LA-DC-6978 (Full Paper)

CONF 651021-1

OCT 27 1965

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NEUTRON RADIOGRAPHY UTILIZING GADOLINIUM ACTIVATION FOILS*

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Presented at
the 25th National Convention
of the
Society for Nondestructive Testing
Detroit, Michigan
18 - 22 October 1965

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ABSTRACT

The response to thermal neutrons of gadolinium foils, in combination with radiographic film, was investigated as a part of the research and development program in neutron radiography at the Los Alamos Scientific Laboratory. The experiments were performed by the Nondestructive Testing Group, which is under the direction of Gerold H. Tenney. Class-I, Class-II and Class-III film speeds are presented as functions of film-foil geometries, and of gadolinium activation-foil thicknesses in the 0.0001- to 0.00623-in. range. Internal-conversion electron and photoelectron emission from gadolinium, applied to the neutron radiographic process, is discussed, as is progress in resolution studies. Optimum film speeds are obtained by utilizing a 0.00027-in.-thick gadolinium foil in a back-film geometry and a 0.0045-in.-thick foil in a front-film geometry. Radiographs of optimum resolution are obtained from the back-film technique.

INTRODUCTION

The investigations described in this paper were conducted as a part of the neutron radiography research and development program of the Nondestructive Testing Group of the Los Alamos Scientific Laboratory. The experiments were designed to yield information on: (a) film-foil response characteristics to thermal neutrons of gadolinium foils ranging in thickness from 0.0001 to 0.00623 in., and (b) resolution data utilizing gadolinium activation foils. This paper describes the experimental results obtained when gadolinium direct-activation foils were used in combination with Kodak Type-M (Class-I), Kodak Type-AA, (Class-II), and Kodak Type-K (Class-III) radiographic films.

In the X-ray energy range commonly used in radiography (from above the K-absorption edge to approximately 300 kvp), the total mass absorption coefficient of the elements generally increases with increasing atomic number.⁽¹⁾ Total X-ray absorption is also a function of the density of the absorbing material. Neutron absorption coefficients exhibit a general trend, not well defined however, in which the coefficients increase in magnitude with decreasing atomic number.⁽¹⁾ In addition, the neutron mass absorption coefficients bear no definite relationship to the object material density. In many cases, the neutron mass absorption coefficients of elements differing in atomic number by one, or of isotopes of the same element, are sufficiently dissimilar to allow differentiation between such elements or isotopes by neutron radiographic or absorption techniques.

The neutron radiographic techniques used in these experiments are similar to those of gamma- and X-radiography in general, with three principal exceptions: Firstly, thermal neutrons, rather than photons, are the penetrating radiation. Secondly, the penumbra from the finite neutron source size and from the neutron scatter due to the reactor graphite thermal column

boundaries cause an image unsharpness. Therefore, it is necessary to place the object being inspected as nearly in contact with the film-foil combination as possible, in order to obtain a well-defined radiograph. Thirdly, the neutron radiation transmitted through the object does not, in general, interact directly with the radiographic film to produce an image. The image must be transferred to the film utilizing some intermediate process that is sensitive to the transmitted neutron flux.

Neutron imaging is accomplished by means of an activation foil in combination with radiographic film placed behind the object, as shown in Fig. 1. The differentially absorbed, primary neutron beam results in a non-uniform foil activation, the degree and pattern of which represent the object response to the incident thermal neutrons. This image is then transferred to the radiographic film by placing the film in contact with, and on either side of, the activation foil. The image is actually recorded by exposing the film to the reaction products emitted by the activation foil.

Activation foil speed is defined, for a particular film-foil geometry, as the net optical density; i. e., film darkening produced on a radiographic film in contact with the foil, for a given incident neutron flux. This definition is valid for neutrons having a particular energy or energy spectrum.

THE (n, γ) REACTIONS IN GADOLINIUM

Gadolinium, one of the rare earth elements, has the physical and nuclear properties listed in Table I, below.

TABLE I⁽²⁾

PHYSICAL AND NUCLEAR PROPERTIES OF GADOLINIUM

Atomic Number	64
Neutron Number (Stable Isotopes)	88, 90, 91, 92, 93, 94, and 96
Atomic Weight	157.26
Mass Density at STP	7.95 gm/cm ³
Total Thermal Absorption Cross Section	4.6 x 10 ⁴ barns
Nuclear Density	3.05 x 10 ²² nuclei/cm ³

Table II shows the percentage abundances of the natural gadolinium isotopes, with their respective isotopic (n, γ) reaction cross sections. The two most important gadolinium isotopes for neutron-imaging purposes are Gd-155 and Gd-157, which have thermal activation cross sections of approximately 70,000 and 160,000 barns, respectively.

TABLE II⁽³⁾ISOTOPIC ABUNDANCES AND ACTIVATION CROSS SECTIONS
OF NATURAL GADOLINIUM

Isotope	Relative Abundance, %	(n, γ) Activation Cross Sections, Barns
Gd-152	0.200	125
Gd-154	2.15	Negligible
Gd-155	14.73	70,000 $\pm$ 20,000
Gd-156	20.47	Negligible
Gd-157	15.68	160,000 $\pm$ 60,000
Gd-158	24.87	3.9 $\pm$ 0.4
Gd-160	21.90	0.8 $\pm$ 0.3

The process utilized in this experiment for the purpose of neutron imaging was that of radiative capture in gadolinium. In this process, a slow neutron is captured by the target nucleus, and the resulting compound nucleus emits excess energy in the form of prompt gamma radiation. The product and the target nuclei are isotopic, and the mass number of the product is always increased by one. If the product nucleus of the capture reaction is a stable isotope of the target element, only prompt gamma emission will result. If the reaction results in an unstable nucleus, the radiation from the reaction will be composed of: (a) the prompt gamma radiation, and (b) the particulate and electromagnetic radiation associated with the unstable isotope.

Two n-capture reactions resulting from thermal neutron bombardment of gadolinium are of interest: $\text{Gd-155}(n,\gamma)\text{Gd-156}$, and $\text{Gd-157}(n,\gamma)\text{Gd-158}$. The target and product nuclei are stable isotopes of the same element in both reactions. The intermediate products are left in an excited state after neutron capture, decaying to the ground state with the emission of prompt photons. It is the interaction of the prompt reaction products, i. e., the prompt gamma rays, with the radiographic film in contact with the activation foil that produces the resultant film darkening and neutron imaging.

EXPERIMENTAL PROCEDURE

The neutron source was the Los Alamos Homogeneous Reactor (SUPO Model, Water Boiler Reactor). The mean source-to-foil distance was 87-5/8 in., through a graphite column having a cross section 4-1/4 in. by 4-1/4 in. The finite graphite source size resulted in a primary neutron beam divergence of approximately 3° at the foil plane. The neutron flux at the center of the foil plane was approximately $8.16 \times 10^6 \text{ n/cm}^2/\text{kW/sec}$,⁽⁴⁾ the beam cadmium ratio at that point being 6.27:1.⁽⁴⁾ The neutron beam contains a 4.9 MeV⁽⁵⁾ gamma

component, the result of the neutron capture reaction in graphite.

Monolayer gadolinium foils were utilized to fabricate an eight-component "stepwedge," the thickness of which varied from 0.00027 to 0.00623 in., in irregular increments. A 0.0001-in.-thick foil was mounted separately because of its fragility. Individual foils were positioned at the same point with respect to the neutron beam, by the perpendicular positioning of a specially designed film holder fabricated of 1/16-in. 1100-H14 aluminum sheets. The film holder was removed from the reaction area and unloaded manually after irradiation. The use of thin aluminum stock for the film holder reduced the amount of aluminum activation, consequently minimizing the radiation exposure to personnel. A back-film radiograph utilizing Class-I film and the eight-component stepwedge is shown in Fig. 2-A. The gadolinium stepwedge was mounted on a hinged flap that facilitated placement of the film on either side of the stepwedge. The data reported in this paper were obtained with films simultaneously in the front-film and the back-film positions.

The reactor was operated at a constant power level during the experiment. The film-foil combinations were placed in the neutron beam by means of an automatically timed, cable-driven transport system. Horizontal positioning of the film-foil combinations in the neutron beam was reproducible to ± 0.0025 in. Mechanical details of the system are illustrated in Figs. 3, 4, and 5.

Neutron exposure was controlled by adjusting the reactor power level and by varying the length of time the film-foil combination was allowed to remain in the neutron beam. The transport system was normally programmed to automatically remove the film-foil combination from the neutron beam after the lapse of a predetermined exposure time. The system control circuit panel is shown in Fig. 5.

After exposure, the films were stored in individual, lead-lined cardboard film holders. This precaution was taken to prevent interference between stacked films, a possible consequence of emulsion-component activation.

Exposed films were hand-developed at 68°F. Special handling brackets were designed to ensure simultaneous film immersion in the development tank. The timing sequence for film development was as follows:

General Electric Supermix Developer	8 minutes
Stop Bath	2 minutes
Kodak Solution B Fixer	8 minutes
Water Wash	30 minutes
Air Dry	

Film optical-density measurements were made with a MacBeth-Ansco color densitometer which had undergone a one-hour warmup period. Net optical densities were obtained by taking the difference between the optical densities of films which had been exposed to the neutron beam: (a) in conjunction with the activation foil, and (b) in the absence of an activation foil.

EXPERIMENTAL RESULTS

The net optical density determinations, as a function of activation foil thickness for Class-I, Class-II and Class-III films, are shown in Figs. 6, 7 and 8, respectively. The curves represent data obtained for front-film and back-film geometries; the actual data points correspond to the following foil thicknesses (in.): 0.00010, 0.00027, 0.00057, 0.00103, 0.00210, 0.00287, 0.00373, 0.00540, and 0.00623.

In all cases, the curves representing the front-film geometry exhibit a peak for foil thicknesses between 0.0035 and 0.0050 in.; in addition, the curves show a sharp rise for foil

thicknesses less than 0.00027 in. No apparent peak shift as a function of neutron exposure in the front-film geometry data was observed for an individual film class. Exact location of the front-film geometry peak was difficult to establish due to data scatter, to the lack of data points at smaller foil-thickness increments, and to the fact that the peak was extremely broad.

The film-foil data representative of Class-I, Class-II and Class-III film response to thermal neutrons for a back-film geometry are illustrated by the lower curves of Figs. 6, 7 and 8, respectively. The back-film data exhibit a sharp peak at a foil thickness of 0.00027 in., followed by a plateau in the foil thickness range from 0.0020 to 0.0040 in. A sharp rise is observed for foil thicknesses less than 0.00027 in.

With regard to the back-film data, the ratio of the peak density to the plateau density remained constant as a function of film class and as a function of neutron exposure over the exposure range covered in the experiment. The peak-to-plateau ratio was found to be indicative of the film contrast, as shown in Table III.

TABLE III

PEAK-DENSITY-TO-PLATEAU-DENSITY RATIO
FOR CLASS-I, CLASS-II AND CLASS-III FILMS
(BACK-FILM GEOMETRY)

Film Class	Film Contrast	Peak-to-Plateau Ratio
Class-I	High	3.2
Class-II	Medium	1.45
Class-III	Low	1.2

The net front-film and back-film optical density, as a function of neutron exposure, for Class-I, Class-II and Class-III films is illustrated in Fig. 9. A 0.00027-in.-thick gadolinium foil was used in obtaining data to determine neutron exposure required to produce a net optical density of 1.5. These data are listed in Table IV for the film classes investigated in this experiment.

TABLE IV

NEUTRON EXPOSURE REQUIRED TO PRODUCE
A NET OPTICAL DENSITY OF 1.5
IN CLASS-I, CLASS-II, AND CLASS-III FILMS
(FRONT-FILM AND BACK-FILM GEOMETRIES)

Film Class	Neutron Exposure (kW-sec)	Neutron Exposure (kW-sec)
	Front-Film Geometry	Back-Film Geometry
Class-I	615	800
Class-II	120	150
Class-III	28	33

The shape of the front-film curve shown in Fig. 8 agrees with the data obtained by Berger ⁽⁶⁾ in the gadolinium foil thickness range from 0.00025 to 0.00200 in. The Los Alamos data indicate that the curve peaks at a foil thickness of 0.004 in.

However, in the case of back-film geometry, there is sharp disagreement between the Los Alamos curve and data of Berger. ⁽⁶⁾ No attempt was made to correlate the characteristic curves shown in Fig. 9 with the data of Berger.

Calculations show that the response curves for front-film and back-film geometries should resemble those illustrated in Fig. 10-A if one assumes a monoenergetic gamma emission from the activation foil under consideration. However, the theoretical

curves disagree with the experimental curves illustrated in Figs. 6, 7 and 8. Preliminary absorption experiments performed earlier by this group, in which mylar absorber films of increasing thickness were placed between the activation foils and the radiographic films, indicated that as absorber thickness increased, the shape of the experimental curves approached that of the theoretical curves. The curves obtained from these experiments are shown schematically in Figs. 10-B and 10-C. The results of these experiments indicate that the sharp rise observed in the front-film and back-film data is, in fact, due to soft particulate emission from the gadolinium foil resulting from neutron capture.

One source of particulate emission from gadolinium has been shown to be internal-conversion electrons,⁽⁷⁾ when the energy of the excited intermediate nucleus is transferred to an orbital electron, resulting in electron ejection from the K or L shell. It has been suggested that a second source of electrons could result from photoelectric interactions of the prompt gamma radiation with the gadolinium foil matrix. The fact that the photoelectric cross section is high for low-energy photons interacting with a high-Z material, such as gadolinium, lends support to this argument.

RESOLUTION STUDIES

At the time the gadolinium foil experiment was performed, no adequate test pieces were available for a study of resolution as a function of foil thickness. Instead of a test piece, a 0.00025-in.-thick gadolinium foil was placed in contact with the film holder in such a manner as to mask half the stepwedge width. The results of this experiment are shown in Fig. 2-B, in which the left-hand and right-hand radiographs correspond to the back-film and front-film geometries, respectively.

An evaluation of the resolution experiment, conducted by Los Alamos film interpreters, indicated that the back-film geometry utilizing a 0.0005-in.-thick gadolinium foil produced the radiograph having the highest resolution.

ANALYSIS OF EXPERIMENTAL DATA

Analysis of the experimental data demonstrated that film speed, and/or resolution of neutron radiographs, can be optimized by the appropriate selection of: (a) film-foil geometry, (b) activation foil thickness, and (c) film class.

With regard to film-foil geometry, optimum film speed was obtained utilizing a 0.004-in.-thick gadolinium foil in conjunction with radiographic film in a front-film geometry. Optimum film speed was obtained with a back-film geometry utilizing a 0.00027-in.-thick activation foil. In the absence of a suitable test plate and of a rigorous examination of neutron-radiographic resolution as a function of activation foil thickness, it was the conclusion of Los Alamos film interpreters that radiographs exhibiting the best resolution resulted when a back-film geometry was employed.

Foil thicknesses, corresponding to the peaks of the film-foil response curves of Figs. 6, 7, and 8, appeared to be independent of film class and, in addition, were not a function of neutron exposure.

For a particular film class, the peak-to-plateau ratio of the back-film curve data remained approximately constant with varying neutron exposures. This ratio was a function of the film class used for the exposure and was indicative of the film contrast.

The mylar absorption experiments clearly demonstrated that the high resolution of neutron radiographs made with gadolinium foils was due to the soft particulate radiation from the gadolinium as a result of neutron capture.

ACKNOWLEDGMENTS

The authors express their appreciation to H. T. Motz and M. M. Thorp of the Los Alamos Reactor Group for their assistance and cooperation throughout the course of the experiment; to R. W. Keil of the Fabrication Section, the Materials Technology Group; to E. J. Vonderheide and R. T. Schmitt of the Nondestructive Testing Group Branch Shop; to A. J. Demny of the Nondestructive Testing Group Branch Engineering Department; and to W. B. Martin, of the Radiation Section, the Nondestructive Testing Group. The authors are also indebted to G. H. Tenney, Group Leader, Nondestructive Testing Group, for his suggestions regarding the preparation of this paper and to H. H. Bybee, Jr.,* a Research Assistant in the Los Alamos Scientific Laboratory Summer Graduate Program, for his significant contribution to the successful completion of this phase of the experiment.

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- Figure 2-A - Gadolinium Stepwedge Radiograph.
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- Figure 3 - Film-Foil Transport System, Los Alamos Homogeneous Reactor.
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FIGURE 1
NEUTRON IMAGING SYSTEM

FIGURE 2A

FIGURE 2B

CMX-1 GT SITE
ORDER NO. 10630

FIGURE 3

GMX-1 GT SITE
ORDER NO. 10628

FIGURE 4

GMX-1 GT SITE
ORDER NO. 10629

FIGURE 5

GMX-1 GT SITE
ORDER NO. 10624

FIGURE 6
NET OPTICAL DENSITY AS A FUNCTION OF FOIL THICKNESS FOR
CLASS I FILM
(NEUTRON EXPOSURE = 825 KW-SEC)

FIGURE 7

NET OPTICAL DENSITY AS A FUNCTION OF FOIL THICKNESS FOR
CLASS II FILM
(NEUTRON EXPOSURE = 200 KW-SEC)

FIGURE 8

NET OPTICAL DENSITY AS A FUNCTION OF FOIL THICKNESS FOR
CLASS III FILM
(NEUTRON EXPOSURE = 30 KW-SEC)

FIGURE 9

NET OPTICAL DENSITY AS A FUNCTION OF NEUTRON EXPOSURE
(FOIL THICKNESS = 0.00027 INCH)

FIGURE 10-A
THEORETICAL FILM-FOIL RESPONSE CURVES

FIGURE 10-B
EXPERIMENTAL FILM-FOIL RESPONSE CURVES AS A FUNCTION OF
MYLAR ABSORBER THICKNESS (FRONT FILM GEOMETRY)

FIGURE 10-C
EXPERIMENTAL FILM-FOIL RESPONSE CURVES AS A FUNCTION OF
MYLAR ABSORBER THICKNESS (BACK FILM GEOMETRY)