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ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois

MASTER

NEUTRON RADIOGRAPHY - A NEW DIMENSION IN RADIOGRAPHY*

by

Harold Berger
Metallurgy Division

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ABSTRACT

In keeping with the metals technology theme of the Pacific Northwest Metals Conference, this discussion will stress the potential advantages of neutron radiographic inspection methods for metals. Primarily these advantages are associated with the relative differences in the absorption of materials for neutrons as compared to gamma or X-radiation. These absorption differences can be used in many cases to obtain improved discrimination between several materials which may be present in an object under inspection. In addition, because of the higher transmission of thermal neutrons through most heavy metals, there are advantages to be gained in employing neutron techniques for the inspection of such materials. A comparison of neutron and other radiographic inspection methods for the inspection of uranium, in particular, is given. It is shown, using available neutron sources, both radioactive and accelerator, that significant gains in exposure time can be achieved over most other radiographic equipment. Using a reactor neutron source, the speed of the neutron inspection method would surpass that of a 22 MEV betatron for uranium radiography. It is also shown that these gains can be achieved while maintaining high image quality, since contrast sensitivities in the order of 1 to 2% have been obtained.

INTRODUCTION

The technical literature for the years prior to 1960 contains relatively few publications on the subject of neutron radiography.[1-4] Although the advantages of the use of neutron radiographic inspection techniques were known and demonstrated during those early years, the fact that neutron sources suitable for such work were not easy to obtain imposed a severe limitation on extended work in that field. More recently, an increasing number of organizations have expressed interest in neutron radiography and are contributing to improved knowledge concerning many of it's characteristics and capabilities.[5-13] This increased activity in the field of neutron radiography has probably been brought about by the increased availability of neutron sources, and by the knowledge that difficult current and future inspection problems will require new and unusual inspection methods.

The advantages of neutron radiographic methods for inspection have been extensively discussed previously.[1,3,4,5,14] Primarily, the advantages are associated with the fact that the relative absorption of materials is different for neutrons than it is for X-radiation. Such absorption differences can be used in many cases to obtain improved detection discrimination between materials. Another advantage is that, because of the relatively low absorption of thermal neutrons in most heavy materials, neutron radiography of large thicknesses of such materials frequently offers the prospect of substantial exposure time reductions over other available techniques. It is primarily with this last advantage that this paper will be concerned.

METHODS FOR NEUTRON RADIOGRAPHY

The experimental procedures employed in the study described here are the same as have been reported previously.[8,14] The neutron beam being used is obtained from a crystal spectrometer used primarily for neutron diffraction[15]

studies at Argonne's CP-5 reactor. The parallel neutron beam is essentially monochromatic, with a wavelength of 1.05 A, and is relatively free of gamma radiation.[8,14] The neutron intensity has been determined by gold foil activation methods to be 3×10^5 neutrons/cm^2-sec. This maximum intensity is obtained only over a relatively small central, oval shaped area, approximately 1" x 1/2". The beam size for radiographic purposes is in the order of 3" in diameter.

The detection techniques employ conventional X-ray film used with materials which emit photographically detectable radiation upon neutron bombardment. Two distinctly separate detection methods, called the direct exposure and transfer methods, have been utilized. The direct exposure method can be used with all of the materials which have proved useful for photographic detection of neutrons because the film and screens are exposed to the neutron image together. Therefore the film is present to record any prompt radiation from the screens and is also present to record radioactive decay radiation, including that from short half-life activities.

The transfer technique employs a screen of some material which becomes conveniently radioactive when exposed to neutrons. Gold, indium, and more recently dysprosium[16] have been particularly useful in our studies. The screen, carrying a radioactive image after neutron exposure, is placed on photographic film and the exposure takes place using only the radioactive decay radiation from the screen. This technique is generally slower than the direct exposure method but it does have the advantages that radiographs having improved image sharpness and contrast[8] can usually be obtained and that the resultant radiograph will not be influenced by radiation other than neutrons which may be either in the beam, or introduced into the beam by the radiographic object.[14]

A number of materials which appear useful for photographic neutron detection methods have been studied. As a result of these studies,[8,16] several detection methods have been recommended for use with neutron radiography.[14] These recommendations for direct exposure neutron radiographic detectors are repeated in Table I. A few revisions have been made in these methods, on the basis of information obtained since the previous recommendations. For applications requiring good image sharpness, the gadolinium screen method is particularly useful since the radiographs obtained using gadolinium have yielded the best image sharpness qualities of any method used in this investigation thus far.

Results obtained with many of these detection methods, both direct exposure and transfer, will be further discussed later in this report.

Before closing this section on methods for neutron radiography, some other techniques which appear potentially useful, should be mentioned. A number of detection methods which may be regarded as nonphotographic have been proposed and studied. A review of several of these, including solid state detectors and spark counters, has been given by Watts.[22] A number of suggestions for neutron sensitive pick-up tubes have also been given, particularly in the patent literature.[23] Still another detection method, namely the use of a sensitive scintillator as a fluoroscopic screen has been suggested.[9] It is difficult to predict at the present time how useful these methods may be for general neutron inspection applications. However they should be considered in setting up a neutron inspection facility.

As far as neutron sources are concerned, it is usually true that neutron radiographic equipment is shown next to a nuclear reactor. Although a reactor has much to recommend it, other sources can be useful. The low neutron yield limitation imposed by most available radioactive sources may no longer be a valid objection to the use of such sources, now that a high yield antimony-

TABLE I

Recommended Photographic Detection Methods for Direct Exposure Thermal Neutron Radiography

Method	Characteristics
Scintillator[a] and type F or similar X-ray film, or Polaroid film.	This is a very high speed method. Detectable images[b] have been reported with thermal neutron exposures in the order of 400 neutrons/mm^2. The disadvantages of this method are that images are relatively grainy and that reciprocity-law failure responses[c] limit the latitude which can be obtained. A third disadvantage is that contrast sensitivity capabilities appear to be in the order of 6 to 10%.
Metal screen methods using 0.0005" gadolinium as a back screen, or double metal screen methods[d] using gadolinium (0.5-2) screens, rhodium front and gadolinium back screens (10-2), or rhodium (10-10) screens.	Slower speed methods[e] which yield improved image sharpness and contrast sensitivity are given in order of decreasing ability for image sharpness.

[a] Scintillators used are B-10 and Li-6 loaded. See references 8, 9, 17, 18, 19, and 21.

[b] Reported neutron exposure was for a film density of 0.5. See reference 9.

[c] The speed of response of the scintillator-film detector tends to decrease as the neutron intensity is lowered beyond a certain limit. See references 20 and 21.

[d] Numbers after double screens refer to thicknesses of front and back screens respectively in thousandths of an inch. The rhodium-gadolinium screen combination is the fastest metal screen configuration studied in our tests. See reference 21.

[e] The speed of these methods will depend upon the film used. For type KK film, total exposures in the order of 3 to 10 x 10^4 thermal neutrons/mm^2 are needed for a film density of 0.5.

beryllium source has been described by Hennelly.[24] Further improvements in radioactive sources, employing transuranic elements,[22] hold additional promise for the future. A number of accelerators also appear potentially useful as sources for neutron radiography and several investigations of neutron radiography using such sources have been,[1,2] and are being made.[6,16] A review of available sources, including neutron yields and approximate installation costs has been given by Burrill and MacGregor.[25] A number of the accelerators listed are capable of supplying a thermal neutron flux in the order of 10^7 neutrons/cm^2-sec. within a moderator. An external thermal neutron beam useful for neutron radiography should be available from such a source. The installed costs listed for a number of these sources are in the order of 20 to 30 thousand dollars which puts them in a price range competitive with high voltage X-ray equipment. For the particular task of inspecting heavy metals, it will be seen that the accelerator neutron source would have an exposure time advantage for many heavy metal inspection situations. A final comment concerning neutron sources for radiography of heavy metals should be made which involves gamma radiation in the imaging beam. It has been previously pointed out[8,14] that gamma radiation in the imaging beam can present problems for general neutron radiographic inspection. If one were attempting to observe hydrogen concentrations within metals, for example, a gamma image superimposed on a neutron image would probably obliterate the desired information. The information could be obtained in this case by employing transfer exposure methods. For the specific problem of inspecting heavy metals however, gamma radiation in the imaging beam may not be a problem, even for direct exposure neutron radiographic methods. The gamma radiation in the neutron beam may even contribute to further exposure time reductions. This is one additional reason why accelerator and radioactive neutron

sources can be recommended for this application. The gamma radiation which results from moderation, and other neutron bombarded objects in the beam, need present no serious problem for heavy metal inspection.

NEUTRON RADIOGRAPHY OF HEAVY METALS

Radiographic inspection of heavy metals frequently involves long exposures using X- or gamma ray equipment.[26,27] In extreme cases, involving large thicknesses and/or equipment limitations, the inspection capability is limited by excessive scatter.[28] Both these problems are much reduced, if thermal neutrons are employed for the radiographic inspection, because of the appreciable increase in transmission of the radiation beam through the object under inspection. A comparison of radiation transmission through natural uranium for a number of different radiations, is shown in Figure 1. Note the relatively high transmission of thermal neutrons through uranium, a rather typical characteristic of thermal neutrons and most heavy metals. Similar comparisons could be made for materials such as lead and bismuth in which the relatively high transmission of thermal neutrons would be apparent. This low absorption of thermal neutrons in many heavy metals makes the use of neutron radiography attractive as an inspection technique.

In spite of this high transmission of thermal neutrons through heavy metals, one is inclined to feel that, comparing actually available equipments, the exposure advantage for neutrons would be slight because of low neutron yields of available neutron sources. In the author's opinion, this is not the case, and the exposure information contained in Figure 2 helps to illustrate this.

Shown in this figure are exposures versus natural uranium thicknesses for a number of X- and gamma ray sources and for several neutron radiographic

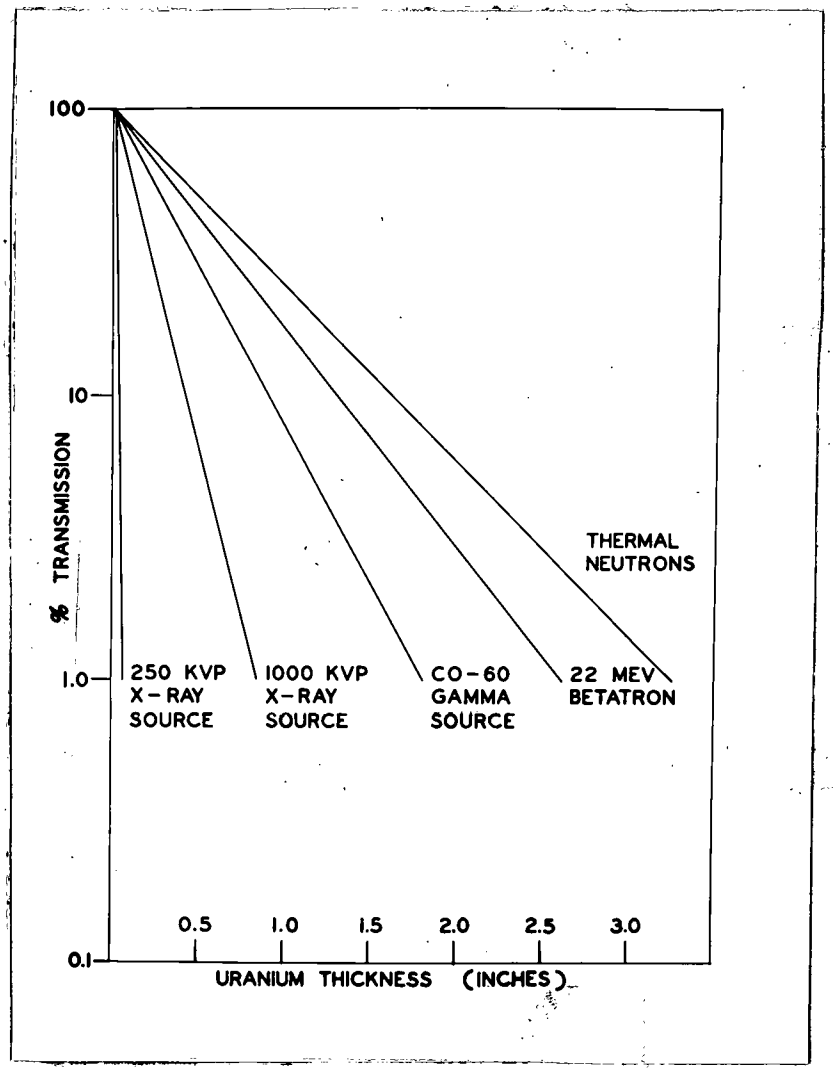

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FIGURE 1

Shown in this graph are the percentages of radiation from various sources which will be transmitted through different thicknesses of natural uranium. Gamma and X-ray data were obtained from reference 26. A half-value thickness of 0.5 inches was used for the neutron data. This value is in reasonable agreement with the absorption data given by Thewlis[3,4] and as determined by Argonne experiments.

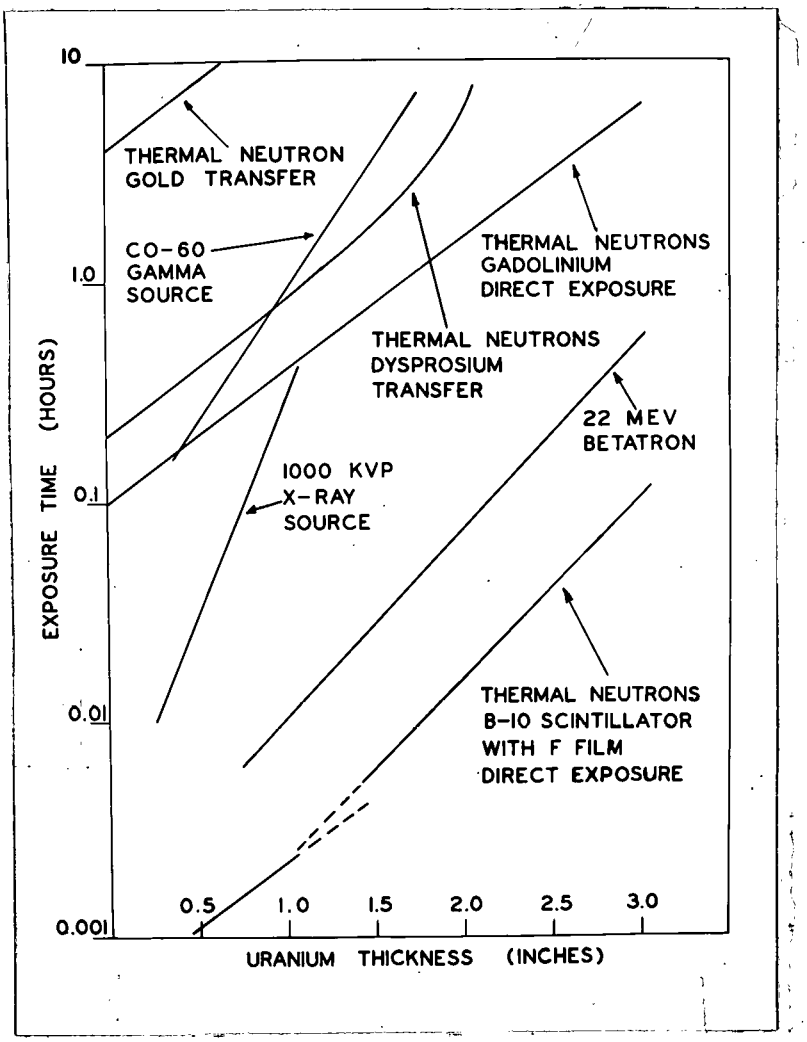

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FIGURE 2

Exposure times versus natural uranium thicknesses are shown for a number of radiation sources and for several neutron techniques. The radiation source data were obtained from reference 26 and were adjusted for Type AA film (using a speed factor increase of 2 times over Type A film). All data shown are for Type AA film except the scintillator technique, where Type F film is indicated on the curve. The cobalt-60 source was 25 curies used at a distance of 30 inches. A GE X-Ray 1000 KVP unit was used with the transmitted beam, 3 ma, 72 inches distance. The betatron was operated at an output of 100 R/min. at a meter and at a distance of 72 inches. The thermal neutron intensity used for all the neutron data was 3×10^5 n/cm^2-sec.

methods. The neutron data were all taken at a thermal neutron intensity of 3×10^5 neutrons/cm^2-sec. Thermal neutron intensities in the order of $10^5/cm^2$-sec should be available from sources having fast neutron yields in the order of 10^{10} neutrons/sec. A radioactive source having this output has been described in the literature,[24] and a number of accelerator sources[25] having such yields are commercially available. For those who may be fortunate enough to have a reactor available as a neutron source, thermal neutron intensities in the order of $10^7/cm^2$-sec should be possible. Intensities such as this, of course, would decrease the neutron exposures shown in Figure 2 by a factor in the order of 30 times, and would put them far ahead of most other available equipment.

As far as comparisons of inspection quality are concerned, the neutron techniques described in Table I and shown in Figure 2 are capable of detecting thickness variations in the order of 1 to 2%. This is true for all the methods discussed except the scintillator technique for which contrast sensitivities in the order of 6 to 10% were found. A neutron radiograph of a 3 inch thick natural uranium sample, approximately 2 inches square is shown in Figure 3. A thickness variation in the order of 1% was easily detected on the original radiograph.

Before progressing too far from the exposure information shown in Figure 2, however, there are a number of comments concerning the neutron detection methods, which should be made. Starting with the two slower neutron techniques shown, the transfer methods, it can be seen that the exposure curve for the dysprosium transfer exposure begins to curve upward for thicker uranium exposures. This occurs because once the dysprosium has been irradiated in a given neutron intensity for three half-lives, further exposure time produces little additional activity in the material. The half-life of dysprosium-165,

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FIGURE 3

This is a reproduction of a neutron radiograph of a 3 inch thick natural uranium sample. The radiograph was taken by a direct exposure technique using gadolinium screens (0.5-1) and AA film. The neutron exposure was 5 hours. The uranium test slab visible in the shadow of the uranium sample is 0.065" thick. The contrast sensitivity shown is 2.17%. On the original radiograph a thickness change of 0.033", representing a contrast sensitivity of 1.1% could be seen. This is a negative print, as are all the radiographs shown in this report. Dark areas are areas of high neutron exposure.

the isotope responsible for most of the dysprosium activity, is 2.3 hours. Therefore there is little to be gained in using dysprosium transfer exposures longer than about 7 hours. The curve for gold would eventually curve upward also, but this point is beyond the scale used in Figure 2. The half-life of gold-198 is 2.7 days.

The exposures for the direct exposure techniques are theoretically capable of being extended in a linear manner to accomodate whatever material thickness may be involved. This is essentially true, except for an inspection material such as uranium, which is radioactive[26] and emits radiation which will fog the film.[26,29] With type AA film and gadolinium screens, and with the natural uranium sample in contact with the cassette, exposures should be limited to about 8 hours to avoid excessive film background. If longer exposures seem to be required, lead screens can be used to lessen this effect.[29]

The other neutron exposure technique shown in Figure 2, the B-10 loaded scintillator, also involves a peculiarity which should be mentioned. Note that the exposure versus thickness curve for this technique changes slope in the region of 1 to 1.5 inches. Beyond a thickness of 1.5 inches, the exposure curve should continually curve upward slightly for increasing uranium thicknesses, although the straight line shown approximates the actual exposure situations. This change occurs because of a reciprocity-law failure response for the scintillator - F film detector.[21] The speed of response of this detector decreases beyond a certain intensity limit. The magnitude of this decrease is such that, irradiating the detector with the same total exposure at 3×10^5 neutrons/cm^2-sec, and at a neutron intensity reduced by 100 times, a relative film exposure reduction in the order of 8 times will be found at the lower intensity. Aside from somewhat complicating exposure calculations,[30] the influence of this reciprocity-law failure response is to decrease the latitude

of the inspection. If an exposure of an object containing a number of different thicknesses were set to record the thinnest section, for example, the neutron intensity transmitted through the thicker portion of the object might not be sufficient to be recorded, because of the decreased response of the detector for that intensity. Further studies of this aspect of neutron radiographic inspection are planned. Generally speaking however, the latitude for neutron radiographic inspection of a material such as uranium will probably be fairly high because of the large thickness for a half-value layer.

An example of a heavy metal inspection problem which was recently encountered in the author's Laboratory, is shown in Figures 4 and 5. The radiographic object is a pressed uranium oxide cylinder, 1 inch in diameter and approximately 2 1/2 inches high, containing a wax filled central cavity. The cylinder was prepared for use as a pycnometer.[31]

The object of the inspection was to locate the internal cavity so that a hole could be accurately drilled through the upper sloping wall to the top of the cavity. Of the radiographic equipment available for this inspection, a 250 KVP X-ray generator, a 5 curie cobalt-60 source and the neutron inspection facility, the information shown in Figures 1 and 2 indicates the superiority of the neutron radiographic method. The high contrast between the wax and uranium made the inspection relatively simple by neutron radiography. Satisfactory results could be obtained using either gadolinium screens (Figure 4) or the faster scintillator method (Figure 5).

CONCLUSIONS

The case for neutron radiographic techniques in the inspection of heavy metals is, in the author's opinion, a strong one. The problems of scatter and long exposure times which are frequently encountered in radiographic inspections of large thicknesses of heavy metals are much reduced using neutron

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FIGURE 4

The neutron radiograph shows a pressed uranium oxide cylinder containing a wax filled central cavity. Note the high neutron absorption of the hydrogen containing wax, and the relatively low neutron absorption of the 1 inch diameter uranium oxide. The nonuniformities of the uranium oxide mixture are also apparent. A 16 minute neutron exposure, using a direct exposure method with gadolinium screens and AA film was used for the radiograph.

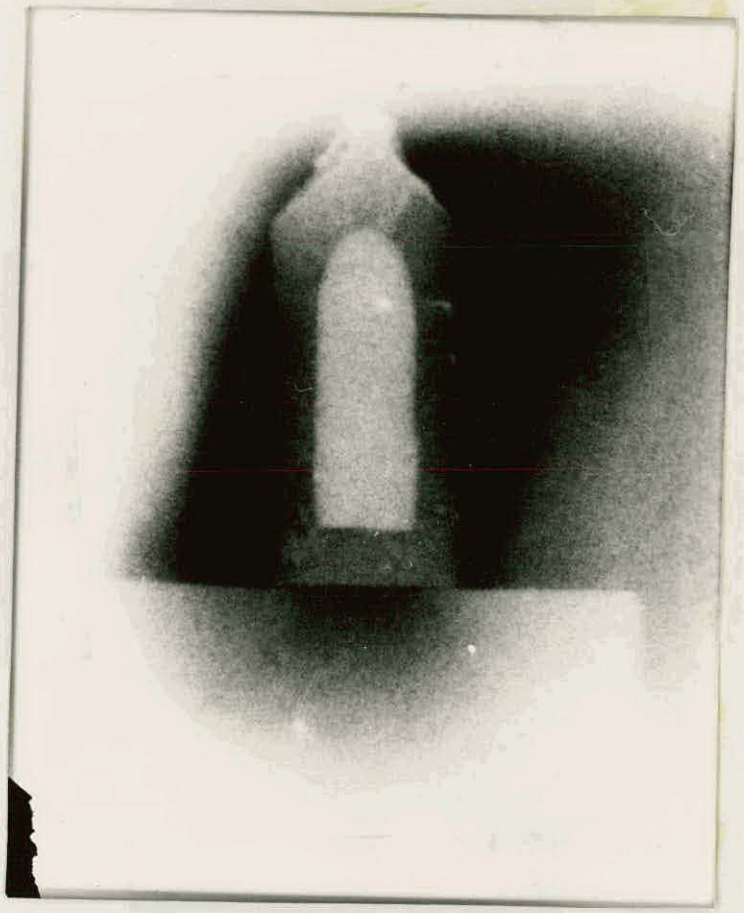

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FIGURE 5

This neutron radiograph, taken using a B-10 loaded scintillator with Type F X-ray film, shows the same object pictured in Figure 4. The neutron exposure time was 8 seconds.

radiography because of the increased transmission of thermal neutrons, as compared to X-rays, through materials such as uranium, lead and bismuth. This increased transmission of thermal neutrons through these materials does result in exposure time gains using readily available sources of neutrons and comparing the exposures to other existing radiographic equipment. Several accelerator neutron sources and at least one radioactive neutron source have been shown to be capable of supplying thermal neutron intensities sufficient for such inspections. The demonstrated contrast sensitivity in the order of 1 to 2% for neutron radiographic inspection of uranium, is indicative of the high quality of inspection possible using neutron radiography.

Results such as these, along with the large number of other application possibilities for neutron radiography[3,4,14,22] inspired the title for this report.[32] The potential usefulness of neutron radiography for heavy metals inspection, the location of materials such as hydrogen, lithium or boron within materials or assemblies, the inspection of hydrogen containing materials such as wood, paper, rubber,adhesives, and, for the same reason, in biological studies, as well as for untold other applications, truly makes neutron radiography a new dimension in radiography. The complimentary techniques of neutron and X-radiography greatly broaden the scope of radiographic inspection.

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REFERENCES

1. Kallmann, H., "Neutron Radiography", Research, 1, 254-260 (1947).
2. Peter, O., "Neutronen - Durchleuchtung", Naturforscher, 1, 557-559 (1946).
3. Thewlis, J., "Neutron Radiography", Brit. J. Appl. Phys., 7, 345-350 (1956).
4. Thewlis, J., "Neutron Radiography", Progress in Non-destructive Testing, 1, 111-126, Heywood, Ltd., London (1958).
5. Burrill, E. A., "The Scientific Application of Particle Accelerators to Nondestructive Testing", Argonne National Laboratory Report ANL-6346, pp. 176-196 (1960).
6. Criscuolo, E. L., and D. Polansky, "Progress Report on Neutron Radiography", Proceedings Missiles and Rockets Symposium, U. S. Naval Ammunition Depot, Concord, California, April, 1961, to be published.
7. Watts, H. V., and C. A. Stone, "An Optical Analog for Neutron Radiography", Trans. Am. Nuclear Soc., 4, 127-128 (June 1961).
8. Berger, H., "Comparison of Several Methods for the Photographic Detection of Thermal Neutron Images", J. Appl. Phys., 33, 48-55, (Jan., 1962).
9. Wang, S. P., C. G. Shull and W. C. Phillips, "Photography of Neutron Diffraction Patterns", Rev. Sci. Instr., 33, 126-8, (Jan., 1962).
10. Leavitt, W. Z., Watertown Arsenal, private communication.
11. Turkanis, M., Nuclear Materials and Equipment Corporation, private communication.
12. Kern, H. B., Martin Marietta Corporation, private communication.
13. Tochilin, E., U. S. Naval Radiological Defense Laboratory, private communication.
14. Berger, H., "A Discussion of Neutron Radiography", Nondestructive Testing, to be published. This publication gives many other references to work in neutron radiography.

15. Sidhu, S. S., L. Heaton and M. H. Mueller, "Neutron Diffraction Techniques and Their Applications to Some Problems in Physics", J. Appl. Phys., 30, 1323-40 (1959).

16. Berger, H., "Neutron Radiography - A Second Progress Report", Argonne National Laboratory Report ANL-6515, and Nucleonics, to be published.

17. Sun, K. H., P. R. Malmberg and F. A. Pecjak, "High Efficiency Slow-Neutron Scintillation Counters", Nucleonics, 14, 46-9 (July, 1956).

18. Stedman, R., "Scintillator for Thermal Neutrons Using Li^6F and ZnS(Ag)", Rev. Sci. Instr., 31, 1156 (1960).

19. Smith, H. G., "Use of Polaroid Film in Neutron and X-Ray Diffraction", Rev. Sci. Instr., 33, 128-9 (Jan., 1962).

20. Ehrlich, M. and W. L. McLaughlin, "Photographic Dosimetry at Total Exposure Levels Below 20 mr", National Bureau of Standards Technical Note 29, October, 1959.

21. Berger, H., "Photographic Detectors for Neutron Diffraction", to be published.

22. Watts, H. V., "Research on Neutron Interactions in Matter as Related to Image Formation", Armour Research Foundation Report ARF-1164-15 (Oct., 1961).

23. Kallmann, H. I. and E. Kuhn, "Neutron Image Converter", U. S. Patent 2,270,373, Jan., 1942. Several other patents are mentioned in reference 1.

24. Hennelly, E. J., "Intense Sb-Be Sources Make 10^{10} Neutrons/Sec", Nucleonics, 19, 124-5 (March, 1961).

25. Burrill, E. A. and M. H. MacGregor, "Using Accelerator Neutrons", Nucleonics, 18, 64-8 (Dec., 1960).

26. Tenney, G. H., "Nondestructive Testing of Uranium", ASTM Special Technical Publication No. 213, 36-48 (1956).

27. Elliott, D. E. and C. M. Knowles, "On The Radiographic Merits of the 1000 PKV Resonant Transformer and the Van de Graaff X-Ray Generators", Nondestructive Testing, 17, 205-209 (1959).

28. Droegkamp, R. E., "Van de Graaff Radiography of High Density Alloys", Nondestructive Testing, 13, 27-30 (Sept.-Oct., 1955).

29. Dutli, J. W. and D. E. Grimm, "Radiography of Materials Used in the Nuclear Energy Field", ASTM Special Technical Publication No. 223, 150-162 (1957).

30. Similar problems are encountered in X-radiography using fluorescent intensifying screens. See "Nondestructive Testing Handbook", edited by R. C. McMaster, Ronald Press, New York, 1959, 1, page 20:13.

31. White, D., Argonne National Laboratory, private communication.

32. The title was suggested by R. B. Socky, Hanford Atomic Products Operation, as appropriate to the theme of the Pacific Northwest Metals Conference and the Seattle World's Fair.