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PROGRESS REPORT ON NEUTRON RADIOGRAPHY

by

H. Berger and
W. J. McGonnagle

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ABSTRACT

The potential advantages of neutron radiography as an inspection method are discussed along with a historical review and discussions of neutron sources and detectors. The results of the current investigation of neutron-image detectors are discussed in regard to photographic speed, relative neutron-gamma response, and resolution comparisons.

Two neutron-image detecting methods are discussed. In one, the direct-exposure method, both the converter screens and the film are exposed to the neutron beam together. The other method, the transfer method, makes use of a radioactive, image-carrying screen, which is transferred to photographic film after the neutron exposure is completed. The direct-exposure method results in increased speed, but has the disadvantages that the film also responds to any gamma radiation in the imaging beam and that, in most cases, improved image resolution can be obtained with the transfer method.

Reference is given to several application possibilities.

I. INTRODUCTION

Neutron radiography is a potentially useful inspection method which has been known for some time, but which has not been widely used. One reason it has not been put to use is the scarcity of information concerning optimum, or even useful, techniques for performing neutron radiography. A program to investigate these techniques and to better determine the potential usefulness of neutron radiography is currently underway. This publication is a progress report on this program.

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*A preliminary account of this work was presented at the Symposium on Physics and Nondestructive Testing held at Argonne National Laboratory, October 4-5, 1960. See Reference 1.

Several things are required in order to obtain a neutron radiograph; these include a source of neutrons, an absorbing object, and a surface sensitive to the image-carrying radiation. In addition, of course, there should be some reason for employing neutron radiography instead of some other inspection method, such as X or gamma radiography.

The reason for considering neutron radiography is of prime importance. This "raison d'être" for neutron radiography comes about because the relative neutron absorption of materials is very different from the relative absorption of these materials for X rays. The mass absorption coefficient for neutrons is quite random when examined in terms of regularly increasing atomic number, whereas the mass absorption coefficient for X rays increases with some regularity if it is similarly examined. This fact is very well demonstrated graphically by Thewlis,[2] who plots mass absorption coefficients for X rays (0.098 Å) and neutrons (1.08 Å) versus atomic number. This plot is shown in Figure 1. It is particularly interesting to find very high neutron mass absorption coefficients for several of the lighter elements, which can present rather difficult problems with X-ray techniques. Notably, light materials such as hydrogen, lithium, and boron, absorb thermal neutrons to a high degree. Other materials having high neutron absorption coefficients include cadmium, samarium, europium, gadolinium, and dysprosium. Absorption differences such as these mean that neutron radiography may present real advantages in certain specific inspection problems.

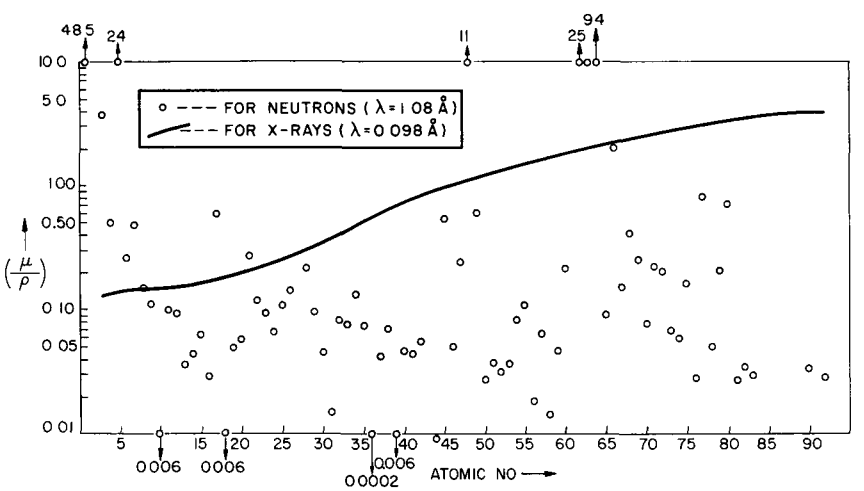

Figure 1

A Comparison Plot of Mass Absorption Coefficients for Neutrons and X Rays (After Thewlis)

It should be mentioned at this point that this discussion will be limited, with one exception, to thermal neutron radiography. The neutron energies involved, then, will be a fraction of an electron volt, and the

corresponding wavelengths will be of the order of one Angstrom unit. The principal reason for not considering neutrons of higher energy is that the relative absorption differences which are prevalent for slow neutrons decrease appreciably for fast neutrons. To the fast neutron, then, most materials are almost equally absorbing. The increased scattering experienced with the use of fast neutrons and the decreased efficiency for fast neutron detection are also factors involved in limiting this discussion to thermal neutrons.

A second limitation on this discussion is concerned with the method of detecting the neutron image. For the most part, photographic detection methods will be discussed. One point which should be mentioned in this connection is that neutrons, fast or slow, have relatively little effect on normal photographic emulsions. The detection techniques which have been employed, for the most part, make use of some intermediate material which is placed next to the photographic emulsion and which emits some photographically detectable radiation upon neutron irradiation. Materials which have been used for such neutron-intensification screens include lithium or boron in conjunction with fluorescent materials (n, α, light reaction), cadmium (n, γ), and beta emitters such as silver, indium, and gold. These methods will be more fully discussed in a later section.

Before discussing neutron sources and neutron-image detectors in detail, let us take a moment to briefly review some previous work in neutron radiography.

II. HISTORICAL BACKGROUND

Successful neutron radiographic experiments were conducted more than 20 years ago in Germany by Kallmann and Kuhn.[3] The neutron source used by Kallmann employed ion bombardment of a suitable target to produce a source of fast neutrons and subsequent slowing down, or moderation, of the neutron beam by water. The arrangement is pictured in Figure 2. The water-free channel and the opening in the boron shield made an exit aperture for the slow neutron beam. The object and film pack were covered with lead to lessen the photographic effects of any gamma radiation in the imaging radiation beam. Kallmann had the greatest success with a detector consisting of photographic film sandwiched between 2 fluorescent

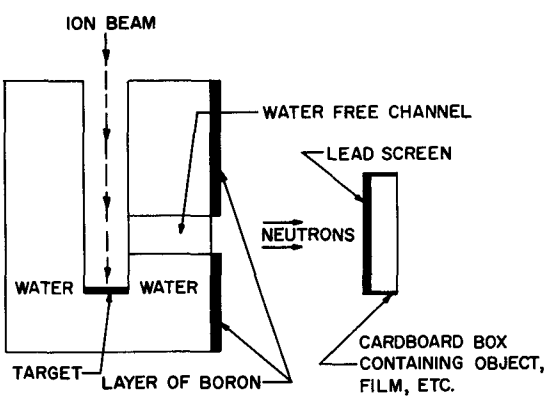

Figure 2

Neutron Source and Arrangement Used by Kallman for Neutron Radiography

layers. The fluorescent layers were boron- or, preferably, lithium-coated, and the resultant alpha emission from the neutron-bombarded lithium resulted in light emission from the fluorescent layers. This light emission produced film blackening. Further improvements were obtained by Kallmann by depositing a thin ($<0.5\ \mu$), aluminum reflecting layer between the lithium or boron and the fluorescent layer.

Kallmann also investigated several other materials which could be used in the photographic detection of neutron images. In particular, he mentioned the gamma emission from cadmium and the beta emission from gadolinium, silver, and indium as being photographically useful for the detection of neutrons. Of these, gadolinium seemed to give the best results.

In this pioneering work in the field of neutron radiography by Kallmann, he also recognized the disturbing effects which result from the gamma radiation which is almost always associated with the neutron beam. Neutron-image detection systems are usually also gamma sensitive, so that the gamma image is also recorded. Since the advantages of neutron radiography are allied with the fact that the relative absorption of X or gamma radiation is different from that of neutrons, the superimposed image due to gamma radiation will tend to obscure the sought-for neutron image. Kallmann concluded that shielding the neutron source did not completely solve this problem, because the additional gamma radiation produced in the shielding materials tended to offset any shielding advantage they might offer. Increased distance of the object and film from the source was one solution to this problem, and since this solution also tended to increase resolution, it is apparently the one he found most satisfactory. He did point out that neutron pictures which were completely free of the disturbing effects of gamma radiation could be obtained by irradiating foils which became radioactive and then transferring these radioactive image-carrying foils to photographic film. Materials such as silver and indium were indicated as being especially suitable for this technique.

Peter[4] extended the work of Kallmann and Kuhn by employing a discharge tube of higher intensity and therefore obtaining a neutron source of higher intensity. The source used by Peter was essentially as pictured in Figure 2 and was said to be 13 kg of radium equivalent. With this increased neutron intensity, Peter could obtain neutron radiographs by exposing X-ray film sandwiched between 0.004-in. silver screens and backed by 0.020-in. cadmium in 1 to 3 min. This compares with the 4-hr exposures reported by Kallmann with his lithium-phosphor-film combination and a neutron source said to be 2 to 3 gm of radium equivalent. Peter also published a picture made by transferring a radioactive, image-carrying silver screen to a piece of film out of the neutron beam. Details of exposure in this latter picture were not discussed.

More recently, an investigation of neutron radiography was conducted in England by Thewlis and Derbyshire.[2,5] A nuclear reactor (BEPO) was used to supply a source of thermal neutrons for these experiments, and the

detection method made use mainly of indium screens and X-ray film. The radiographs published by Thewlis were of comparable resolution quality to many gamma-ray radiographs. In addition to obtaining radiographs of improved quality over the earlier work of Kallmann and Peter, Thewlis also attempted to point the way toward applications of this inspection method. Among the application areas indicated as being attractive were the inspections of neutron shielding material and heavy metals, and the use of neutron radiography in biological studies. It is the high neutron absorption of hydrogen which makes this technique of potential value in this latter application. Neutron radiography is, of course, ideally suited to the inspection of neutron shielding materials, such as Boral sheet. The method also seems to offer advantages in the inspection of heavy materials, such as uranium, because of the reduced exposure time which should result from lower neutron absorption coefficients. The greatest use, however, would seem to be in radiographing objects made of materials which do not lend themselves to other inspection means. A neutron radiograph of a piece of waxed string in a 2-in. lead block was used by Thewlis to illustrate this point very effectively.

III. NEUTRON SOURCES

With this background of previous work in this field, let us now look at some of the neutron sources which might be useful for this application. Although there are many source variations possible, the different types of neutron sources can be rather conveniently grouped as (1) radioactive sources, (2) accelerators, and (3) nuclear reactors. From experience with other imaging systems, some desirable characteristics for a neutron radiographic source can be indicated. For good qualities of image resolution the source should originate from a small area, or have enough intensity so that objects can be radiographed at a considerable distance from source to film in a reasonable time. The source should also provide a uniform radiation intensity over the desired inspection area. A further requirement is that the radiation beam should not contain a masking radiation present in any significantly detectable intensity. In the case of a neutron radiographic source, the ratio of gamma to neutron radiation should be low.

Practically speaking, the first general requirement, that of a small originating area for the neutron source, is difficult to achieve for thermal neutrons. Thermal neutrons are usually obtained from neutron sources of higher energy by allowing the neutrons to diffuse through a moderating material.[6] For thermal neutron sources, light materials such as hydrogen, deuterium, beryllium, or carbon are normally employed as the scattering or moderating material. The slowing-down process in this case is the result of collisions between the neutrons and these light nuclei. Appreciable thickness of moderating material is usually required to accomplish this slowing-down process and, as a result, thermal neutron sources are generally large in size compared with the focal spot of an X-ray tube, for example.

In addition to this comment on neutron sources in general a few words about each type of neutron source might be in order.

Radioactive neutron sources of practical size usually have low neutron yields, making them rather unattractive for this type of application. Sources of high yield are of the Ra-α-Be type and produce of the order of 10^7 n/sec-c. These neutrons, however, are fast neutrons, which include energies up to 13 Mev. After moderation and collimation, the thermal neutron yield for radiographic use would be appreciably less. Some idea of possible exposure times with such a source may be gained by a comparison with the work of Kallmann, who used a neutron source said to be 2 to 3 gm of radium equivalent. His exposures were of the order of 4 hr. An additional problem encountered with radioactive (α, n) sources is that they have a relatively high ratio of gamma to neutron radiation.* This would pose a shielding problem in the use of such sources for neutron radiography, or would indicate the use of a method such as a radioactive foil to eliminate the masking effect of the gamma radiation.

Radioactive photoneutron (γ, n) sources employ primarily beryllium or deuterium, since these have relatively low thresholds for the (γ, n) reaction (1.67 and 2.23 Mev, respectively). These (γ, n) sources combine a γ-emitting isotope (whose γ energy is above the threshold for the (γ, n) reaction) with either beryllium or deuterium. These sources usually have a rather low neutron yield** and offer no particular advantage over the (α, n) sources for this application.

High-voltage accelerators are capable of producing much more intense neutron beams than practical radioactive sources. As in the case of the radioactive source, however, moderation and collimation of the neutron beam is required in order to make it useful for most neutron radiography. Arrangements such as that used by Kallman and Peter (see Figure 2) accomplish this purpose.

Many neutron-producing reactions are possible with accelerators, including the (γ, n) reaction discussed previously. In this case, high-energy X rays provide the photon energy necessary for the reaction. Reactions used more generally for neutron production, however, involve the acceleration of positive ions toward various neutron-yielding targets. The neutron yields will be dependent upon the target material, the accelerated particle, the accelerating voltage, and the intensity of accelerated particles. In general, the fast neutron intensity available from normal accelerators will be of the order of 10^6 to 10^{11} neutrons/cm^2-sec.

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*One exception to this rule is the Po-α-Be source. The relatively short half-life (138 days) is a major disadvantage, however.

**Recently a 10^{10} n/sec Sb-Be source has been described.(7)

By comparison with these other sources, very high neutron intensities are available from nuclear reactors. For example, the CP-5 reactor at Argonne has a neutron flux of 3.5×10^{13} neutrons/cm^2-sec in the region of highest flux. Other reactors have neutron intensities appreciably higher than this. Although it is true that the neutron flux which is useful for neutron radiography will be of much less intensity by the time it is collimated and brought out to a useful location, the nuclear reactor still yields a higher useful thermal neutron flux than would be obtained from normally available radioactive sources or accelerators. For this reason, it can be concluded that a nuclear reactor should be the preferred source for use in this application.

However, from a practical standpoint, the accelerator seems to offer the best possibility for more generally available neutron sources for neutron radiography. Although the neutron intensities may be somewhat lower than those obtainable with a nuclear reactor, they still can be of the order of magnitude of those used in this investigation at Argonne (10^5 n/cm^2-sec) and can therefore produce neutron radiographs in a reasonable exposure time (10 min or less).*

IV. NEUTRON ABSORPTION

The absorption of neutrons in material is a combination of what might be called true absorption (absorption of a neutron by a nucleus) and neutron scatter. Both processes remove neutrons from the incident neutron beam and therefore contribute to absorption. These factors are both considered in the total cross section for the material.

The absorption of neutrons is given by the familiar absorption equation:

$$I/I_0 = e^{-N\sigma_t x} \quad , \tag{1}$$

where I/I_0 = the fraction of the neutron beam transmitted through an absorber of thickness x

e = Naperian logarithm base

N = number of nuclei per cm^3 in the absorbing material

σ_t = total cross section in units of cm^2 (1 barn = 10^{-24} cm^2)

x = absorber thickness in cm.

More extensive discussions of neutron absorption are given in the literature,[11] as are tables of neutron cross sections.[12] It should again be emphasized that the fact that the relative neutron absorption of materials

*Further work on a comparison of various types of neutron sources for use in neutron radiography is planned. In the meantime, the reader desiring more information is referred to several reviews in the literature.[8,9,10]

is different from that of X radiation is what makes neutron radiography a potentially useful inspection technique. The high neutron absorption of several light elements seems to be particularly advantageous, as is the potential ability to discriminate between several materials which have similar X-ray absorption coefficients.

V. PHOTOGRAPHIC NEUTRON IMAGE DETECTION

A. Background Information

In addition to the work mentioned previously on neutron radiography, there have been several other investigations of the photographic detection of neutrons. There are, for example, publications concerned with radiation dosimetry,[13-16] recording of neutron diffraction patterns,[17,18] and other applications.[19-21] As has already been mentioned, neutrons have little influence on normal photographic emulsions, so that intermediate materials, which emit some photographically detectable radiation, are normally used next to the film. The intermediate materials reported in these references include the alpha emitters boron or lithium combined with a phosphor, cadmium and gadolinium which emit gamma radiation when bombarded with slow neutrons, and beta emitters (such as indium, silver, gold, and rhodium). Photographic neutron detection has also been studied with boron- or lithium-loaded film emulsions and standard emulsions.

The characteristics of several of these neutron photographic-image-intensification materials are shown in Table I. All of the columns shown in the table are of importance in choosing an intensifier material for use in neutron radiography. First of all, the relative abundance of the particular isotope involved should be high, so that the naturally occurring material of that element will contain a large number of the desired nuclei. The reaction cross section should also be high, in order that the probability of the reaction be large. The radiation emitted should be photographically effective and, finally, the half-life of the material (if it is a radioactive reaction) should be convenient. The reported relatively poor photographic response found with the use of gold, for example, stems from the long half-life (2.7 days) of the material, and our reluctance to consider exposure times which would be long enough to approach a saturation activity in the gold screens.

The data in the table indicate that, in some cases, several possible reactions* are likely to produce photographically detectable radiation from the intensifier or converter material. For example, in the case of silver,

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*Although it is not shown directly in Table I, (n, γ) radiation adds appreciably to the photographic response of the materials shown as radioactive when they are used in the direct-exposure method.

the approximately equal division of naturally occurring material between the two stable isotopes Ag^{107} and Ag^{109} and the relatively high reaction cross sections for the 2.3-min Ag^{108} and 24.2-sec Ag^{110} activities means that both these isotopes will be responsible for much of the photographic effect obtained with silver screens for neutron radiography.

Table I

CHARACTERISTICS OF SEVERAL NEUTRON PHOTOGRAPHIC IMAGE INTENSIFIERS

MATERIAL	ISOTOPE INVOLVED IN REACTION	RELATIVE NATURAL ABUNDANCE (PER CENT)	CROSS SECTION FOR REACTION (THERMAL NEUTRONS VELOCITY = 2200 m/s) (BARNS)	REACTION	RADIATION PRODUCING PRINCIPAL PHOTOGRAPHIC EFFECT
LITHIUM	Li-6	7.52	910	Li-6(n, α)H-3	α EMISSION OR PREFERABLY LIGHT EMISSION EXCITED IN AN ASSOCIATED PHOSPHOR BY THE α.
BORON	B-10	18.8	3,770	B-10(n, α)Li-7	SAME AS LITHIUM
RHODIUM	Rh-103	100	12	Rh-103(n)Rh-104m	ISOMERIC TRANSITION TO Rh-104 OF HALF LIFE 4.5 MINUTES
			140	Rh-103(n)Rh-104	BETA EMISSION (2.5 Mev) OF HALF-LIFE 44 SECONDS
SILVER	Ag-107	51.35	44	Ag-107(n)Ag-108	BETA EMISSION (1.77 Mev) OF HALF-LIFE 2.3 MINUTES
	Ag-109	48.65	2.8	Ag-109(n)Ag-110m	BETA EMISSION OF HALF-LIFE 270 DAYS
			110	Ag-109(n)Ag-110	BETA EMISSION OF HALF-LIFE 24.2 SECONDS
CADMIUM	Cd-113	12.26	20,800	Cd-113(n, γ)Cd-114	GAMMA RADIATION PLUS SECONDARY RADIATION
INDIUM	In-115	95.77	145	In-115(n) In-116m	BETA EMISSION (ENERGY 1 Mev AND LESS) OF HALF-LIFE 54.1 MINUTES
			52	In-115(n) In-116	BETA EMISSION (ABOUT 3 Mev) OF HALF-LIFE 13 SECONDS
GOLD	Au-197	100	96	Au-197(n)Au-198	BETA EMISSION OF HALF-LIFE 2.7 DAYS

Data for this table were obtained from references 6 and 12 and from Table of Isotopes by D. Strominger, J. M. Hollander, and G. T. Seaborg, Rev. Mod. Phys., 30, 585 (April 1958)

Relatively little work has been done to make a comparison of the relative photographic effect (speed) of these various materials, nor has much been reported on comparing qualities of image resolution for these materials. These questions have been the subjects of this investigation of neutron radiography, and some of these results can now be reported. All of the data reported herein have been taken with a monochromatic neutron beam from the Argonne Neutron Spectrometer.[22] In this neutron-diffraction equipment, a collimated neutron beam from the reactor (Argonne Research Reactor CP-5) is brought out to a monochromatizing crystal. The monochromatic neutron beam from the crystal emerges through a set of defining slits and a beam monitor to the neutron-diffraction sample. The diffracted beams are detected by a BF_3 counter on the movable spectrometer arm. Most of the monochromatic neutron beam is transmitted through, or around, the sample and is caught by a beam catcher at a

distance of about 5 to 6 ft from the sample. This beam at the beam catcher, having a maximum intensity in the order of 10^5 thermal neutrons/cm^2-sec has been employed for these tests reported below. A schematic drawing of this arrangement is shown in Figure 3.

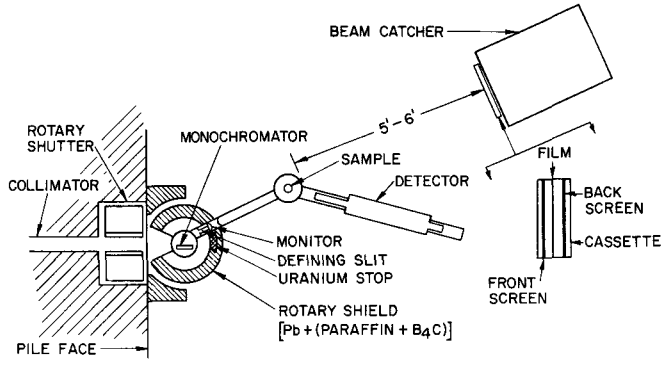


Figure 3

Schematic Drawing of Neutron Spectrometer
Showing Arrangement Used for
Neutron Radiography

This neutron beam has a rather small area of uniform maximum intensity, covering approximately a $1\frac{1}{2}$ x 3-in. oval at the beam catcher. Scatter increases the overall coverage to about 3 x 5 in. In spite of this small coverage area, many radiographs have been taken with this beam and, in fact, the beam was particularly useful in obtaining film-density data for the various neutron-intensification materials. The ratio of gamma to neutron intensity was quite low as far as film-blackening effects are concerned,* and the fact that the neutron beam was monochromatic made absorption calculations relatively simple. The (de Broglie) wavelength of the neutron beam used for the majority of these measurements was (1.05 ± 0.01) Å, which corresponds to an energy of 0.055 ev.

B. Experimental Determination of Thicknesses of Converter Screens for Optimum Film Density

The first efforts of this current research program attempted to determine experimentally the thickness of the various intensifying materials which yielded the greatest film density for a comparable neutron exposure. For the beta emitters, an estimate for the optimum thickness can be made on the basis of the range of the energy of the beta particle. However, the intensifier thickness which produces the optimum photographic density can be difficult to determine strictly on a theoretical basis,

*A sheet of No Screen film exposed to the neutron beam without screens for a period of 3 times as long as that used for normal neutron exposures (18-min versus 6-min normal exposure) showed no detectable image.

because the decay schemes usually include several beta and gamma emission energies. For the gamma emitters, such as cadmium, the optimum thickness should essentially absorb the neutron beam, assuming the resultant screen thickness does not appreciably absorb the gamma radiation.

The data presented about optimum screen thickness were obtained by sandwiching X-ray film between known thicknesses of intensifier screen material and exposing this combination to the neutron beam for a known total neutron exposure. In the case of the screens which became radioactive upon neutron exposure, the screens were permitted to remain in contact with the film for a period of at least 3 half-lives after the neutron exposure was completed. Films and screens were exposed to the neutron beam contained in an aluminum-front, spring-loaded X-ray cassette. In so far as possible, films exposed in a particular set of experiments were processed together to eliminate discrepancies due to developing differences. Films were developed in Kodak Liquid X-Ray Developer ($20 \pm \frac{1}{4}$°C) for 5 min without agitation. Density measurements were taken with a Macbeth-Ansco Densitometer (model 12A) having a density range from 0 to 6.0.

Figure 4 is a graph of the data used to determine the optimum screen thickness for cadmium intensifiers. In the upper portion of Figure 4, the net density produced on KK X-ray film is plotted against the thickness of the back intensifying screen (the screen farthest from the neutron source). In each curve shown, the front screen is held constant and the thickness is indicated on the curve. It can be seen that the curves all tend to level off for a back screen thickness of 0.020 in. and that the highest density curve is obtained for a front screen thickness of 0.010 in. The lower curve is a plot of film density versus front screen thickness for a back screen thickness of 0.020 in. This curve shows a broad peak in density for a front screen varying in thickness from 0.010 to 0.015 in. From these data, it has been concluded that the optimum film density for exposure of KK film and cadmium screens to 0.05-ev neutrons is produced by a combination of a 0.010-in. front screen and a 0.020-in. back screen. The shorthand designation used in this article to indicate such an arrangement is 10-20 Cd, where the first number refers to the front screen thickness in mils, the second number to the back screen thickness in mils, and the chemical symbol identifies the material.

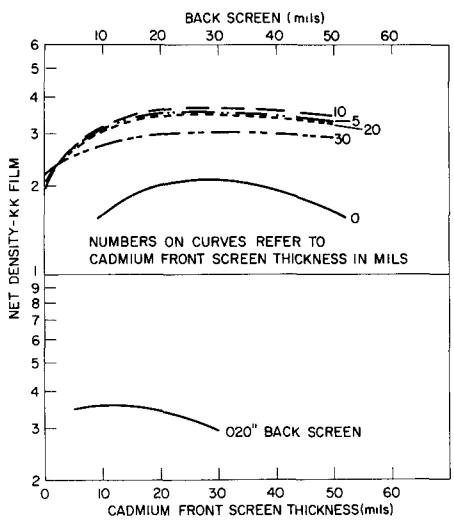


Figure 4

Determination of Optimum Cadmium Screen Thickness for Constant Neutron Exposure

It is interesting to note that 0.010-in. cadmium absorbs about 95% of a 0.05-ev neutron beam. The additional density produced by the back screen seems to be the result of electron emission and secondary radiation generated in the cadmium by the hard gamma radiation from the cadmium (n, γ) reaction. This is somewhat confirmed by the fact that there is little change in film density produced by replacing the cadmium back screen by a 0.010-in. lead screen, and by the fact that some increase in density is obtained by using the normal cadmium front and back screens but sandwiching the film between 0.005-in. lead screens.

The thicknesses of cadmium screens used for photographic neutron detection by other workers varied from 0.010 to 0.040 in. The neutron-dosimetry investigations reported by Dessauer[23,24] indicated that the optimum cadmium screen thicknesses for use in mixed radiation fields should be 0.020-in. front and back screens. Since Dessauer applied his screens by folding a single cadmium sheet around the film pack, however, he may not have investigated front and back screens of different thicknesses.

It should be mentioned that the optimum screen thicknesses determined in this report take into account only relative photographic speed. It is recognized that thinner screens might very well yield improved resolution.

Optimum screen thicknesses for rhodium, silver, and indium have also been determined in a similar manner and the data are shown graphically in Figures 5-7. Results from these curves indicate that there is little to be gained by increasing the screen configurations beyond 10-10 for rhodium, 18-18 for silver, and 20-30 for indium.

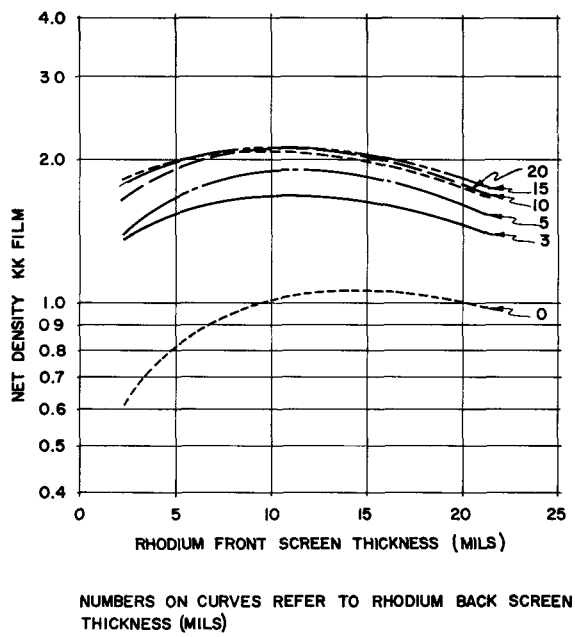


Figure 5

Determination of Optimum Rhodium Screen Thickness for Constant Neutron Exposure

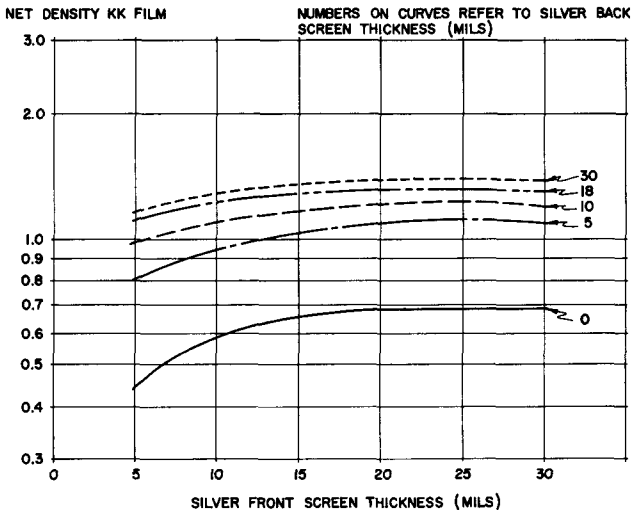
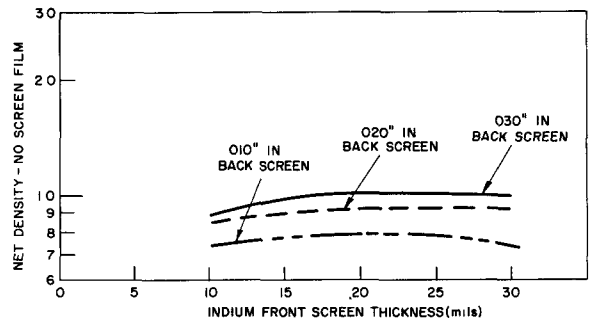


Figure 6

Determination of Optimum Silver Screen Thickness for Constant Neutron Exposure

Figure 7

Determination of Optimum Indium Screen Thickness for Constant Neutron Exposure



Optimum thicknesses for the other metal screens used (gold and gadolinium) have not yet been determined experimentally. The screen thicknesses used for gold (6-10) are of the order of the range of the emitted beta particles, whereas the thicknesses used for gadolinium (1-2) were chosen to essentially absorb the incident neutron beam.

C. Relative Photographic Speed Comparisons

The relative photographic speed of several of these materials used in the direct-exposure method has been investigated for a particular exposure (6 min) to a particular neutron flux (9.4×10^4 n/cm^2-sec),* and the data are summarized in Table II. Cadmium has arbitrarily been rated 1.0.

Although one cannot put too much reliance on these photographic speed figures, they do indicate the order of magnitude of decrease in exposure times which can be obtained by using the materials with high relative photographic speed.

*Flux measurements were made by gold-foil activation.

Table II

RELATIVE PHOTOGRAPHIC SPEED OBTAINED WITH VARIOUS NEUTRON IMAGE DETECTORS

Detector Configuration	Relative Photographic Speed[a]	
B^{10}-loaded Scintillator[b]	38.6 (type F film)	
B^{10}-loaded Scintillator[b]	12.5	No Screen Film
Rhodium[c] (10-10)[d]	1.46	
Indium[c] (20-30)[d]	1.1	
Cadmium (10-20)[d]	1.0	
Silver[c] (18-18)[d]	0.76	
Gold[e] (6-10)[d]	0.28	

[a] Relative photographic speed obtained by comparing film density and relative exposure on characteristic curves. Cadmium is arbitrarily set equal to 1.0. All neutron exposures were 6 min in a neutron flux of $9.36 \times 10^4 n/cm^2$-sec unless otherwise indicated.

[b] A special scintillator screen similar to the type NE402 from Nuclear Enterprises, Ltd. was used for these tests. Relative exposure data were obtained by shorter exposure comparisons with similar exposures using cadmium screens.

[c] Neutron exposures of radioactive screens and film were followed by a 3-half-life radioactive decay transfer period in which the screens were allowed to remain in contact with the film. Decay times used were 13.2 min for rhodium, 2.7 hr for indium, and 6.9 min for silver.

[d] Numbers refer to front and back screen thicknesses, respectively, in mils.

[e] The radioactive decay transfer period for the gold screens, after the neutron exposure, was one half-life (2.7 days). The speed value for gold would not be doubled by allowing the gold to decay 3 half-lives or more because of the (n,γ) film exposure during the neutron exposure.

Variations from the values shown would be expected for different exposure situations because of reciprocity-law failures for the scintillator,* and because of different times involved for the radioactive materials to reach saturation activities. In the data for this table, silver has almost reached saturation activity (2.3-min half-life, 6-min exposure); longer exposures would probably tend to change the relative speed ratings for rhodium, indium, and gold by allowing these materials to reach a greater percentage of saturation activity.

An indication of the total neutron exposure required to obtain a given film density under these various conditions can be gained from the fact that the 6-min exposure for the 10-20 cadmium configuration using No Screen film yielded a total film density of 1.5. The total neutron exposure involved in this case was 3.4×10^7 n/cm^2. A fair approximation for the neutron exposure required to yield a total film density of 1.5 by means of the other neutron detectors listed in Table II can be obtained by dividing this exposure for cadmium (3.4×10^7 n/cm^2) by the speed number in Table II. On this basis, then, a neutron exposure of only about 8×10^5 n/cm^2 is required using the B^{10}-loaded scintillator and type F film. An easily recognizable neutron image using this configuration can be obtained with a total neutron exposure of the order of 2000 n/mm^2 with a neutron intensity of the order of 10^5 n/cm^2-sec.

For the other materials, a further increase in speed over the No Screen film data, of approximately 1.5, can be obtained by using type KK X-ray film.

As indicated above, the neutron flux and the neutron exposure time have some bearing on these speed figures. However, rather large variations in flux and time are probably necessary in order to detect appreciable differences in the relative speeds indicated. Recent tests using 2-min exposures to a neutron flux of about 3×10^5 n/cm^2-sec yielded approximately the same speed values.

This information on relative photographic effects confirms comparisons of intensifier materials by other workers. Rausa[13] has indicated that rhodium yields greater film density than cadmium for the same neutron exposure. Dessauer[24] further indicated that the order of relative photographic effectiveness was rhodium, indium, and cadmium, and Kallmann's work pointed out that the effectiveness of cadmium and silver was about the same.[3]

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*The scintillator used was a commercial adaptation of the B^{10}-loaded ZnS phosphor described by Sun et al.[17] Only one scintillator screen was available, and in the configuration used for these tests it was employed as a back screen. This scintillator is contained in a different physical package than the one discussed in a previous report.[1] The separation of the film and scintillator is very small (<0.001 in.) permitting good light collection.

D. Relative Response of Detectors to Neutrons and Gamma Radiation

From the neutron exposures required to obtain a total film density of 1.5, as calculated from the speed data of Table II, and from similar data on gamma detection for these various methods of neutron detection, a comparison of relative neutron-gamma detection can be made. These data are shown in Table III. The data for this table were obtained in the following manner. The neutron exposure to obtain total film density of 1.5 was calculated by dividing the required neutron exposure for cadmium screens by the relative speed. The same neutron-detecting configurations were also exposed to cobalt-60 gamma radiation. From the exposures and densities obtained, and from the film characteristic curves, the gamma exposure required to obtain a total film density of 1.5 was calculated.

Table III

RELATIVE RESPONSE OF SEVERAL DIRECT-EXPOSURE PHOTOGRAPHIC DETECTION METHODS TO NEUTRON AND GAMMA-RAY EXPOSURES

Detector Configuration (numbers refer to front and back screen thicknesses, in mils)	Film Type	-A- Total Neutron Exposure to Obtain Total Film Density of 1.5[a] n/cm^2	-B- Total Gamma[b] Exposure to Obtain Total Film Density of 1.5[a] mr	Ratio A/B, $\frac{n/cm^2}{mr}$ $(\times 10^{-4})$
B^{10}-loaded Scintillator	F	8.8×10^5	150	0.59
B^{10}-loaded Scintillator	No Screen	2.7×10^6	366	0.75
10-10 Rhodium	No Screen	2.3×10^7	347	6.7
20-30 Indium	No Screen	3.1×10^7	363	8.5
10-20 Cadmium	No Screen	3.4×10^7	349	9.8
18-18 Silver	No Screen	4.5×10^7	354	12.7
6-10 Gold	No Screen	1.2×10^8	263	46.4

[a] See text for method of calculation. Neutron intensity used was 9.36×10^4 n/cm^2-sec.

[b] Gamma source used was cobalt-60. Gamma intensity was approximately 500 mr/hr.

The final column in Table III, the ratio of the neutron exposure required for density 1.5 to the similar figure for gamma exposure, is a figure of merit for relative neutron-gamma exposure for these film techniques. The lower the figure, the more useful the method will be for detecting neutrons in a mixed neutron-gamma radiation field. On this

basis, then, the scintillator is very good,* followed by rhodium, indium, cadmium, silver, and gold. In other words, in a mixed radiation field, the best direct-exposure detector (of those tested) to use for neutron radiography is the B^{10}-loaded scintillator and type F film. It is this detector which would show the least relative response to gamma radiation. Since the primary advantage in employing neutron radiography is the difference in relative absorption of materials to neutrons and to X or gamma radiation, it is desirable that the detector used have a relatively low response to gamma radiation, in order that the desired neutron image not be masked by the gamma image.

An imaging method which can be used to eliminate completely the effects of any gamma radiation in the neutron beam is the radioactive-foil technique, discussed by Kallmann and used by Peter. In this method only a screen of a material which becomes radioactive is exposed to the neutron image. This radioactive, image-carrying screen is then transferred to photographic film. One problem with this technique is that the neutron intensity should be relatively high in order to produce usable activities in the detecting screen materials. In the neutron flux available for this study (10^5 n/cm^2-sec), rhodium and silver did not yield useful film densities even if irradiated and allowed to remain on the film for periods greater than 3 half-lives for each operation. Indium does appear useful, but requires combined exposure and transfer times of the order of hours. Long times are also required in the case of gold foils.

These results on the usefulness of the radioactive screen materials for the transfer imaging method follow directly from the information contained in Table I. In the case of silver, the highest cross section is associated with a 24.2-sec activity. The high cross section indicated for rhodium is also associated with a short half-life activity (44 sec). These activities approach a saturation value in a very short time, and likewise an appreciable percentage of the decay activity is obtained in a short time. In the case of the transfer imaging tests reported here, a large percentage of the activity is lost in the 30 sec required to move the radioactive, image-carrying foil from the neutron beam to the film. Even more importantly, relatively little activation of the foil can occur before a saturation value is approached. Therefore, an appreciably higher neutron flux seems to be necessary in order to use rhodium or silver successfully for a transfer imaging technique.

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*Recognize that these figures for the scintillator may change for other exposure situations because of reciprocity-law failures. The data for this comparison were obtained with approximate intensities of 10^5 n/cm^2-sec for thermal neutrons and 500 mr/hr for Co^{60} gamma radiation. The comparison is valid only for this situation. Nevertheless, the trend should be true over a wide range of similar values.

E. Resolution Comparisons for the Various Detecting Methods

With the neutron beam available for this study, the transfer imaging technique has really not been needed to obtain a useful neutron radiograph (because the gamma radiation in the imaging beam was very low). However, the transfer method has been found to be very useful in the matter of improved image resolution over that of the direct-exposure method. Part of this improvement comes about because, as a matter of convenience in alignment, it has been found necessary to use a single converter screen in the transfer method. However, direct exposures taken with single screens also show somewhat poorer image sharpness than those taken by the transfer method. The poorer resolution yielded by the direct-exposure method then seems to be the result of scatter within the film emulsion and backing material.* A comparison of direct exposures and a transfer neutron radiograph is shown in Figures 8 and 9. The object pictured in each case is a 0.100-in.-thick cadmium bar containing several drilled holes varying in diameter from 0.040 (#60 drill) to 0.0135 (#80 drill) in. Figure 8 is a transfer picture taken on KK film,** using 0.020-in. indium as the screen material. Figure 9 shows direct-exposure radiographs for various film-screen methods. The resolution in all these pictures appears to be useful, except in the case of the direct exposures with cadmium and gold screens.

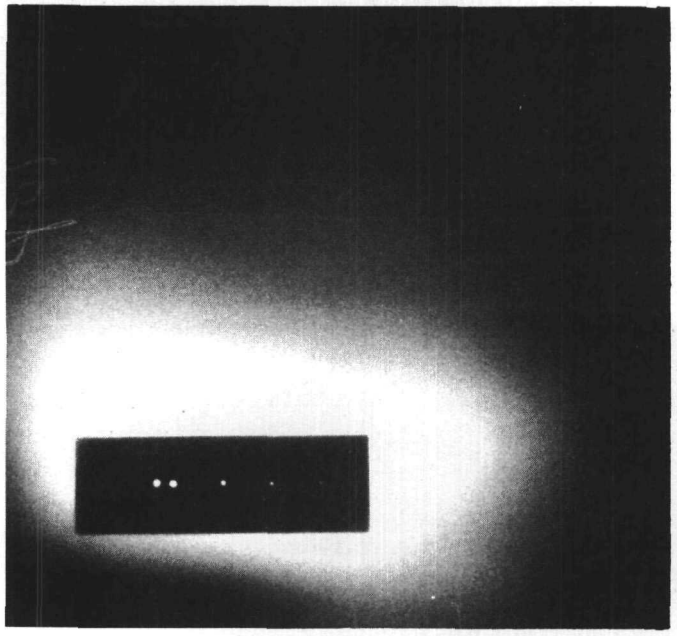

Figure 8

A transfer neutron radiograph of a cadmium test piece containing a series of holes varying in diameter from 0.040 (#60 drill) to 0.0135 (#80 drill) in. The radiograph was taken by exposing 0.020 in. of indium to the neutron beam for 13.5 min (in a maximum flux of 3×10^5 n/cm-sec) and then transferring the indium to KK film overnight.

*Recent data indicate that hard gamma radiation from the (n, γ) reactions in these converter materials is responsible for much of the image unsharpness obtained on a direct exposure.[25]

**All radiographs shown (except Figures 9 and 10) are positive prints. White areas are areas of high exposure.

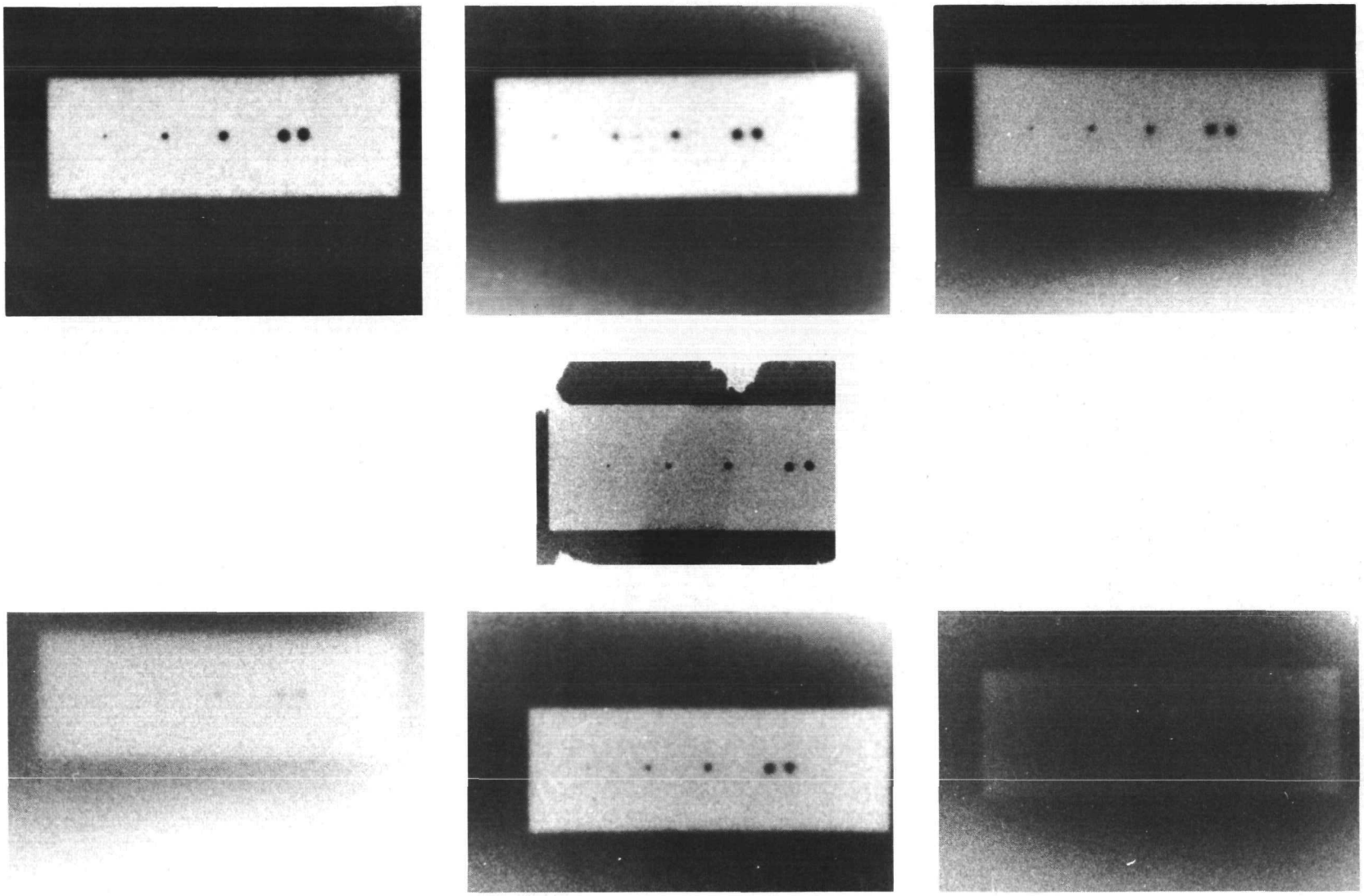

Figure 9

This is a series of direct neutron exposures of the same object described for Figure 8. The screen thicknesses used were those given in Table II. The intensifier materials used were: left to right: top row: B^{10} scintillator, rhodium and indium; middle row: gadolinium (1-2 screens); bottom row: cadmium, silver and gold.

To date, resolution comparisons of these various methods have been hampered by the difficulty of visual comparisons. A recording densitometer has recently become available for more objective studies of resolution. Preliminary results with this instrument confirm the visual impressions that transfer pictures yield improved resolution over direct-exposure radiographs. Exceptions to this rule involve direct-exposure pictures using the B^{10}-loaded scintillator and gadolinium screens. The B^{10} technique appears to yield resolution qualities about equal to those obtained by the transfer method, and direct-exposure radiographs taken with gadolinium screens appear to yield the best sharpness qualities of any of the methods studied.

In the case of cadmium, the poor image resolution can be explained by the fact that the Cd^{113} (n,γ) Cd^{114} reaction yields gamma radiation of very high energy. Several gamma energies are found from this reaction, including energies up to about 9 Mev.[26] The inherent image unsharpness obtained with X or gamma radiography with radiation of such energies is known to be high (in the order of 0.5 mm).[27]

It should be pointed out that the resolution achieved with a neutron image detector may be appreciably influenced by the materials contained in the object being inspected and by the materials in the vicinity of the detector, as well as by the detector itself. As was indicated in the discussion on neutron absorption, scatter of the neutrons by the absorbing object may be a major portion of the neutron absorption for a particular material.[28] For example, this is true of hydrogen, which has a total mass absorption coefficient for 1.08-Å neutrons of 48.5 cm^2/gm, of which 48.4 cm^2/gm is due to scattering.[2] This is an extreme example, but, in general, scattering accounts for much slower neutron absorption, particularly among the light nuclei.[6] This scattered radiation, whether it originates in the inspection sample, the detector, or the material in the vicinity, can contribute to overall fog and to decreased image resolution.

VI. DISCUSSION OF EXPERIMENTAL STUDY OF PHOTOGRAPHIC NEUTRON DETECTORS

The experimental study has yielded information on the relative speed, the relative response to neutrons and gamma radiation, and the qualities of image resolution that can be obtained with several photographic neutron-image-detector techniques. From the data presented, it must be concluded that the B^{10}-loaded scintillator, used in a direct-neutron-exposure method with type F film, has the most promise. This method has yielded the best speed, the best relative neutron-gamma response, and good resolution qualities. One problem with this detector is that the phosphor used in it (ZnS:Ag) tends to agglomerate. The nonuniform distribution of the phosphor in the screen has yielded mottled pictures. This is demonstrated in Figure 10, which shows 2 views of the neutron beam. The figure at the left was

taken with the scintillator and No Screen film. The lower corner of the beam in each case was absorbed by $\frac{1}{4}$-in. steel. The differences in the pictures, namely, the mottled areas of the scintillator picture, are caused by the agglomeration of the ZnS in the scintillator.*

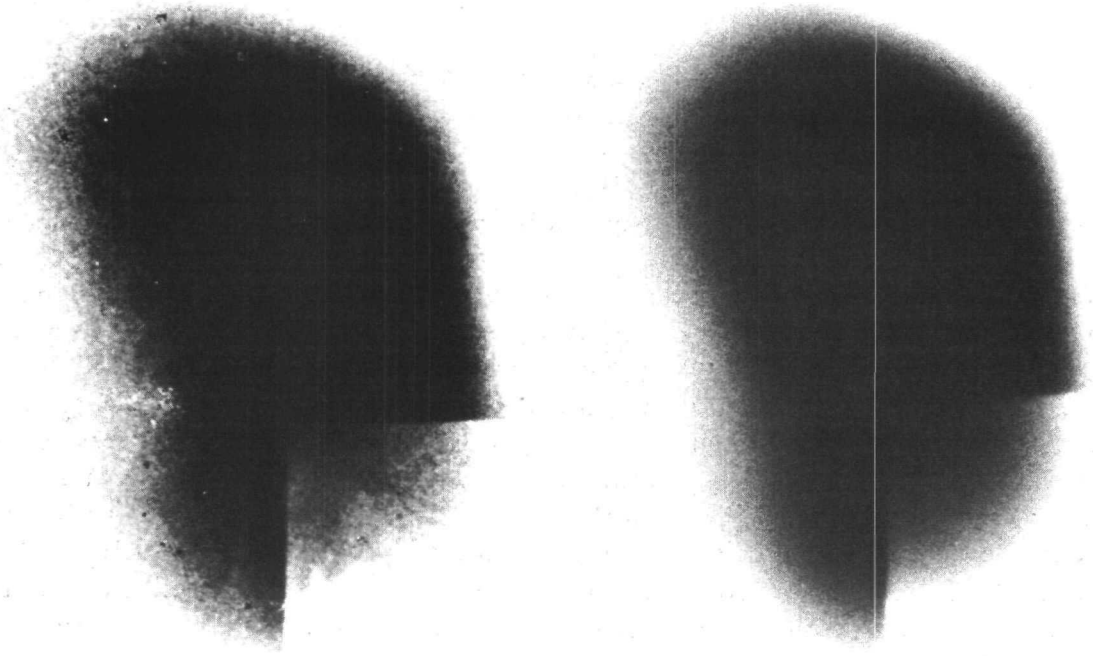

Figure 10

Shown are two views of the neutron beam detected by direct neutron exposures using 20-30 indium (right) and a B^{10} loaded scintillator (left), each using No Screen film. One edge of the beam in each case was absorbed by $\frac{1}{4}$-in. steel.

Of the other materials tested, rhodium appears to be the most useful for the direct-exposure method. This material has shown the best speed and relative neutron-gamma response of the metal screens. Indium, cadmium, and silver must be grouped next as far as speed and relative neutron-gamma response are concerned, but the poor resolution obtained with cadmium will limit its usefulness. As far as resolution is concerned, the excellent results obtained with gadolinium should make this material very useful.

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*An attempt is presently being made to prepare a more uniform scintillator. This work is being done through the cooperation of W. R. Allison, Nuclear Enterprises, Ltd. Lithium scintillators (Reference 29) are also under study.

In the neutron flux available for this study, indium and gold are useful in the radioactive transfer imaging method, whereas rhodium and silver are not useful because the short activities associated with these materials are limiting factors. The transfer method appears to offer an advantage other than the elimination of gamma detection in the neutron radiograph, in that improved resolution can be obtained by this method.

VII. PRELIMINARY RESULTS OF SOME RECENT EXPERIMENTAL WORK

As part of this program, it is planned to investigate other neutron sources for application to neutron radiography. The things sought in other sources include a more uniform neutron flux over a larger inspection area, and beams of other neutron energy. A start in this direction has been made in an initial trial with a Van de Graaff generator. The $Li^7(p,n)Be^7$ reaction was used at a voltage of 2.5 Mev and a proton current of 10 μamp. The moderating configuration used in this trial was unsatisfactory (6 in. of water in front of the thick lithium target), and further efforts are planned using improved moderating configurations. In this initial trial, the best imaging results were obtained with the fast neutron beam, moderated only by the thick target and the air path. The effective flux was relatively low (about 10^4 n/cm^2-sec at 12 in. from the target) and seemed to follow the inverse-square law over a range from 6 to 24 in. At a 36-in. distance, the effective flux was higher than anticipated, and this rise may have been caused by backscatter. Best resolution results were obtained at the 6-in. distance, indicating that scatter of the beam in the air path may be causing some difficulty.

Detector-wise, it is planned to investigate several rare earth materials for their possible usefulness as intensifier screen materials. Some preliminary results with gadolinium indicate a relative speed about equal to silver (based on data in Table II) and very good resolution qualities. The good resolution probably results from the fact that very thin screens can be used (0.001 and 0.002-in. screens were used in this preliminary test) because of the very high neutron cross section for gadolinium. Materials being prepared for use at the present time include samarium and dysprosium in addition to gadolinium.

VIII. APPLICATIONS

Relatively little has been done to apply neutron radiography as an inspection technique. The stress in this investigation has been placed on a better understanding of the techniques which can be used to obtain neutron images. However, in the course of this work, several pictures have been taken which indicate possible areas of application.

The high neutron absorption of hydrogen makes neutron radiography of potential value in biological studies. Figures 11 and 12 indicate the type of neutron radiographs which can be obtained for biological samples with present techniques. The radiographs show primarily areas of hydrogen concentration in a grasshopper (Figure 11) and a jaw bone and teeth sample (Figure 12). Although such pictures appear to be of general interest, specific application of neutron radiography in the field of biology will probably require techniques yielding much improved resolution.

Figure 11

This direct neutron exposure of a grasshopper was taken using 20-30 indium screens and AA film.

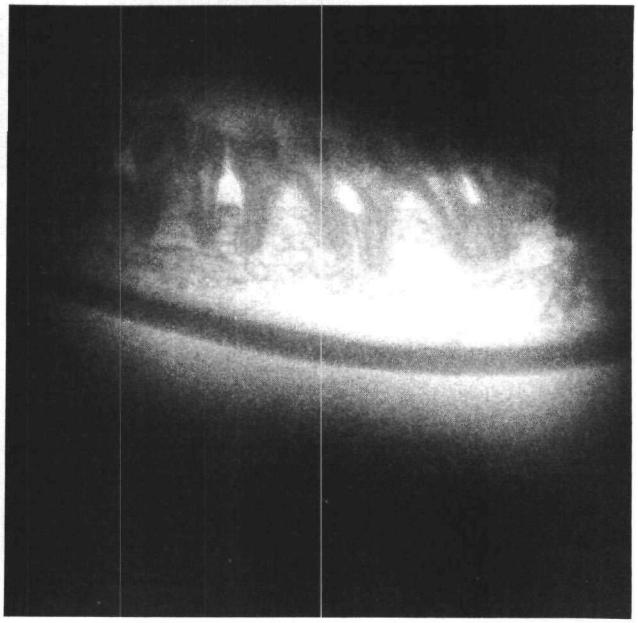


Figure 12

This neutron radiograph of a jaw bone was taken by a transfer technique using 0.020-in. indium and type KK film. Neutron exposure was 13.5 min; transfer time was overnight.

Even with the resolution presently achievable, neutron radiography appears ready for application to the "unusual" problem, to the problem which takes advantage of the differing absorption between neutrons and X rays. Figure 13 illustrates this point by showing the image of a sheet of rubber $\frac{1}{32}$ in. thick on $\frac{1}{4}$-in. steel. Other inspection applications which have been suggested by the absorption differences between neutrons and X rays include the determination of the uniformity of sintered boron carbide deposits in boron carbide-zirconium elements, the location of zirconium hydride deposits in single-crystal zirconium, and the inspection of metals in general for hydrogen localized areas.

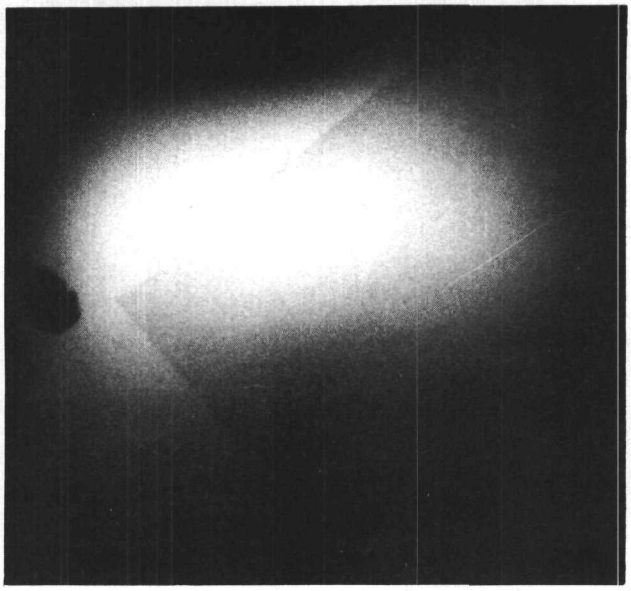
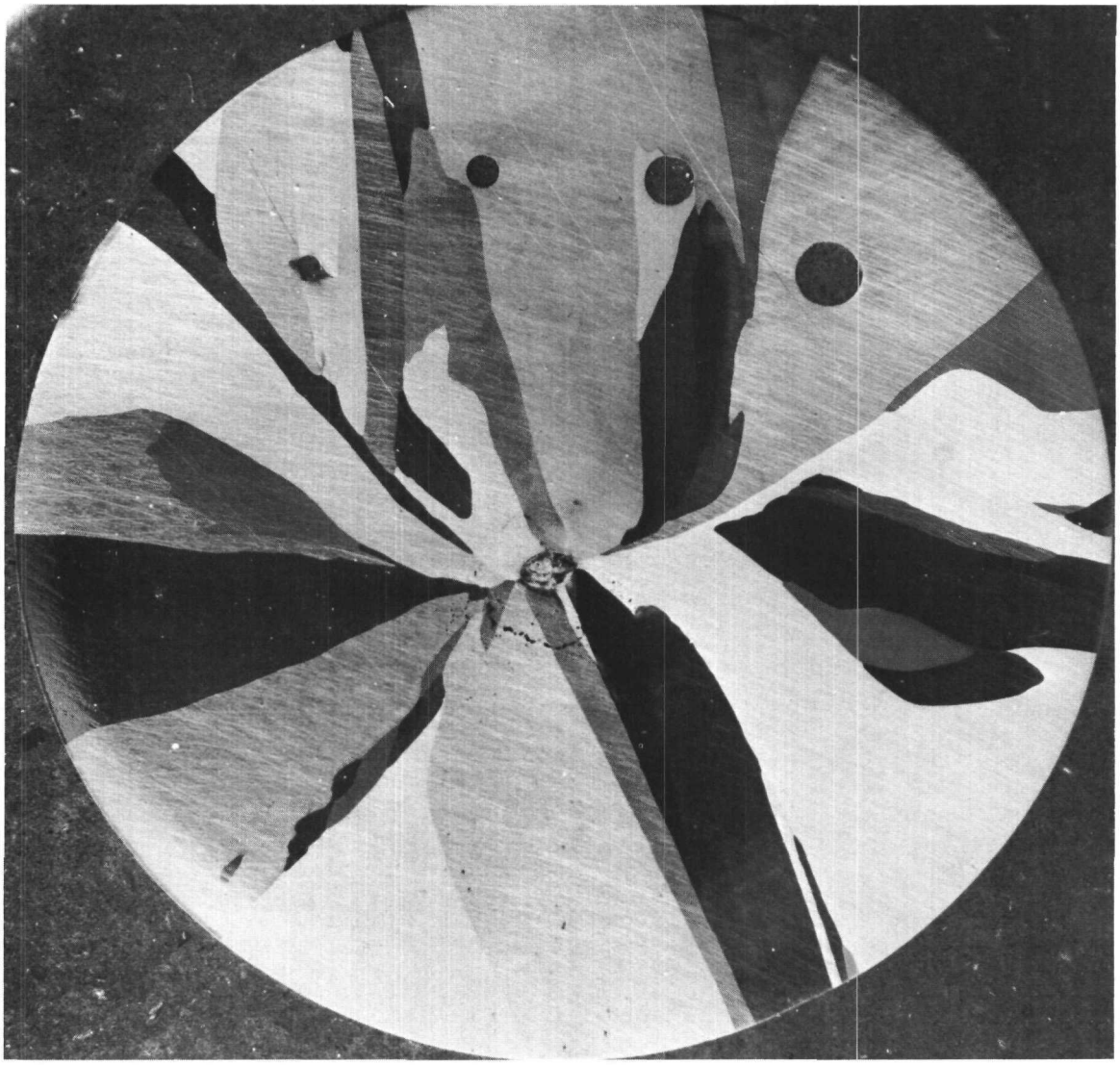
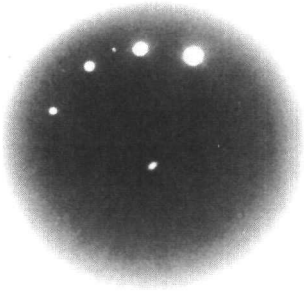


Figure 13

This neutron radiograph was taken by a transfer technique using a 0.020-in. indium and KK film. The entire beam was filtered through $\frac{1}{4}$-in. steel. The square image shown is that of $\frac{1}{32}$-in. rubber. The oval shaped absorbing object at the point of the rubber square is a piece of 0.001-in. gadolinium.

Another interesting possibility is demonstrated by Figure 14 which shows 3 different views of a 0.100-in.-thick nickel disk which was cut from a cast rod. The upper picture, taken at 4.8 X magnification, (and reproduced here at 75% reduction) shows an optical picture of one surface of the nickel disk after polishing and etching. The lower pictures are an X ray (left) and a neutron radiograph (right) of the same disk. The surface crystal structures are well shown in the metallographic sample picture. The X ray shows, upon close examination, some of the boundaries between these different areas of crystal orientation. The neutron radiograph displays the crystal orientation differences within the sample. The differences in neutron transmission within the sample follow from the work of Beyer, Nix, and others[30,31] who have demonstrated in their work on materials, such as iron, nickel, and copper, that single crystals have greater neutron transmission than polycrystalline samples and that larger grain size areas have higher neutron transmission. This ability to picture internal grain size and crystal orientation differences may be of appreciable value in some metallurgical studies.





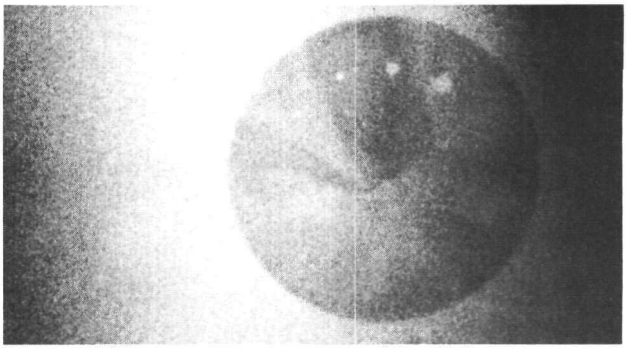


Figure 14

This figure shows three views of a 0.100-in. thick nickel disk cut from a cast rod. The large picture (taken at 4.8 X magnification and reproduced here at 75% reduction) shows surface crystal orientation after polishing and etching. The lower pictures are an X ray (left), showing only the boundaries between different crystal orientation areas, and a neutron radiograph (right), showing internal crystal structure.

IX. CONCLUSIONS

The neutron radiographs presently being produced can be obtained in reasonable exposure times and can yield resolution qualities generally equivalent to those obtained with gamma radiography. Both these properties seem well enough advanced so that neutron radiography can now be considered for application as an inspection method. The problems to which neutron radiography may be applicable should take into account the absorption differences between neutrons and X radiation, because the advantage which the use of neutron radiography may have in any specific problem will probably be associated with these differences.

X. ACKNOWLEDGEMENTS

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