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NEUTRON RADIOGRAPHIC INSPECTION OF HEAVY METALS AND HYDROGENOUS MATERIALS*

by

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ABSTRACT

In this experimental study the possibility of using thermal neutron radiography for inspecting heavy metals and hydrogenous materials has been examined. The data include exposure curves, contrast sensitivities and an assessment of the influence of higher energy neutrons and interferring gamma radiation on image quality. It is shown that, in the case of homogeneous materials, neutron radiography presents definite advantages for the inspection of heavier metals such as uranium, bismuth and lead, and that the images obtained in such inspections are influenced very little by other radiation in the thermal neutron beam. This is somewhat less true for intermediate metals such as steel and tungsten, although in these cases too, some exposure time advantage can usually be gained. Nevertheless, neutron radiographic inspection of these intermediate materials may be limited to those cases in which some complication, such as radioactivity of the sample, is involved. Thermal neutron inspection of hydrogenous materials having a thickness greater than about an inch is not recommended and may be useful primarily in special cases such as one in which the light material is in some combination with a heavier, X-ray absorbing material.

I. INTRODUCTION

Neutron radiography has been a potentially useful inspection method since the early work of Kallmann and Kuhn,[1,2] and Peter[3] pointed out many application possibilities and some useful techniques. The later extension of that work by Thewlis and Derbyshire[4] gave additional application examples and produced several neutron radiographs of excellent quality. More recently, with the greater availability of neutron sources, neutron radiography has been investigated at several laboratories.[5-10]

This renewed interest in neutron radiography will undoubtedly bring about increased application of this inspection method. Among the potentially promising areas of application for neutron radiography are the inspections of heavy metals and hydrogenous materials. This paper will be concerned primarily with these possibilities.

After a brief description of methods used for neutron radiography, exposure curves for a number of materials will be given. The exposure curves are given for several exposure conditions and detection methods. Contrast sensitivity, and the influence of fast neutrons and gamma radiation on image quality are also discussed.

II. EXPERIMENTAL METHODS

All of the references cited thus far mention techniques which can be used for neutron radiography. The radiation sources mentioned include accelerator,[2,3,5] radioactive[7,10] and nuclear reactor[4,6,8,9] neutron sources. The detection methods discussed are primarily photographic, although some other types are described.[1,9]

In this study, photographic detection methods have been utilized with nuclear reactor neutron sources. The detection methods have been described

elsewhere[8,11] but a brief summary will be given here.

Since neutrons have relatively little influence on photographic emulsions, screens of material which convert the neutron image into one of radiation which is photographically detectable are used. These include prompt emission materials which emit alpha or gamma radiation immediately upon neutron bombardment, and potentially radioactive materials which make use of radioactive decay radiation to expose the photographic film.[a]

Examples of the first type of screen material include boron and lithium, alpha emitters which are usually used combined with a phosphor, and cadmium and gadolinium, which are gamma emitters. Rhodium, silver, indium, dysprosium and gold are examples of the radioactive screen materials. All of these materials can be used to detect a neutron image by exposing the screen and film together to the neutron beam. This has been termed the direct exposure method. This is a fast detection method but it has the disadvantage that the film also records interferring radiation which may be in the beam or be emitted from the object.[8]

A second detection technique is called the transfer method. In this technique, the photographic film is not exposed to the neutron beam at all. The neutron image is detected by a screen of potentially radioactive material which is then transferred to a film loaded cassette and allowed to decay. This method is slower than the direct exposure method but has the advantage that interferring gamma radiation in the beam, or emitted from the object, will not confuse the resultant neutron radiograph.

This ability of the transfer method to eliminate the influence of gamma

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[a] Commercially available X-ray film has been used in this work.

radiation from the final radiograph can be important in many cases, since one of the great advantages of neutron radiography is that the relative absorption of thermal neutrons and gamma radiation in materials is very different.[2,4] If one were trying to locate a hydrogenous material within a metal assembly, for example, it would probably be necessary to eliminate the effect of gamma radiation in the beam, since a gamma image superimposed on a neutron image would lessen the desired contrast. In inspecting a homogeneous material, however, the use of a transfer method might not be necessary since the gamma image might even be useful in reducing exposure time.

The neutron energy region which appears to be most generally useful for radiography is the thermal energy region[4,8] encompassing neutron energies up to several electron volts. Two nuclear reactor neutron sources have been utilized in this study, both of which supply a thermal neutron beam. One of these sources supplies a monochromatic neutron beam having an energy in the order of 0.05 ev. This beam, obtained from a crystal monochromator located at Argonne's CP-5 reactor, has been described previously.[8,11] The beam is essentially free of gamma radiation and covers an area about 3 in. in diameter. The beam intensity is 3×10^5 neutrons/cm^2-sec.

The second source supplies a more intense thermal neutron beam, 10^7 neutrons/cm^2-sec., over an area of 2 1/2 x 4 in. This beam, obtained directly from Argonne's Juggernaut reactor, contains gamma radiation having an intensity of about 50 R/hr. and some neutrons of higher energy. The cadmium ratio[12] with a 0.020 in. cadmium cover is 3.6:1.[b] A further description of this neutron radiography facility has been given in connection with a reported application study.[13]

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[b] The cadmium ratio was obtained by determining the neutron intensity using a bare gold coil and a 0.020 in. thick cadmium covered gold foil,

III. EXPERIMENTAL DATA

With one exception, the exposure curves to be presented here were obtained using the Juggernaut reactor facility. That neutron beam, containing significant intensities of gamma radiation and higher energy neutrons, is representative of neutron beams which might be generally available for radiographic purposes.

Exposure curves for tungsten, cold rolled steel, natural uranium and lead are given in Figs. 1 - 3, for three different detection methods. Exposure curves for bismuth would be similar to those shown for lead but with slightly reduced exposure times. Unless otherwise indicated, all the exposure curves in this report yield a total film density of 1.5 on Kodak Type AA film and all films were developed in Kodak Liquid Developer, 5 minutes, without agitation.

A transfer method using a 0.010 in. thick dysprosium metal screen was used for the curves in Fig. 1. A three half-life decay[c] was permitted before the films were developed. The exposure curves begin to curve upward as the neutron exposure times approach several half-lives, because the dysprosium activity is approaching saturation.(14)

A direct exposure method using a 0.0005 in. thick gadolinium metal screen as a back screen[d] was used for the curves in Fig. 2. An unusual feature observed in this set of exposure curves is the levelling off shown by the tungsten exposure curve. Some tendency for this is also shown on the curve for steel, although the effect is appreciably less than that shown for tungsten. This appears

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[c] The half-life of Dy-165 is 2.3 hours.

[d] This detection method has been shown to be a high resolution technique. See reference 15.

to be the result of prompt (n,γ) radiation emitted from the inspection material.[16] Note that the tungsten exposure curve for the transfer method, Fig. 1, eliminates that effect. Also shown on this set of exposure curves for direct exposure methods is a curve for steel taken with a direct exposure technique employing a 0.010 in. thick rhodium front screen and a 0.002 in. gadolinium back screen. This technique has been shown to be a fast metal screen detection method[17] having relatively good resolution.[15]

The very fast speed results shown in Fig. 3 do not, for the most part, represent actual experimental data since exposure times of less than a few seconds are not readily reproducible by our present methods. The exposure curves, however, do represent what might be done using a boron-10 loaded scintillator[8,11] and Type F X-Ray Film in a direct exposure method.

This last detection method, although very fast, does not yield as good contrast sensitivity as the methods represented in Figs. 1 and 2. The contrast sensitivity observed using scintillator detection methods has ranged generally from 6 to 10 per cent. The metal screen methods used either in a direct exposure or transfer method, on the other hand, have usually yielded contrast sensitivities in the order of 2 per cent. The penetrameter sensitivity curves shown in Fig. 4 show some typical results for a direct exposure method using a 0.0005 in. gadolinium metal back screen technique used for inspection of natural uranium. The penetrameter sensitivities indicated were for observation of the 2T hole, T being the thickness of the penetrameter and representing a percentage of the inspection material thickness.

Except for the slight peculiarity introduced by prompt (n,γ) radiation from some of the inspection materials, the results obtained with neutron radiographic inspection of heavy metals do not present complications. This is not the case for neutron inspection of relatively large thicknesses of hydrogenous materials.

The exposure curves shown in Fig. 5 for tempered, laminated Masonite demonstrate some of the problems encountered in inspecting relatively large thicknesses of hydrogenous material. Both these exposure curves tend to level off as the thickness of the inspection material becomes greater than about 2 in. The direct exposure curve (lower curve) tends to level because the gamma radiation in the beam is attenuated very little by the hydrogenous material and its effect becomes significant as the exposure times increase. Some effect on the direct exposure curve is probably produced also by the fact that some higher energy neutrons in the beam are moderated in the Masonite, and therefore are more likely to be detected than if the hydrogenous material were not there.

This same explanation appears to account for the levelling of the transfer exposure curve. As more Masonite is introduced, more higher energy neutrons are moderated and activate the detecting foil. Both this effect, and the influence of the beam gamma radiation on direct exposure detection methods can be eliminated if the neutron beam can be made free of gamma radiation and fast neutrons. This is demonstrated in Fig. 6. These exposure curves for the same inspection material were obtained using the gamma-free, monochromatic thermal neutron beam described in the preceeding section of this report.

The fact that the disturbing influences of gamma radiation and fast neutrons on the exposure curves for hydrogenous or other light material can be eliminated is encouraging. From a practical standpoint, however, a radiographic thermal neutron beam which did not contain significant intensities of higher energy neutrons and gamma radiation would be difficult to obtain. Even more important, the effects of these interferring radiations are not the primary problems encountered in the neutron inspection of large thicknesses of hydrogenous material.

The primary problem appears to be introduced by the fact that the large absorption of thermal neutrons by hydrogen is due primarily to scattering. This multiple scattering of the beam makes it difficult to detect small thickness changes in larger masses of hydrogenous material. In Masonite and similar materials, thickness changes of about 4 per cent have been detected for base material up to 1/2 in. In the range between 1/2 and 1 in., the thickness variation detectable increases to about 8 per cent. For material thicknesses greater than an inch, neutron radiography appears capable of detecting only 10 to 20 per cent thickness variations. This situation exists for both the neutron beams described in this work. The effect, therefore, does appear to be explained by the multiple scattering of thermal neutrons within large masses of material. The Masonite blocks used for these tests were 3 by 5 inches in cross section. It is possible that significantly improved results could be achieved with physically smaller test samples since there would then be less tendency for scattered neutrons to strike the detector.

IV. DISCUSSION AND CONCLUSIONS

Neutron radiographic examination of relatively large thicknesses of heavy metals can be used for inspections requiring 2 per cent contrast sensitivity. Although few problems appear to be encountered with imaging thermal neutron beams containing significant intensities of higher energy neutrons and gamma radiation, some decrease in contrast will undoubtedly result from prompt (n,γ) emission when using direct exposure techniques. At least part of this pronounced effect shown for neutron inspection of tungsten may be due to undercutting and scatter because of the small sample sizes[e] available. Lack of

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[e] Tungsten exposure curves were obtained by radiographing rod shaped samples having a 3/4 in. diameter. Boral sheet masks were used to keep most of the direct beam from striking the detector.

suitable test pieces for tungsten also made contrast sensitivity measurements difficult. The (n,γ) effect observed for the neutron inspection of steel, however, in which sample size and test pieces were not problems, was such that contrast sensitivities degraded from 2 per cent for 1 to 2 in. thick material to about 2 1/2 to 3 per cent at thicknesses in the order of 5 to 6 inches. In spite of this rather small change in observed contrast sensitivity, the prompt (n,γ) radiation emitted from steel must be fairly significant since the direct exposure curves in Fig. 2 show that faster results can be obtained for steel than for uranium.

It was first believed that the use of the faster rhodium-gadolinium screen method might contribute to a reduction of the prompt (n,γ) effect for the larger steel thicknesses since the photographic film could be present in the imaging beam a shorter time and would therefore record less of the unuseful prompt (n,γ) radiation from the inspection material. However, the approximate factor of two in speed improvement for this technique, as shown for steel thicknesses up to about 3 in. in Fig. 2, did not appear to be enough to yield a significant contrast sensitivity improvement for larger steel thicknesses. For larger thicknesses the two steel exposure curves shown in Fig. 2 begin to approach each other as the influence of the prompt (n,γ) radiation begins to become more important. There has not, therefore, been a detectable contrast sensitivity improvement gained by using the faster detection method.

Materials which yield less prompt (n,γ) radiation can be inspected without this difficulty. Natural uranium has been inspected to a thickness of 3 in. and lead to a thickness of 6 in. without encountering any problem. The useful inspection thicknesses of materials such as tungsten and steel may be limited by this effect, however.

Of course, transfer methods can then be used to eliminate this effect completely. Although in this case too, some limiting thickness will be encountered

when the metal foil used to detect the neutron image becomes saturated. For example, Fig. 1 indicates that, in a thermal neutron intensity of 10^7 neutrons/cm^2-sec., using a 0.010 in. thick dysprosium metal screen transferred to Type AA film and developing as indicated one could inspect tungsten up to about 2 1/2 in., steel up to about 3 in., and natural uranium up to about 3 1/2 in. These limits could be extended by using a higher neutron intensity, by using a faster film or perhaps by using a detecting foil having a longer half-life. In this latter case one might gain because longer exposure times could be profitably used. The high activation cross section for dysprosium, however, makes it unlikely that much would be gained unless a material with an appreciably longer half-life was employed. Even using gold, for example, with a 2.7 day half-life and exposure times up to about 8 days it is unlikely that these thickness limits could be appreciably extended for similar conditions. Such exposure times would present a number of practical difficulties.

In addition to these problems, the use of thermal neutrons to inspect hydrogenous material is further complicated by the facts that higher energy neutrons in the beam may be moderated by the inspection material and by the fact that the imaging neutrons themselves are subject to multiple scattering within the sample. It appears now that neutron radiographic inspection of hydrogenous material having a thickness greater than about an inch will not be capable of yielding useful contrast sensitivity.

To compare these results with other radiographic methods one must conclude that X-radiographic techniques have greater promise for relatively large thicknesses of hydrogenous material. Neutron radiographic inspection of such material may be useful for smaller thicknesses and particularly for situations in which the hydrogenous material is in some combination with heavier material which is relatively opaque to X-radiation. Another situation in which neutron inspection of hydrogenous materials (adhesives, for example) may be especially useful is

one in which they are combined with a material which yields a disturbing pattern on an X-radiograph, a material such as Fiberglas, for example. Radiographic tests with these materials indicate that the low neutron absorption of silicon and oxygen, combined with the high neutron absorption of hydrogen can be used to great advantage in an inspection problem involving hydrogenous material combined with a Fiberglas or similar material.

Neutron radiographic inspection of several of the heavier metals appears to offer definite advantages in exposure time over other radiographic methods. Inspection of 1 1/2 in. of natural uranium using 25 curies of cobalt-60, for example, would require about a 4 hr. exposure.[18] Fig. 2 indicates an exposure of less than 6 minutes would be required using a direct exposure neutron radiographic method. The use of the double metal screen technique using a rhodium front screen and a gadolinium back screen would yield an additional speed increase of about a factor of two over this result, with little loss in resolution. Even more pronounced speed comparisons between neutron and X-radiography could probably be made for such materials as lead, and bismuth, whose neutron absorption is less than that of uranium.

In any given situation the actual gains in exposure time will depend on material thicknesses and the type of equipment available. Generally speaking, however, there will be a significant exposure time advantage for the neutron technique when material thicknesses greater than an inch are involved. This will be true generally for neutron intensities in the order of 10^5 thermal neutrons/cm^2-sec, or more.[19] This neutron intensity can be obtained from nuclear reactor sources and also from accelerator[20,21] and radioactive[10,21] neutron sources.

In the case of steel, the exposure time comparisons are not quite as startling, although some exposure time advantage does remain, especially for thicknesses of a few inches. For example, X-radiographic inspection of 2 in. of steel requires about 6000 Mas at 250 KVP, or a time of about 10 minutes at 10 ma.[22] A comparable quality direct exposure neutron radiograph requires about half that exposure, and an additional factor of two could be gained using the rhodium-gadolinium screen combination mentioned earlier. Even more pronounced exposure comparisons are possible for tungsten. Nevertheless, because of prompt (n,γ) problems, neutron radiographic inspection of these materials may present advantages primarily in cases in which the inspection material is radioactive[10,13] or in other cases in which the situation is complicated.

V. ACKNOWLEDGMENTS

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FIGURE 1

Exposure curves for thermal neutron radiographic inspection of various thicknesses of tungsten (W), cold rolled steel (Fe), natural uranium (U) and lead (Pb) are shown. The detection method for these curves was a transfer technique using a 0.010 in. thick dysprosium metal screen. All transfers were to Type AA film for 3 half lives or more. In Figs. 1 through 3 the exposure curves for bismuth (not shown) would be similar to those for lead but would require slightly s neutron exposure time for each sample thickness. The thermal neutron intensity for these ves, and for Figs. 2 through 5 was 10^7 neutrons/cm^2-sec.

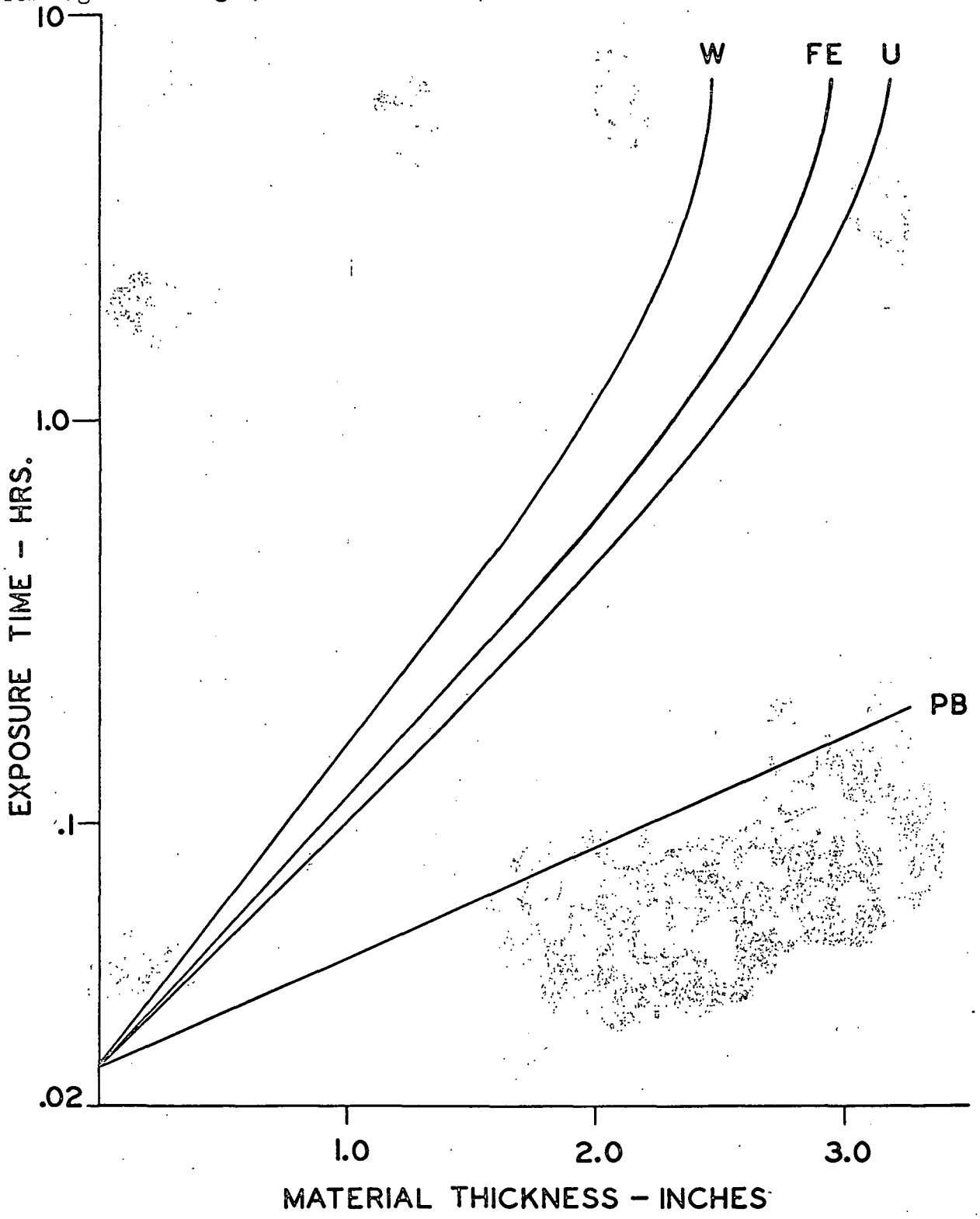

FIGURE 2

These exposure curves for the same materials shown in Fig. 1 are for a direct exposure neutron radiographic technique using a 0.0005 in. thick gadolinium metal screen used as a back screen with Type AA film. An exposure curve for steel using a rhodium-gadolinium screen combination with Type AA film is shown by the dashed curve. The levelling of the tungsten exposure curve and the relatively fast results indicated for steel appear to be caused by prompt (n,γ) radiation emitted from the inspection material.

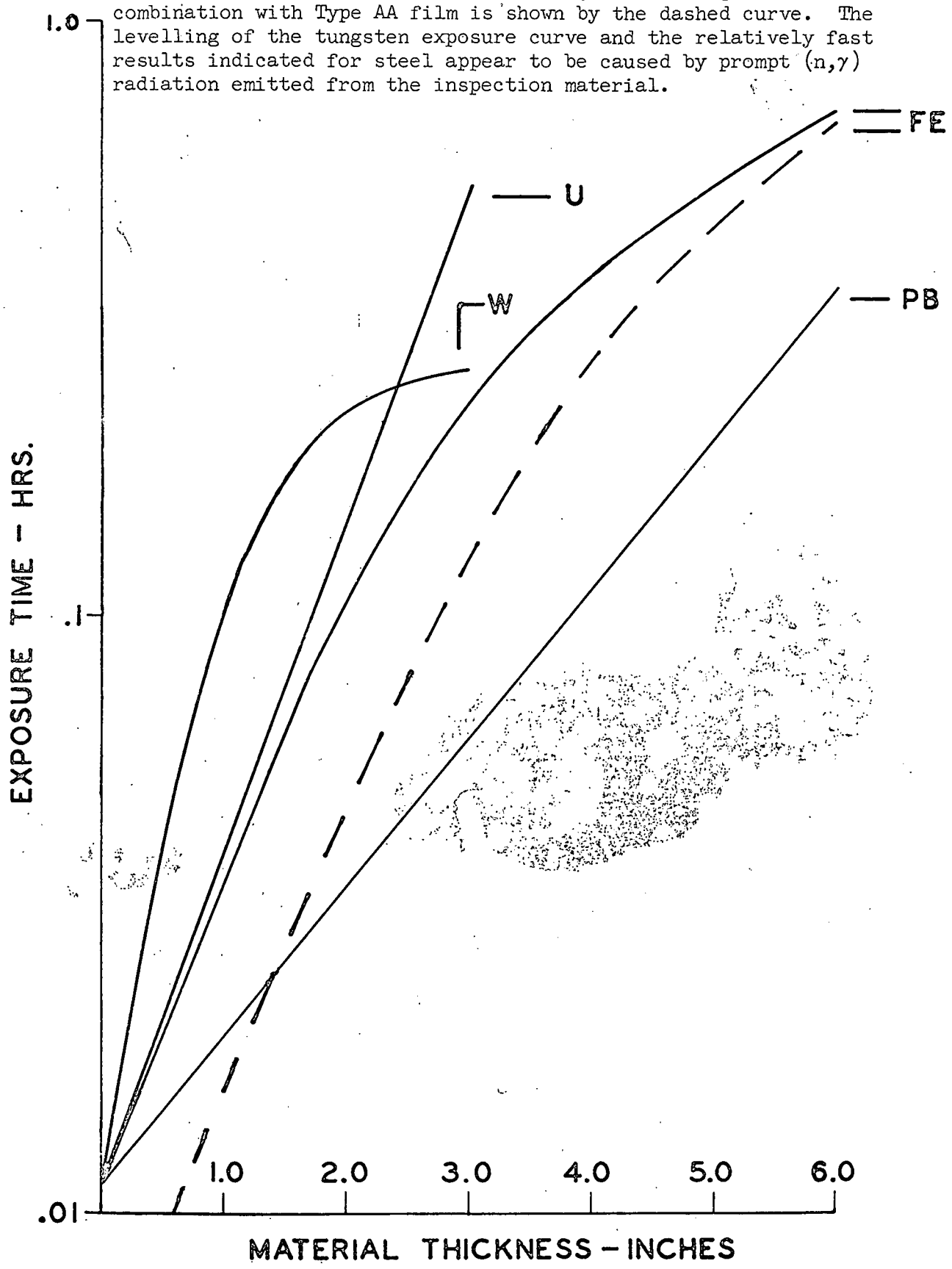

FIGURE 3

These exposure curves, again for the same materials shown for Fig. 1 were taken using a boron-10 loaded scintillator and Type F film in a direct-exposure neutron radiographic technique.

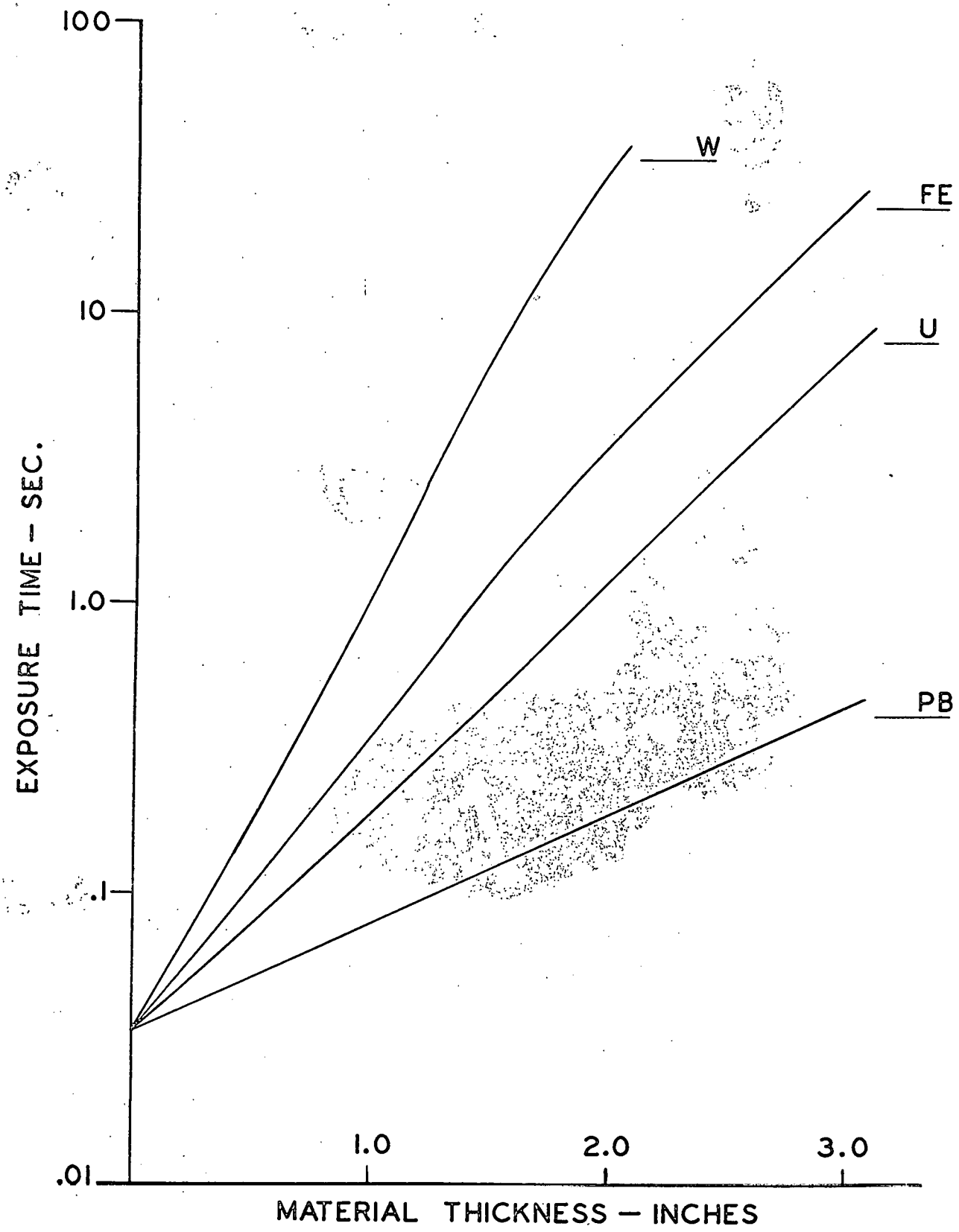

FIGURE 4

The two straight line exposure curves for natural uranium were taken to yield total AA film densities of 1.5 and 2.0, as indicated on the graph. In addition, contrast sensitivity loops yielding 2% and 3% are shown. Exposures within the loops should yield a contrast sensitivity at least that indicated on the loop. All these curves were taken by a direct exposure neutron radiographic method employing a 0.0005 in. thick gadolinium back screen technique. Best contrast sensitivity was obtained if films were agitated during development.

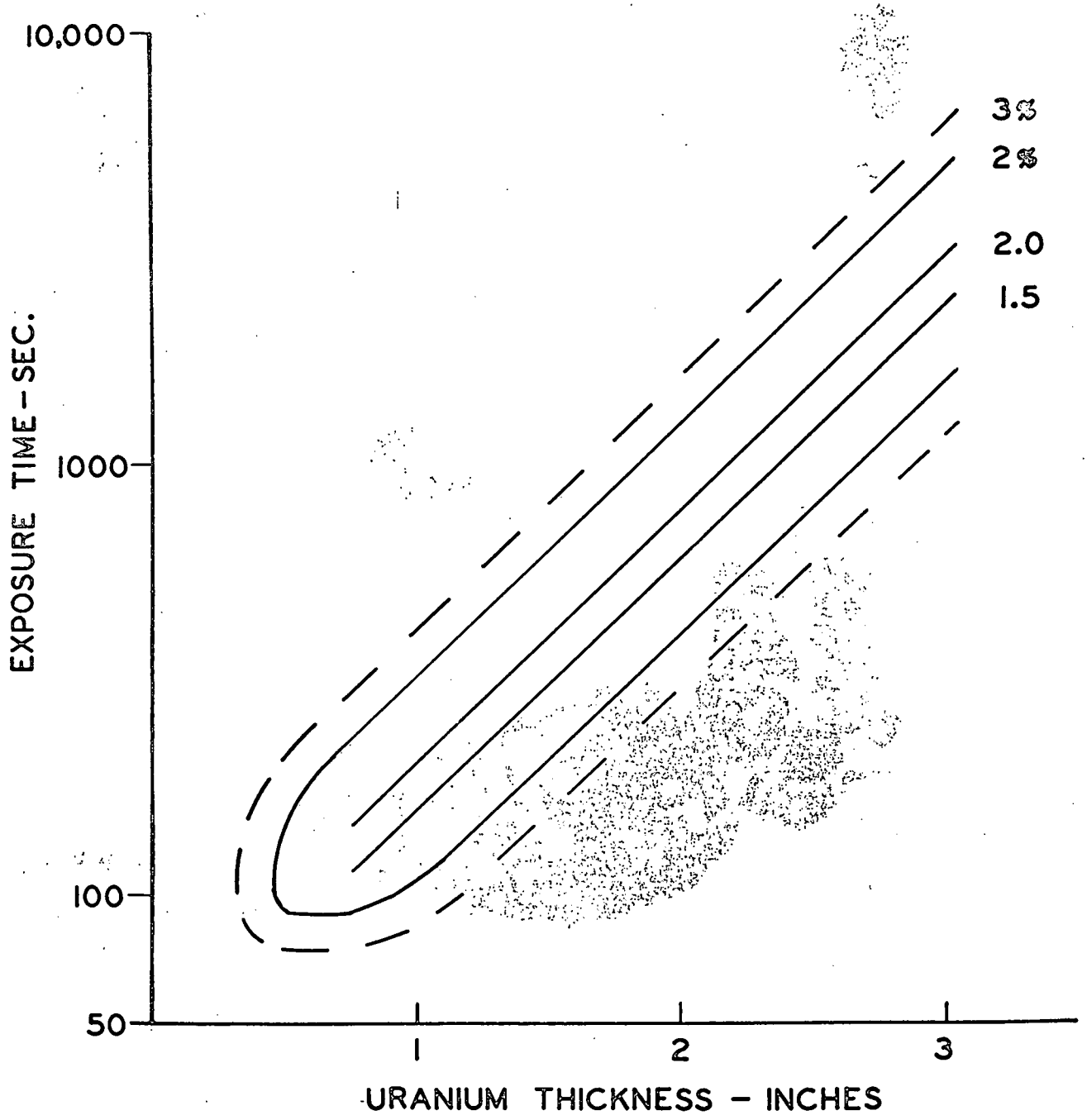

FIGURE 5

Neutron exposure curves for tempered, laminated Masonite for two exposure techniques are shown. The lower curve was taken using a 0.0005 in. gadolinium back screen direct exposure technique with Type AA film. The upper curve was taken by a transfer method using a 0.010 in. indium metal screen transferred to Type AA film for 3 half-lives or more. Neutron exposure curves for a number of other hydrogenous materials such as Bakelite yield similar results.

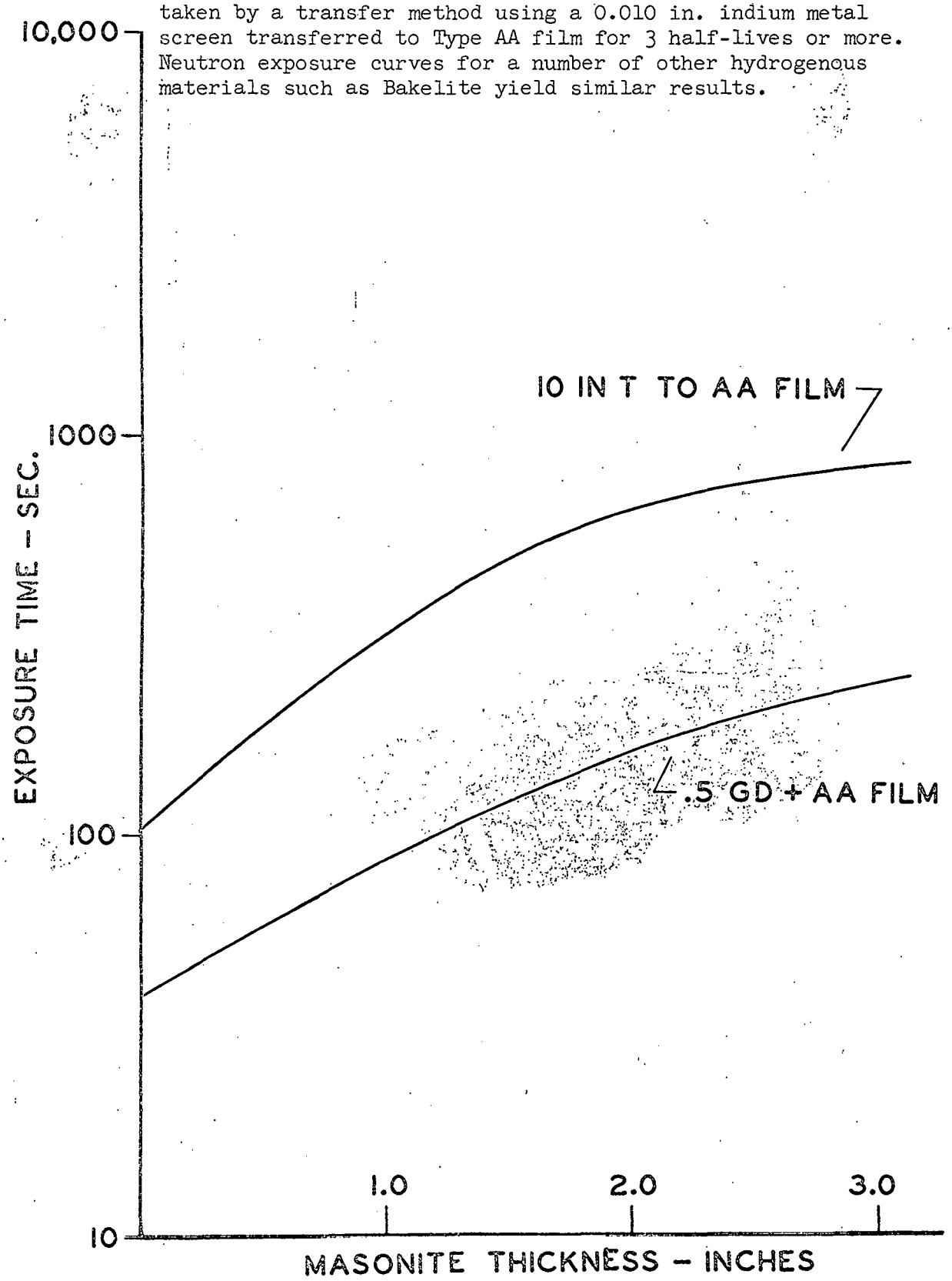

FIGURE 6

These neutron exposure curves for tempered, laminated Masonite were taken using a monochromatic, gamma-free thermal neutron beam having an intensity of 3×10^5 neutrons/cm^2-sec. The lower curve was taken by a direct exposure method using double gadolinium screens, 0.0005 in. front screen and 0.002 in. back screen, with Type AA film. The upper curve was taken by transferring 0.010 in. indium metal screens to Type AA film for 3 half-lives or more.