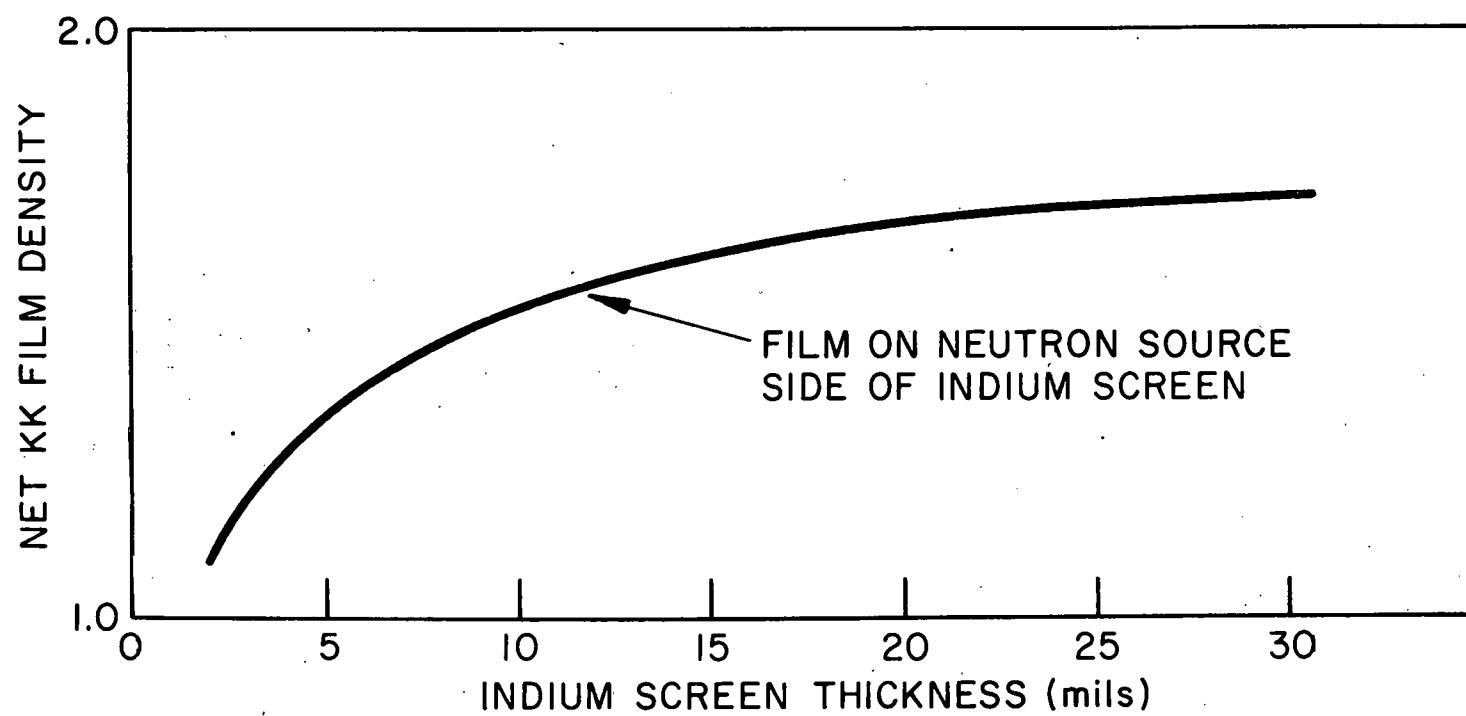
the fact that the influences of gamma radiation in the imaging beam, and of radiation (other than neutrons) which may be emitted from the radiographic object are eliminated, transfer radiographs have the added advantage that they produce sharper pictures than direct exposures (with the noted exception of gadolinium).

Specifically comparing indium and gold, much sharper radiographs can be obtained by the transfer method than by the direct exposure method. Explanations for this result include the facts that some scatter is detected on the film during a direct exposure and that decreased contrast is obtained because the film is present to record prompt radiation emission from the absorbing object under study. A major cause for the change in sharpness however, appears to be the fact that, in a direct exposure, the film is present to record relatively hard prompt  $(n, \gamma)$  radiation from the converter screen itself.<sup>8</sup> In the transfer method, film darkening is accomplished by much softer radiation, thereby yielding improved image sharpness.

Between these two materials used for transfer radiographs, gold seems to yield somewhat better sharpness than indium. This result also may have its explanation in terms of radiation hardness. In each case, much of the film exposure would be caused by beta emission. However there would also be an appreciable photographic effect yielded by the radioactive decay gamma radiation. For In-116 a rather large portion of the total gamma emission occurs for gamma energies greater than 1 MEV. This is not the case for Au-198, in that the energy of the most prominent gamma emission is 0.41 MEV.<sup>20</sup> This difference in gamma energy may explain the improved image sharpness results found for gold transfer methods over those using indium.

For the indium transfer method, Figure 8 shows that the best speed is found for about a 0.020" thickness, a thickness which is also the approximate range of the emitted beta energy. Figure 9 shows a similar result for gold transfers, with

FIGURE 8



Determination of single indium screen thickness for best neutron photographic speed using the transfer exposure method. The information shown was obtained by exposing different thickness indium foils to the neutron beam for a constant neutron exposure, transferring the foils to a film loaded cassette, and comparing the film densities obtained after a three half-life transfer period.

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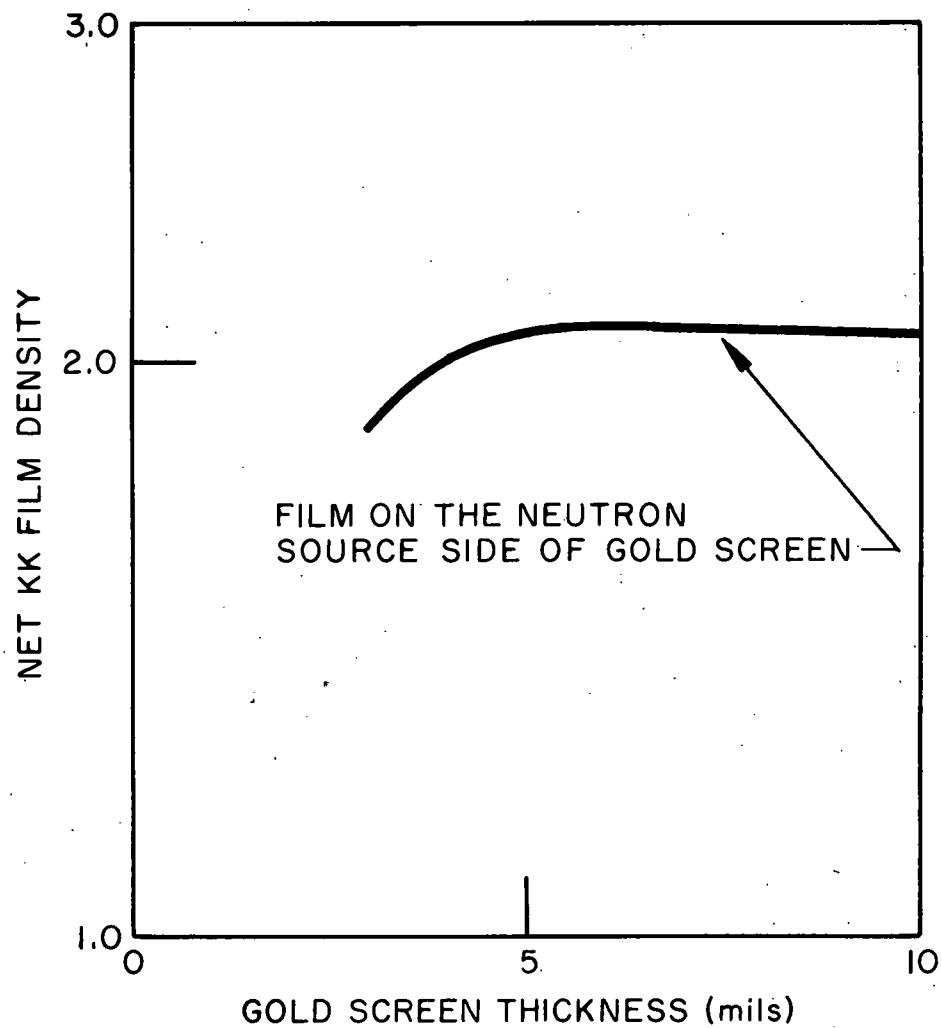


FIGURE 9

Determination of single gold screen thickness for best neutron photographic speed using the transfer exposure method. The information shown was obtained by exposing different thickness gold foils to the neutron beam for a constant neutron exposure, transferring the foils to a film loaded cassette, and comparing the film densities obtained after a three half-life transfer period.

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the best speed occurring for a thickness in the order of 0.005". Both these curves are for front films. The exposure of film on the back side of the screen (not shown) begins to fall off at about the points where these curves level off because of the combined effects of absorption of the neutrons within the screen material and the range of the emitted beta.

The image sharpness data for the transfer radiographs indicate little change in sharpness for transfers of gold in thicknesses of 0.003" and 0.005". The 0.005" screen would therefore be recommended because of its increased speed. The comparable data for indium show little detectable change in image sharpness for screens 0.010" or thinner (down to 0.002"). The 0.010" indium screen then appears to be a useful compromise between speed and image sharpness.

Further comparing these two materials, it should be mentioned that indium, because of its shorter half-life, does yield useful film blackening in less elapsed time than does gold, both in regard to neutron exposure time and to transfer time. However, for similar reasons, indium cannot be used (in neutron intensities such as those used here) with slow, fine grain film such as type M because a saturated activity is reached for a relatively short neutron exposure (about 3 hours). Gold, on the other hand, having a 2.7 day half life can be profitably exposed for the order of a week. Exposures in the order of 2 1/2 days plus a week transfer time have yielded good film densities for gold transfers to type M film.

Both these techniques then appear to be useful methods for neutron radiography. Typical exposure information is being given in another publication.<sup>10</sup>

#### C. Other Neutron Imaging Observations

The purpose of this section of this report is to briefly discuss some other observations made in regard to this work. These include some preliminary trials with other neutron sources and some observations made using X-radiographic intensification methods with neutron converter screen materials.

Our work with other neutron sources includes the use of a one-half curie Ra- $\alpha$ -Be source and a Van de Graaff generator employing the  $\text{Li}^7(\text{p},\text{n})\text{Be}^7$  reaction. The high gamma to neutron ratio output of the radioactive source has made direct exposure methods using this source difficult and the relatively low neutron yield has resulted in very light film exposures for transfer methods even using 3 half life exposures and transfers for gold screens to KK film. Although our results with this particular radioactive source have not been encouraging it seems reasonable that usable neutron radiographs using higher yield sources (such as recently described by Hennelly)<sup>21</sup> could be obtained. This possibility of using radioactive neutron sources for neutron radiography is further discussed by Watts.<sup>6</sup>

The radiographic work with the Van de Graaff generator has been tried here using only the  $\text{Li}^7(\text{p},\text{n})\text{Be}^7$  reaction, employing 2.5 MEV, 10 microamperes proton current and a thick lithium target. In these initial trials, the best, and approximately equal, imaging results were obtained using two different methods. In one, the neutron beam was moderated only by the thick target and airpath, while in the second, the neutron beam was moderated by 2 inches of surrounding paraffin and the beam was brought out through a 1" diameter cylinder which did not "look at" the fast neutron source. The neutron intensity from each of these methods was such that at 12" from the source, exposures approximately 10 times as long as those needed with the monochromatic neutron beam at the reactor were required. The effective neutron intensity at that distance was therefore in the order of  $10^4$  thermal neutrons/cm<sup>2</sup>-sec.<sup>22</sup> The gamma intensity under these conditions was relatively high, and it is estimated that 20 to 30% of the film exposure was the result of gamma radiation.

Although our results with the accelerator neutron source are also not too encouraging, it must be emphasized that, at this time, relatively little effort

has been made here to use such other neutron sources for radiography. Further, and more extensive investigations employing these other source type are presently in progress at other laboratories.<sup>23,24</sup>

When metal screens and films are exposed directly to the neutron beam to record the neutron image, a large portion of the photographic effect is produced by gamma radiation, both in the form of prompt ( $n, \gamma$ ) associated with neutron capture and the gamma radiation associated with the radioactive decay of the metal screen. Therefore, intensifying methods used in X-radiography would seem applicable to these neutron detecting methods. The influence of both lead screens and fluorescent screens on these neutron radiographic detection methods has been briefly investigated using the monochromatic thermal neutron source described earlier.

Par speed X-ray screens, used with the metal converter materials have led to speed increases in the order of 25 to 50%, with little obvious loss in image quality. These speed increases were found for single metal converter screens and a single fluorescent screen sandwiched around the film. These tests were made with types KK and AA X-ray films and even better speed improvements might be expected with a film such as type F film.

Best results using lead screens also were found by sandwiching the film between the single converter screen and the intensifier screen (lead in this case). For both lead and fluorescent screens less intensification was found by sandwiching the film between two extra intensifier screens because then the screen between the converter and the film tended to absorb some of the softer radiation emitted from the converter screen. With the addition of a single 0.005" lead screen, speed increases in the order of 50% over only the converter screen and film alone have been found for cadmium and gadolinium and in the order of 10 to 20% for rhodium, indium and silver. Since much of the photographic effect con-

tributed by cadmium and gadolinium screens is due to prompt  $(n,\gamma)$  radiation, the larger speed increases with the use of a lead screen with these materials seems reasonable. There appears to be no significant loss in image quality with the use of the lead screen.

This last observation is now being further studied, particularly in regard to the use of a lead intensifying screen and a gadolinium converter screen. The relatively large increase in speed and excellent image sharpness of this combination is very attractive. The large increase in speed appears to occur because the soft radiation from the converter screen is still permitted to easily reach the film emulsion along with the added contribution of the secondary radiation from the lead screen caused by the harder radiation which would otherwise not have been detected by the film.

#### DISCUSSION

Gadolinium appears to possess all the qualities desired of a metal screen for use in direct exposure neutron radiography. The tendency of this material in its naturally occurring form to become radioactive is negligible, meaning that additional transfer time after the neutron exposure is completed is not necessary and that there is very little, if any possibility of "double exposures" by using the same screen for two consecutive radiographs. More important, the material has a very high cross section for thermal neutrons (46,000 barns) so that it can be used in small thicknesses with good speed. This is one factor involved in the excellent film sharpness that has been obtained with gadolinium. The second factor is that film blackening appears to be caused to a great extent by relatively soft radiation.<sup>25</sup> Measurements made here show this in the following manner.<sup>8</sup>

If two films are placed in front of a gadolinium screen in a direct exposure neutron radiograph, the film farther from the screen will have about half the

relative exposure of the film next to the gadolinium screen. By contrast with the other metal direct exposure screen materials, it is found that, for a similar test, the film farther from the screen will have 75 to more than 90% of the relative exposure of the film placed next to a rhodium, indium, silver or cadmium screen. This increase in exposure for films placed a distance away from these other converter materials shows that more of the radiation emitted from these other screens is hard, penetrating radiation. It is true that this percentage relative exposure does vary somewhat with screen thickness because, with a thicker screen, more of the radiation is filtered by the screen itself before reaching the film, and is therefore harder. However, for most of the other screen materials, the use of thinner screens (and therefore somewhat softer radiation) becomes impractical because the increase in exposure required for those thin screens means that more scattered and secondary radiation will be recorded on the film, thereby decreasing the image quality. Even comparing similar screen thicknesses it is found that much more of the emitted radiation is absorbed in the film immediately adjacent to a gadolinium screen than for the other metal screen materials.

The soft radiation emitted from gadolinium and the high cross section for this material, account for the fact that films placed on the neutron source side of the gadolinium have greater relative exposure than back films (see Figure 7). Most of the radiation emitted from the gadolinium appears to come from the first 0.00025" thickness. As the screen becomes thicker, the soft radiation emitted toward the back film is filtered by the remaining screen thickness, yielding both less relative exposure and poorer image sharpness for the back film as contrasted to the front film.

For the other metals, the much lower neutron reaction cross sections involved mean that the radiation emitted from the thicknesses used will be more

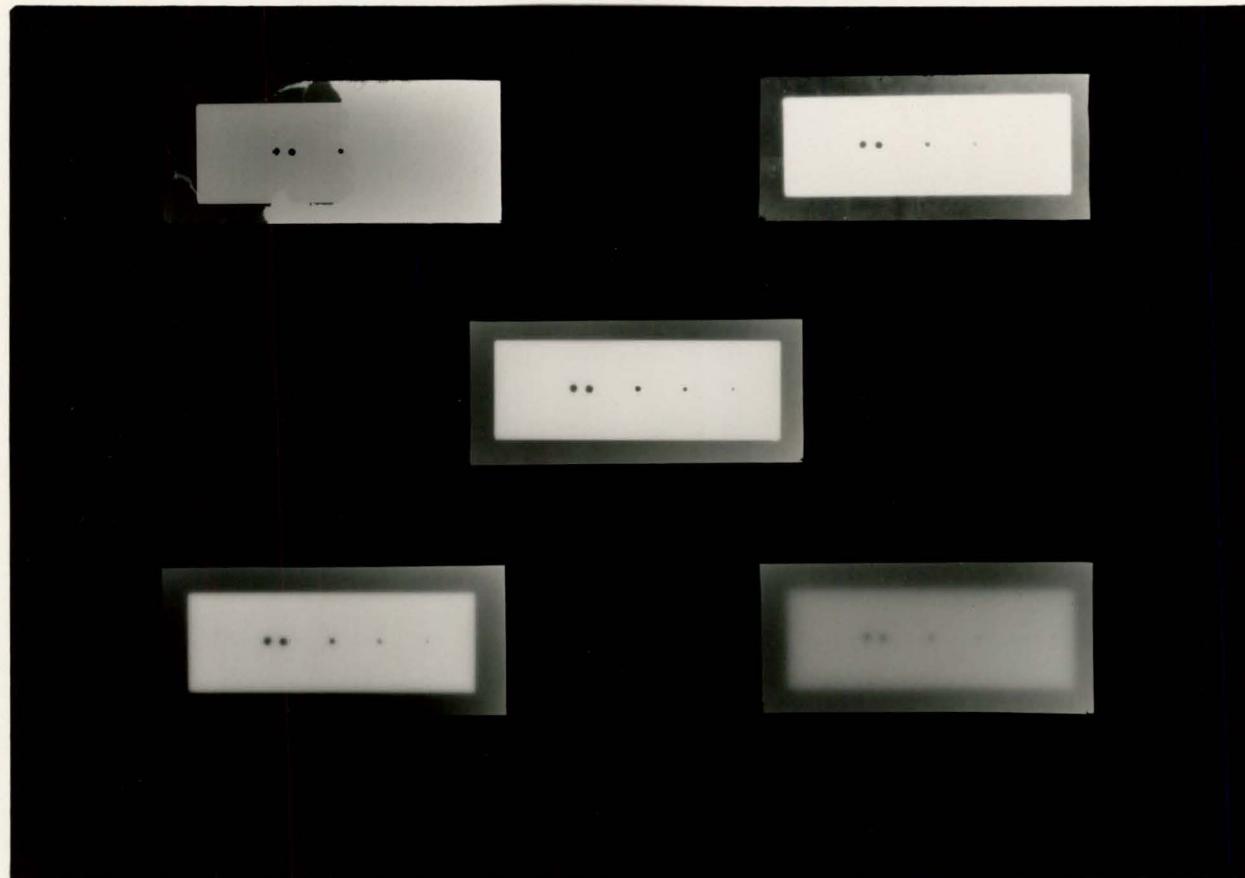
or less equally emitted throughout the thickness of the material. Slightly more radiation will be emitted from the front surface and, what is more important from the sharpness point of view, the radiation emitted from the front surface will be stronger in softer radiation. The fact that greater density is found for back films rather than front films with these other materials, would seem to be the result of secondary radiation in the converter screen generated by the gamma emission of the screen itself. Metal intensifying screens used for gamma or X-radiography are known to yield greater intensification for films placed behind the screen rather than for front films.<sup>26</sup>

An overall comparison of image sharpness qualities among the materials studied thus far rates gadolinium direct exposures, gold and indium transfers, the B-10 loaded scintillator and rhodium, indium, silver, gold and cadmium direct exposures in order of decreasing sharpness. A comparison of radiographs made with several of these materials is shown in Figure 10. Eliminating the scintillator from the following discussion, and making use of data such as that mentioned earlier, in which several films were exposed together with a converter screen, it is generally found that good image sharpness results are found for materials which emit softer radiation. This result is consistent with image unsharpness studies made with X-radiation of various energies.<sup>27</sup>

A disturbing aspect of the data presented in this report is that little difference in image sharpness was found for rather large variations in converter screen thickness. It is obvious for neutron radiography, as in the similar technique of autoradiography,<sup>28</sup> that as the screen, or sample, is reduced in thickness, the resultant radiograph will have greater contrast and better sharpness. These two qualities will both be improved because the radiation which reaches the film will originate from a thinner section, giving the radiation less opportunity to spread before reaching the film.

For the direct exposure methods, the increased neutron exposure

FIGURE 10



A reproduction of several neutron radiographs of a cadmium test piece (0.100" thick) containing several drilled holes varying from 0.0135" to 0.040" in diameter. The radiographs were taken on AA film using a 0.001" gadolinium screen and a front film (odd shape is that of available gadolinium screen when radiograph was taken), top left, 0.005" gold in a transfer method, middle; double rhodium screens (10-10), bottom left; and double cadmium screens (10-20), bottom right. The B-10 loaded scintillator is shown at the top right.

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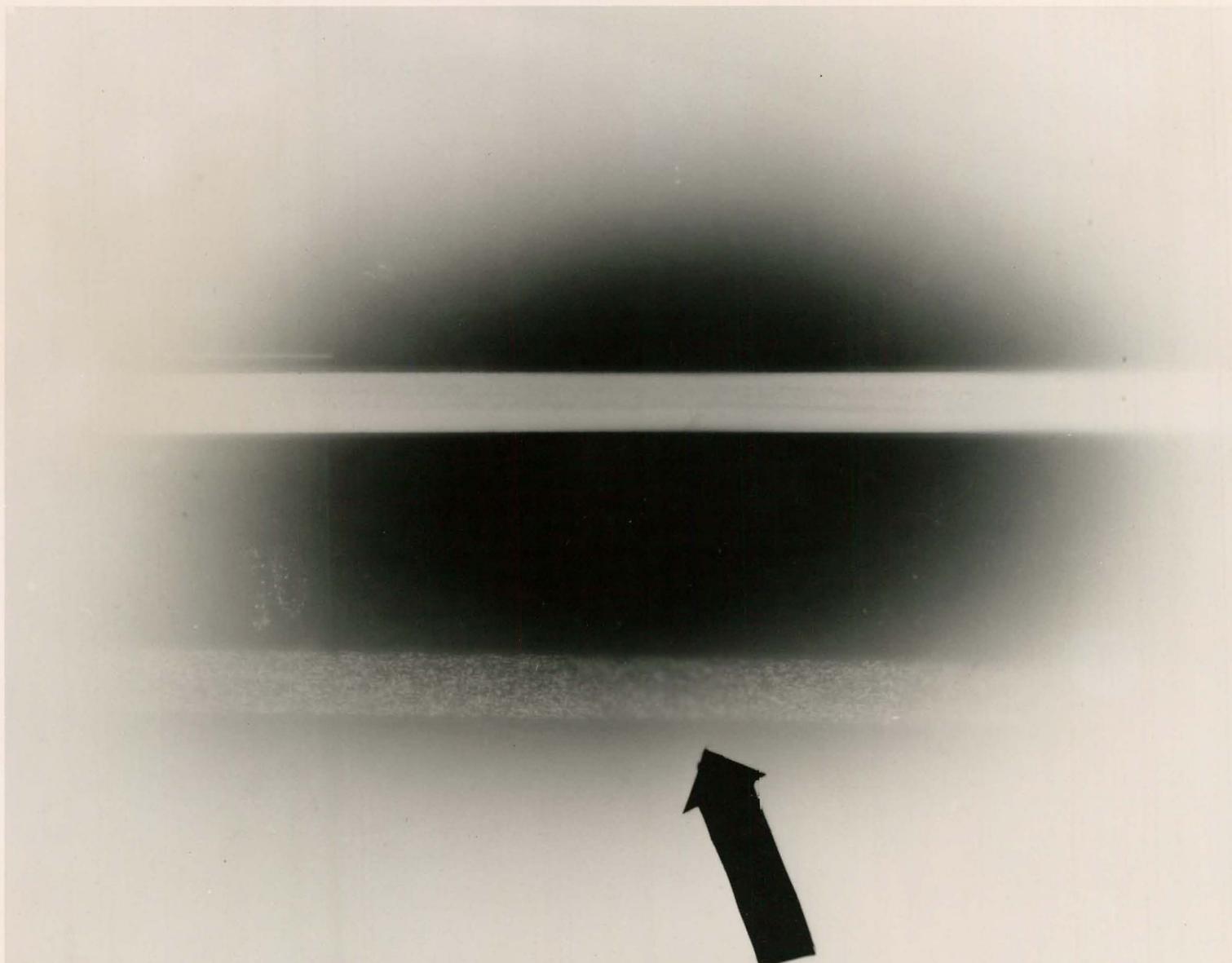
times necessary for the thinner screens may have allowed more secondary and scattered radiation to influence the film, thereby also influencing the sharpness result. This explanation however, does not seem entirely appropriate for the transfer radiographs. A possibility that does present itself is that it may be only by using much thinner screens for transfer radiography that significant improvements in image sharpness can be obtained. Other possible points to consider include our methods for maintaining good film-screen contact and for determining the image sharpness of the resultant radiograph.<sup>8</sup> Our techniques in both these areas are presently being re-evaluated.

Good screen-film contact is, of course, a very important condition which must be maintained for best results. That this is one of the places in which improvement in our present methods appears necessary is shown by the transfer neutron radiograph of samples of boron carbide reactor poison elements given in Figure 11. The radiograph was taken by exposing the objects<sup>29</sup> and a 0.005" gold screen to the neutron beam for 2 1/2 days, then transferring the radioactive gold to type M film in a spring loaded X-ray cassette for 8 days. Areas of poor film-screen contact on the radiograph are obvious. That these unsharp areas on the radiograph shown were the result of poor screen-film contact is more or less confirmed by the fact that the film exposed on the other side of the radioactive gold screen was essentially the reverse of the one used for Figure 11, as far as image sharpness is concerned. Vacuum cassettes are presently being considered as a means of overcoming such difficulties.

#### CONCLUSIONS

Since this discussion is in the nature of a progress report it seems natural to make some comparisons between results which can be obtained now and those which were reported last year. From the point of view of the radiographs themselves, the most obvious improvement is in image sharpness. This is illustrated in

FIGURE 11



A reproduction of a neutron radiograph of two rectangular cross section, zirconium clad, boron carbide reactor poison elements. This is a negative print, as are all the radiographs shown, so dark areas represent areas of high neutron intensity. The whitish spots within the shadow of the bar shaped objects (particularly in the image of the thinner of the two samples, lower view) show the locations of the boron within the samples. The arrow points out a poor film-screen contact area which resulted in poor image sharpness.

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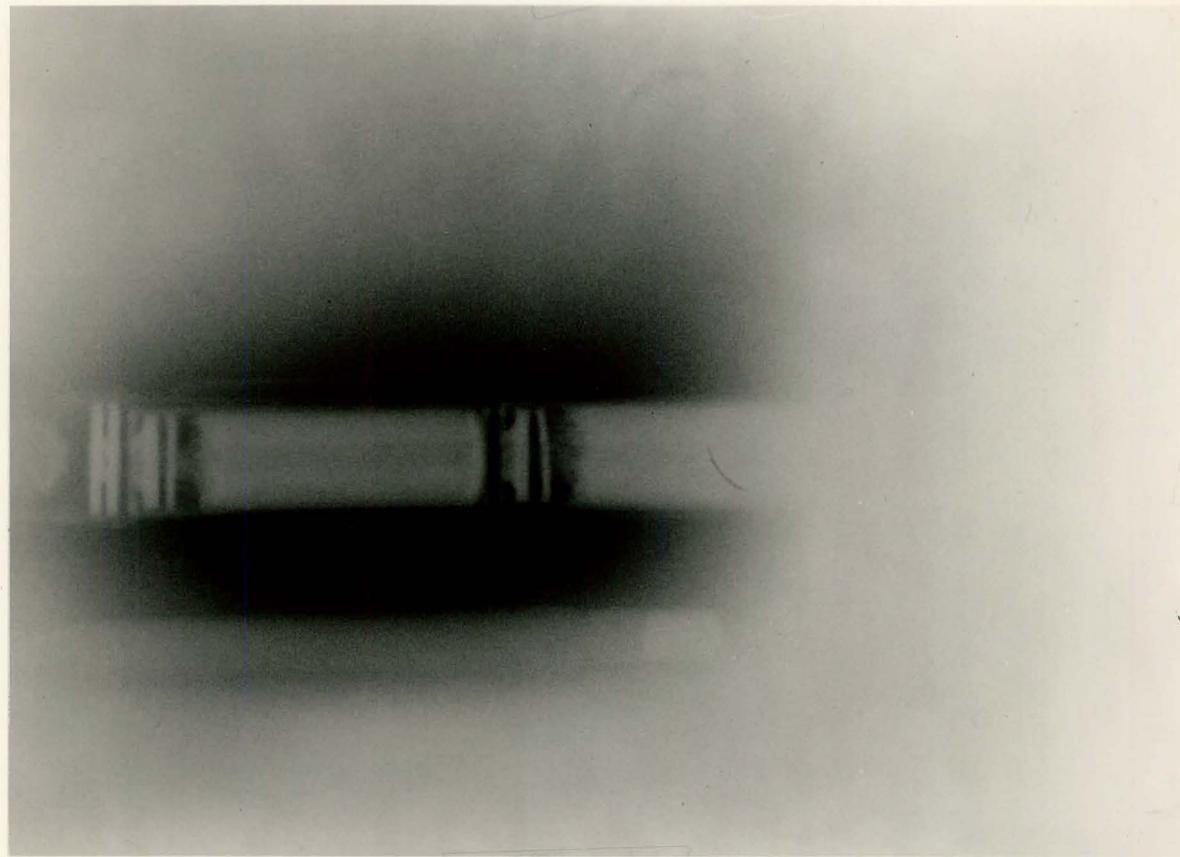
Figures 12 to 15 which show some comparisons of recent radiographs to those published in the first report.<sup>1</sup>

This improved image quality now makes a discussion of possible areas of application for neutron radiography more attractive. Although a more detailed discussion of neutron radiographic application possibilities is planned for publication elsewhere,<sup>10</sup> some discussion of the objects pictured in the radiographs shown in this report does seem appropriate.

Figure 2 was shown primarily to demonstrate the mottled image background which is characteristic of the particular B-10 loaded scintillator used in these tests. The lines on the picture are also of some importance in that they were caused by diffracted neutron beams from a powdered nickel sample. A film showing more lines from a similar sample (film was closer to the sample) is shown in Figure 16. The use of film techniques for neutron diffraction work has not been uncommon for checking alignment and general set up. In addition, film has been used for recording neutron Laue patterns.<sup>30</sup> This extension of the use of film techniques for neutron diffraction powder patterns may have some technical advantages in certain situations.<sup>31</sup> Although the use of film techniques for neutron diffraction may not be considered an application of neutron radiography, such use of neutron imaging methods may be an important by-product.

Figure 11, in addition to demonstrating the importance of good film-screen contact, also easily shows the distribution of boron within the inspection samples, a problem which would present difficulties with other test methods. Here, it illustrates the fact that absorption differences between neutrons and X-rays can be used to advantage. The X-ray absorption of the materials within the object (zirconium and boron carbide) is approximately the same, while the neutron absorption is appreciably different, the neutron absorption of the boron being much higher than that for any of the other materials present. Such a study of the distribution of a high neutron cross section material within a mixture or alloy with other, low neutron cross section materials, is a natural

FIGURE 12



A reproduction of a neutron radiograph of a pocket flashlight taken using a double silver screen direct exposure method (18-18 screens) and No Screen film. This radiograph was taken in 1960 and was used as Figure 6 of reference 1. The neutron exposure time was 6 minutes.

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FIGURE 13



A reproduction of a neutron radiograph of the same object as shown in Figure 12. This was taken recently by a direct exposure method using a 0.002" gadolinium screen and type M film. The exposure time was 2 1/2 hours. These neutron radiographs look very different than X-radiographs of the same object, primarily because of the different absorption of the plastic components within the flashlight and the hydrogenous material in the batteries.

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FIGURE 14

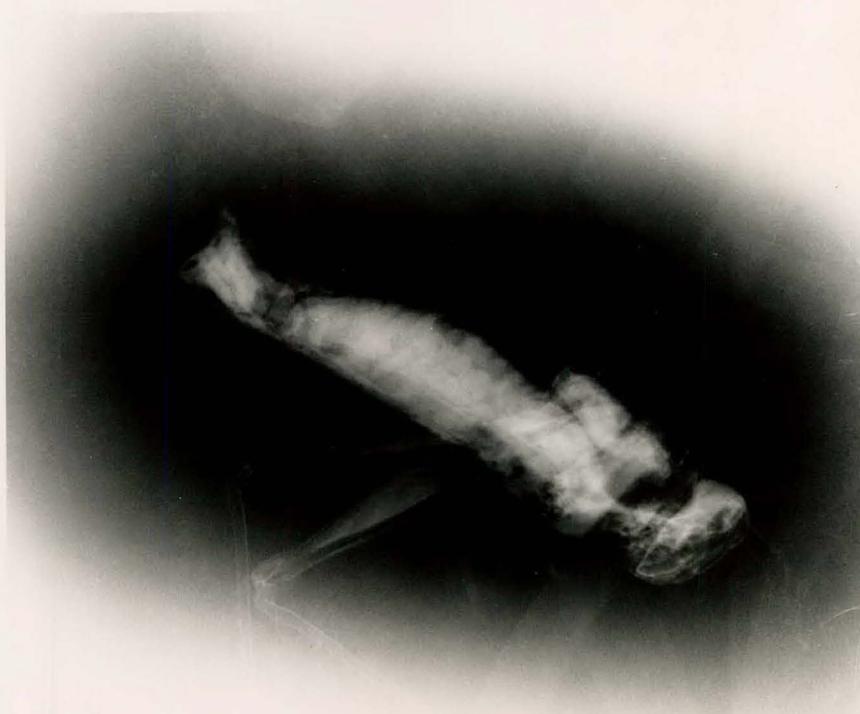


A reproduction of a neutron radiograph of a grasshopper taken with double indium screens (20-30) and AA film by a direct exposure method. The neutron exposure was 40 minutes. This radiograph was taken in 1960 and was used as Figure 9 of reference 1.

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FIGURE 15



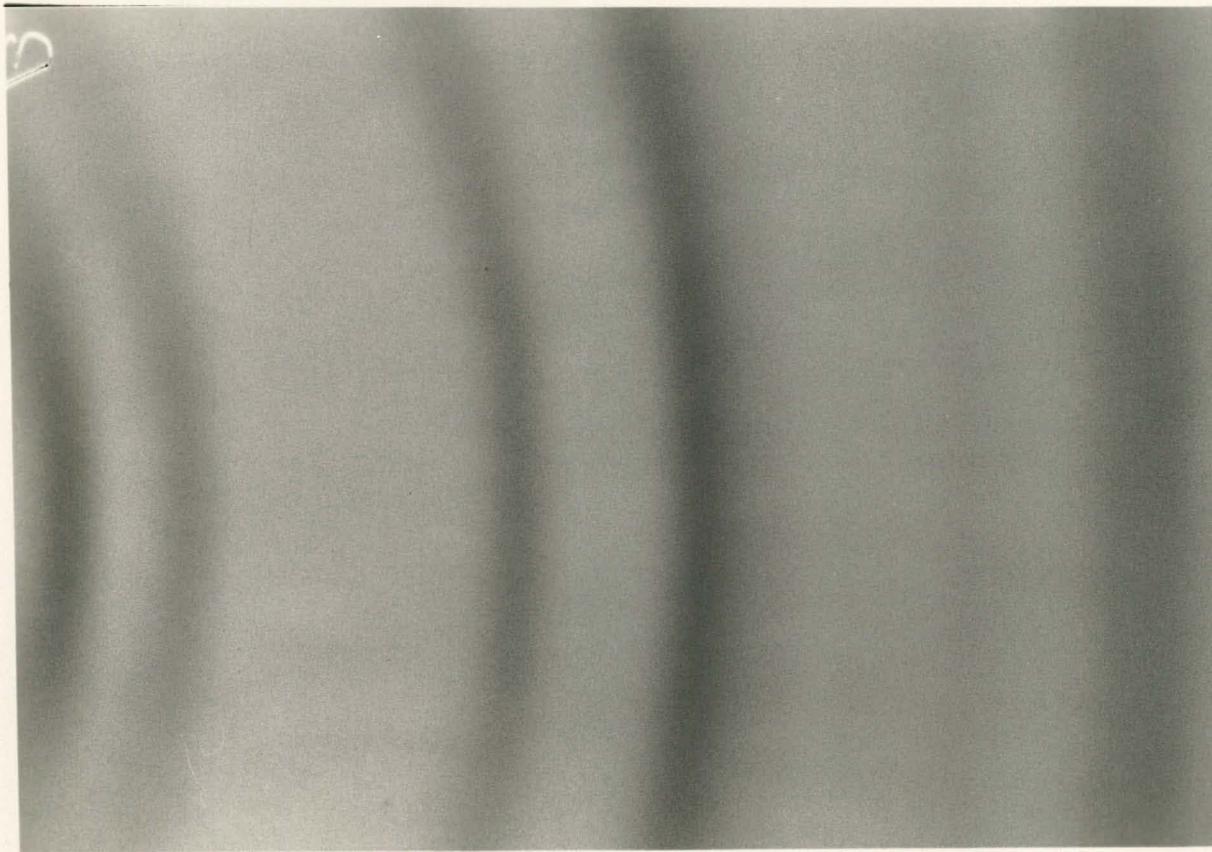
A reproduction of a recent neutron radiograph of a grasshopper taken by the direct exposure method using a 0.002" gadolinium screen and type M film. The neutron exposure was 2 hours.

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FIGURE 16



A reproduction of a neutron diffraction pattern of a powdered nickel sample taken with double rhodium screens (10-10) and type KK film in a flat cassette. The cassette was approximately 6" from a relatively thick sample (7/16" dia.). Exposure time for this picture was 15 hours. The diffracted lines shown begin with the 111 reflection on the extreme left.

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application for neutron radiography.

Other application possibilities are discussed in the previously mentioned publication.<sup>10</sup> The reader is also referred to the more complete list of possible areas of application for neutron radiography given by Watts.<sup>32</sup>

The past year's efforts have also led to some clarification of the requirements for good photographic neutron detection converter materials. For the direct exposure method gadolinium seems about perfect because of its very high cross section and because it emits relatively soft radiation. An additional requirement for a transfer technique material would be a convenient half-life for the radioactivity. Very recent tests with dysprosium screens indicate that this rare earth metal may satisfy these requirements.<sup>33</sup> Tests with both these promising materials are continuing.

#### ACKNOWLEDGMENTS

The author would like to express his appreciation to Dr. W. J. McGonnagle for suggesting this research problem and for his cooperation during its investigation, to Mr. I. R. Kraska for his assistance throughout this study, to Dr. S. S. Sidhu and his neutron diffraction co-workers for their helpful discussions and for their cooperation in making neutrons available, and to Dr. A. B. Smith and the Van de Graaff operators for their cooperation in the use of that equipment.

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15. The neutron beam used for most of this work was essentially parallel. Therefore no attempts have been made to place the object a distance ahead of the cassette in order to obtain geometrical magnification of the image.
16. The base of the speed value table was changed from silver (reference 1) to cadmium because it was felt that cadmium, with little or no radioactivity involved, would be a useful base over a very wide range of exposure conditions.
17. As pointed out in reference 1, a large portion of the film darkening for a direct exposure neutron radiograph occurs during and very shortly after the exposure. This is due to the combined effects of high cross sections for the short half-life activities and to the photographic effect of the prompt ( $n, \gamma$ ) radiation from the converter screen itself. These facts appear to account for the agreement in the literature on the order of speed for these metal screens over a wide range of conditions.

18. Argonne data tend to confirm the high speed of the 4 part ZnS(Ag) and 1 part LiF mixture used by Shull. Our results have shown a peak in speed for 4:1 and 5:1 mixtures by weight, using natural LiF.
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