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CALIFORNIUM-252 AS A SOURCE FOR THERMAL NEUTRON RADIOGRAPHY

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ABSTRACT

Thermal neutron radiography is compared to other methods for nondestructive evaluation; it is shown to offer advantages in many aerospace and nuclear inspection areas. One example concerns problems such as detection of nonbonds or cracks open to the surface, where liquid contrast agents can be used to help image defect areas. Descriptions and economics of neutron sources including reactors, subcritical assemblies, accelerators and radioactive sources are given. All the sources can be considered for in-plant inspection use; the higher yield sources offer initial economic advantages. Radioactive sources are well suited for field applications. The inspection effectiveness of ^{252}Cf has been demonstrated. This source seems particularly useful where a peak thermal flux of $\geq 10^7$ n/cm$^2 \cdot$s is needed.

1. INTRODUCTION

Radiography with neutrons is now being used as a method for non-destructive evaluation[1] in the explosive, nuclear, space and aircraft industries. This inspection work is performed with neutrons because other nondestructive evaluation methods, including x-radiography, do not supply all the desired information about the object.

Neutrons offer advantages in many of these cases because most metals are relatively transparent while several light materials, such as hydrogen, lithium and boron have high attenuation. Sensitivity to hydrogen opens up many application areas involving common materials such as adhesives, fluids, plastics and rubber. The relative attenuation properties of the elements for thermal neutrons and medium energy x-rays are given in Fig. 1.

In addition to the relative reversal of attenuation for the light and heavy materials for thermal neutrons and x-rays, Fig. 1 shows widely varying neutron attenuation for several groups of neighboring elements. Neutrons provide a simple radiograpic method for visualizing cadmium or many rare earth materials, for example, in assemblies containing several neighboring elements. Other advantages for neutron inspection include relative insensitivity to gamma rays (so that highly radioactive material may be inspected), differentiation capability for certain isotopes, and the advantage that new types of contrast agents may be used.

These capabilities of thermal neutron radiography are compared to those of x-radiography (see Fig. 1). The neutron technique also offers advantages as compared to other methods for nondestructive evaluation[2]. A comparison of the capabilities and limitations of the common methods for industrial nondestructive evaluation (NDE) is given in Table I. Compared to the other NDE methods, neutron radiography provides a direct means to determine such things as proper part placement in assemblies (including rubber, plastic or explosive components, for example), location and cure of adhesives and resins, detection of corrosion, discrimination capability for certain isotopes and the capability for inspecting radioactive material. In addition, by the use of contrast agents such as penetrants or gadolinium doped liquids, the neutron detection of nonbonded areas and cracks can be very much improved[3-5]. These types of flaws normally are difficult to detect by conventional radiography; with neutrons they can be visualized.

2. SOURCES FOR NEUTRON RADIOGRAPHY

There are many sources that have been used for neutron radiography and gaging. These include reactor, accelerator, radioactive and subcritical assembly sources. A summary of the general characteristics of these various types of sources is given in Table II. The limits of available intensity, as well as the other observations, are not firm but they give an indication of what may normally be expected from each type of source.

Most thermal neutron radiographic work has been done with reactor sources; confirmation of this may be obtained from several recent reviews[1,6-11]. However, the use of nonreactor sources is increasing. Accelerators used for thermal neutron radiography include low voltage, Cockroft-Walton generators employing the (d-T) reaction, linear accelerators employing the (X,n) reaction, and Van de Graaff* or Dynamitron* accelerators in which a beryllium target is bombarded with deuterons or protons. Radioactive sources used for thermal neutron radiography include ^{124}Sb-Be, ^{241}Am-Be, ^{241}Am-^{242}Cm-Be and ^{252}Cf. Other radioactive neutron sources have been considered for neutron radiography; characteristics of many radioactive sources are given in Table III.

The subcritical assembly or neutron multiplier is a more recent source for this type of inspection. It is described at this meeting[14] and has been discussed in the literature[15-17].

The recently published proceedings of the 1975 meeting on applications of neutron radiography and gaging[1] may provide an indication concerning the sources actually being used. There are 14 specific application papers in the proceedings. The sources used for this application

*The naming of specific products or apparatus throughout this report is for identification only and does not constitute an endorsement of any particular product or equipment.

work were distributed as follows (one paper described work with two sources): reactor, 6; ^{252}Cf, 5; Van de Graaff accelerator, 3; and ^{241}Am-Be, 1. It can be seen that non-reactor sources predominated in this instance and that ^{252}Cf was a popular choice. At a more recent technical meeting (October, 1975) four of the seven papers presented in a session on neutron radiography[18] included data on ^{252}Cf.

All these sources yield primarily fast neutrons. For thermal neutron radiography and gaging, and the good material (or isotope) discrimination sensitivity that goes with thermal neutrons as opposed to fast neutrons, it is necessary to slow the neutron velocities. This is normally done by a surrounding light material moderator such as water, oil, paraffin, or plastic. Since the efficiency of moderation depends on source factors such as physical size and neutron energy spectrum, the method of comparing source cost by comparing the total fast neutron yield (as was done in Table III) is not completely descriptive. An additional factor that must be taken into account is the thermalization factor, namely the ratio of the total fast neutron yield from the source in neutrons/s, to the peak thermal neutron flux in the moderator (in units of $n/cm^2 \cdot s$). These differ widely for the various sources; a low thermalization factor is favorable because more of the source neutrons are available for extraction from the moderator as a thermal neutron beam.

A source such as ^{124}Sb-Be has a very low thermalization factor, 45, because the neutrons emitted are reasonably low energy (25 keV) initially; the mean energies of emitted neutrons from most other sources are well in the MeV range[19]. The 14MeV neutrons emitted from (d-T) reaction accelerators are difficult to moderate; the thermalization factor[11,19] is in the range 600 to 1000. Hawkesworth has provided a summary of thermalization factors for several types of sources. His results are shown in Table IV. Combining the neutron yield and the thermalization factor, one can make a more realistic comparison of source costs for thermal neutron radiography and gaging[19,10]; results are shown in Fig. 2. These data were tabulated in 1971; however, the relative costs appear to be valid now.

Although one can draw a line through the average of all the points in Fig. 2, it will be seen that the average cost per neutron (peak thermal neutron flux) tends to decrease as higher intensity sources are considered. For example, the two reactor data points at the upper right give about $10^6 n/cm^2 \cdot s$ flux per dollar of basic equipment cost; the cluster of sources at lower center produce about $100 n/cm^2 \cdot s$ per equipment dollar, some four orders of magnitude poorer.

When ^{252}Cf is compared to other neutron sources economically, both favorable and unfavorable comparisons can be found. The 1 mg ^{252}Cf source indicated in Fig. 2 gives about $10^3 n/cm^2 \cdot s$ per dollar of encapsulated source cost. Below that data point the cluster mentioned above yields an order of magnitude fewer neutrons per dollar. On the other hand, directly above the 1 mg ^{252}Cf source are a number of accelerator sources with approximately the same basic source cost of $25 to $30,000. These give as many as $2.3 \times 10^3 n/cm^2 \cdot s$ per dollar, a figure that does not include[17] the use of a uranium multiplier block around the target. A thickness of 2cm of uranium around the target has been shown to increase the neutron flux by a factor of 3 or more[22,23], by adding neutrons by (n,2n), (n,3n) and fission reactions. This would raise the accelerator to a point where it would provide more than $6 \times 10^3 n/cm^2 \cdot s$ per dollar of equipment cost, a factor of about 6 more than a similarly priced ^{252}Cf source.

3. DISCUSSION AND CONCLUSIONS

There are many factors to be considered in a neutron source to be used for thermal neutron radiography or gaging. Some indication of basic source acquisition costs has been given; that is one important factor. Another factor concerns operating cost. A radioactive source gives a steady output but it slowly decays. Replacement cost must be considered.

Accelerator sources have operating costs associated with them in terms of target or tube replacement and maintenance costs for electronic and vacuum systems. Although the output of an accelerator is normally less stable than that of a radioactive source, it offers the advantage that it can be turned off, thereby minimizing some safety problems. The fact that it can be turned off is also important for relating the operating cost to the use factor. If a source will be used much of the time, an accelerator may be expensive because of tube or target replacement costs. If, however, a source is planned for less extensive use, the constant decay of a radioactive source can be a high expense item.

Reactors provide relatively high intensity, stable sources, but also involve several operators, licenses and safeguard provisions, thereby adding to the cost of operation. An advantage is that a reactor can be stopped, as can the accelerator.

Subcritical assemblies or neutrons multipliers, like reactors, are sufficiently complex that the sources start at a relatively high cost. Like the reactor, these sources can be very stable and repeatable in output. They require less in the way of operators than a reactor, but do also require safeguard provisions.

Certainly all these sources, and perhaps new source concepts such as plasmas[24], have a place in neutron radiography and gaging. The growth of neutron inspection methods in industry will depend on the availability of sources (and associated equipment) that can be used in a manufacturer's plant and in the field, as x-radiography is used today. All the sources have the potential for in-plant use. All these sources also have the potential for field use. Both reactors and accelerators have been mounted on trucks for mobile operation.

In practice, however, the radioactive sources offer the greatest potential for field neutron radiography and gaging. For small yield sources giving $10^6 n/cm^2 \cdot s$ or less peak thermal flux in a moderator, several sources seem feasible and economical, including ^{241}Am-Be, ^{241}Am-^{242}Cm-Be, ^{252}Cf ^{242}Cm-Be, ^{244}Cm-Be, ^{210}Po-Be, and ^{239}Pu-Be. For these lower yield sources that are attractive for field use, the possibility of a truly portable source employing the (α,η) reaction should also be mentioned. For efficiency of neutron production it is necessary to mix well the alpha source and the neutron target (since the alpha range is so short). However, if the alpha source or the target were a fluid (gas or liquid), good mixing could be obtained and yet the two could be separable, thereby improving portability (since one could easily shield the alpha source). The neutron target, ^{18}O, offers such a possibility since it could be used as a fluid and drawn off to leave an easily shielded alpha source. Experiments with these sources[25] have shown neutron yields as high as one-third those of the best α-Be neutron source mixtures. Giving up 2/3 of the possible neutron yield, even if that performance could not be improved, might not be too much to pay for a truly portable neutron source.

On the other extreme, there are occassions when a field radiographic application demands short inspection times. It has been shown that ^{252}Cf sources well over a milligram in size can be used for such applications[4]. For field sources that yield more than $10^7 n/cm^2 \cdot s$ peak thermal flux, ^{252}Cf offers a distinct advantage because of its low heat generation[12], a fact that permits the use of large sources without problems of source cooling.

Additional advantages offered by ^{252}Cf as a source for neutron inspection are the reasonable half-life (2.6y), steady output, and relatively low gamma radiation output. Certainly a present advantage is the significant amount of research and development that has been accomplished with ^{252}Cf. Thus, ^{252}Cf as a source for both laboratory and field use for neutron radiography and gaging has been effectively demonstrated[26]. Neutron sources seem destined for increasing industrial inspection use; ^{252}Cf offers advantages, particularly in the higher intensity area. Future predictions of more than $9 million worth of ^{252}Cf neutron radiography equipment (all involving mg level sources) in the next five years[27] could prove to be unduly pessimistic.

This report has been concerned only with thermal neutron radiography because that is the energy range where the major activity has been concentrated. However, interest in radiography with neutrons in the cold, resonance and fast energy ranges is on the increase[6-11]. Cooled moderators provide a way to obtain cold neutrons from a source such as ^{252}Cf. Energy tailoring in the moderator-collimator assembly can increase the yield of resonance neutrons. The bare source itself provides fast neutrons. Therefore, as interest in nonthermal neutron radiography increases, one can expect ^{252}Cf to continue to play an important role.

Table 1. Comparison of Common Nondestructive Evaluation Methods

Method	Characteristics Detected	Advantages	Limitations
Visual-Optical	Surface characteristics such as finish, scratches, cracks or color; strain in transparent materials.	Often convenient; can be automated.	Can be applied only to surfaces, through surface openings or to transparent material.
Liquid Penetrant	Surface openings due to cracks, porosity, seams or folds.	Inexpensive, easy to use, readily portable, sensitive to small surface flaws.	Flaw must be open to surface. Not useful on porous materials.
Ultrasonics	Changes in acoustic impedance caused by cracks, nonbonds, inclusions, or interfaces.	Can penetrate thick materials; excellent for crack detection; can be automated.	Requires coupling to material either by contact to surface or immersion in a fluid such as water.
Radiography	Changes in density from voids, inclusions, material variations; placement of internal parts.	Can be used to inspect wide range of materials and thicknesses; versatile; film provides record of inspection.	Radiation safety requires precautions; expensive; detection of cracks can be difficult.
Magnetic Particles	Leakage magnetic flux caused by surface or near-surface cracks, voids, inclusions, material or geometry changes.	Inexpensive, sensitive both to surface and near-surface flaws.	Limited to ferromagnetic material; surface preparation and post-inspection demagnetization may be required.
Eddy Currents	Changes in electrical conductivity caused by material variations, cracks, voids, or inclusions.	Readily automated; moderate cost.	Limited to electrically conducting materials; limited penetration depth.

Table II. Characteristics of Thermal-Neutron Sources

Type of Source	Typical radiographic intensity (a)	Resolution	Exposure time	Characteristics
Radioisotope	10^1 to 10^4	Poor to medium	Long	Stable operation, medium investment cost, possibly portable.
Accelerator	10^3 to 10^6	Medium	Average	On-off operation, medium cost, possibly portable.
Subcritical Assembly	10^4 to 10^6	Good	Average	Stable operation. Medium to high investment cost, portability difficult.
Nuclear reactor	10^5 to 10^8	Excellent	Short	Stable operation. Medium to high investment cost, portability difficult.

(a) Neutrons per sq cm per s.

Table III. Some radioactive sources for neutron radiography and gaging

Source	Reaction	Half-Life	Cost[1] in thousands of dollars	Average neutron energy (meV)	Neutron yield (n/s-g)	Gamma dose (rads/hr at 1 m)[2]	Gamma ray energy (MeV)	Comments
^{124}Sb-Be	(γ,n)	60 days	25	0.024	2.7×10^{9}	4.5×10^{4}	1.7	Short half-life and high γ background, available in high intensity sources, low neutron energy is an advantage for thermalization
^{210}Po-Be	(α,n)	138 days	20	4.3	1.28×10^{10}	2	0.8	Short half-life; low γ background
^{238}Pu-Be	(α,n)	89 years	310	∿4	4.7×10^{7}	0.4	0.1	High cost, long half-life
^{241}Am-Be	(α,n)	458 years	1500	∿4	1×10^{7}	2.5	0.06	Easily shielded γ output, long half-life, high cost
^{241}Am-^{242}-Cm-Be	(α,n)	163 days	-	∿4	1.2×10^{9} (80% Am, 20% Cm)	Low	0.04 0.06	Increased yield over ^{241}Am-Be for relatively little more cost but with a short half-life
^{242}Cm-Be	(α,n)	163 days	-	∿4	1.46×10^{10}	0.3	0.04	High yield source, but half-life is short
^{244}Cm-Be	(α,n)	18.1 years	35[3]	∿4	2.4×10^{8}	0.2	0.04	Long half-life, low γ background are attractive. Source can also be used as a spontaneous fission source, with about half the neutron yield. Because ^{244}Cm is produced in nuclear fuel, this radiosotope could be widely available as a by-product material.
^{252}Cf	Spontaneous fission	2.65 years	200[4]	2.3	3×10^{12}	2.9	0.04 0.1	Very high yield source, present cost projected future cost makes it attractive, small size and low energy are advantages for moderation

[1]Cost of the radionuclide only is given; see Reinig [12]. The cost is normalized to a source total yield of 5×10^{10} n/s.
[2]The γ-ray dose is normalized to a neutron yield of 5×10^{10} n/s.
[3]The cost is based on a proposed cost of \$170/g, as quoted by Stewart, Horwitz & Youngquist [13].
[4]The cost is based on the present proce of \$10/μg, unencapsulated.

Table IV. Thermalization of Fast Neutrons

Type of Source	Thermalization Factor[a]
d-T accelerator reaction	600
d-D acceleration reaction	200
^{241}Am-Be radioactive source	200
d-Be accelerator reaction	100[b]
^{252}Cf radioactive source	100[b]
^{124}Sb-Be radioactive source	45

[a] Approximate values based on work at the University of Birmingham, see reference [20].

[b] Approximate values from the literature.

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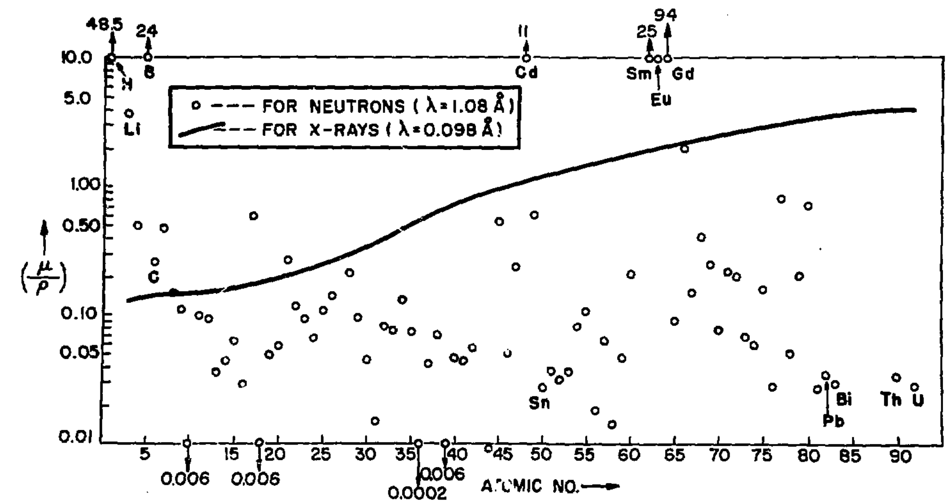

Figure 1. Absorption coefficients of the elements for both x-rays (solid line) and thermal neutrons (dots) are shown on a logarithmic scale.

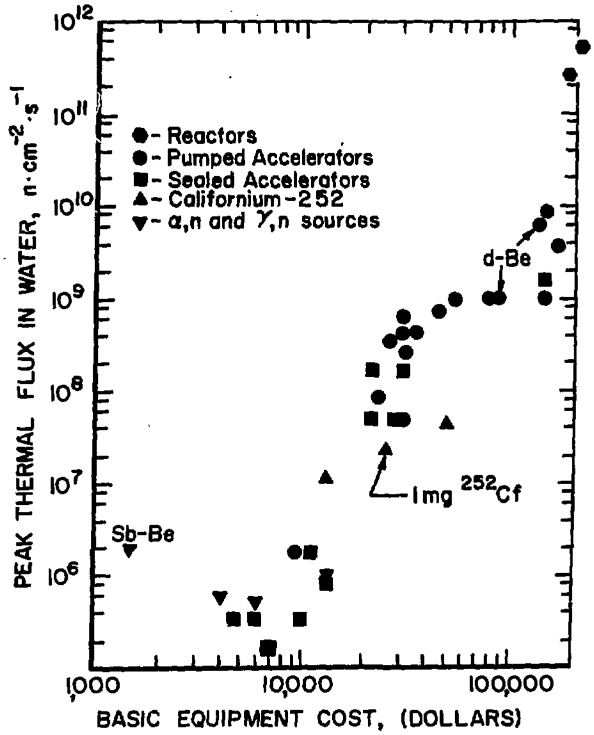

Figure 2. Peak thermal neutron flux in a surrounding water moderator is plotted against cost of the basic source for a variety of sources. Courtesy - M. R. Hawkesworth, University of Birmingham, ref. [20].