REPORT A66-14

...ACKSCATTERING AND RADIATION-INDUCED X-RAYS FOR ...RFACE COMPOSITION AND STRUCTURE

by

SIGMUND BERK

June 1966

AMCMS Code 5025.11.294

Distribution of this document is unlimited.

U.S. ARMY MUNITIONS COMMAND

UNITED STATES ARMY
FRANKFORD ARSENAL
PHILADELPHIA, PA.

Abstract

The use of promethium-147 beta backscattering to measure the thickness of plastic coatings on copper was determined. Backscattering radiation was also used to determine the structure and surface composition of copper specimens. A preliminary study of the use of beta-particle backscattering and beta-particle-induced X rays for measuring corrosion is described.

Radiation Backscattering And Radiation-Induced X Rays for Measuring Surface Composition and Structure

By SIGMUND BERK

Pitman-Dunn Research Laboratories, Frankford Arsenal, Philadelphia, Pennsylvania

Introduction

The literature on beta and gamma backscattering and radiation-induced soft X rays has become extensive. In October, 1964, the U.S. Atomic Energy Commission and the Illinois Institute of Technology sponsored a symposium on low-energy X rays and gamma sources and their application.[1] A number of papers on radiation backscattering were presented at the second Geneva Conference.[2] R. H. and D. C. Müller contributed some excellent work on the relative backscattering of the elements and of a number of compounds.[3, 4, 5] Their work resulted in a systematic study of radiation backscattering and produced a number of analytical applications of the method by others.[6]

In this article a number of applications of a promethium-147 beta-particle backscattering gauge are described. The effect of thickness of plastic films on metal was measured by backscattering radiation. Also, the contamination of copper surfaces and their surface structure were determined by beta-particle backscattering. Results of preliminary experiments on the use of backscattering by beta particles and beta-particle-induced Kα X rays for measuring corrosion are reported.

Beta-Particle Backscattering

A brief description of the characteristics of beta particles and of beta-particle backscattering is given below. Beta particles are high-speed electrons which have their origin in the nucleus of certain radioactive elements. Of the large number of radioisotopes (more than 900), there are only 16 pure beta-particle-emitting radioisotopes commonly available, and of these only a small number are suitable for sources for backscattering studies. The most commonly used are listed in Table I. Aebersold and Fowler[7] limit the number of (beta- and gamma-emitting) radioisotopes suitable for transmission and reflection gauges to six (Sr-90, Tl-204, Kr-85, Co-60, Cs-137, Ir-192). Of these only the first two are pure beta-particle emitters.

TABLE I

Pure beta-particle-emitting radioisotopes useful in backscattering

Radioisotope	Half-Life	Maximum Energy (Mev)
Sr-90	25y*	0.61
Y-90	2.5d*	2.18
Tl-204	4y	0.76
Pm-147	2.6y	0.223
S-35	87d	0.167
C-14	5568y	0.155
P-32	14.3d	1.7
H-3	12.5y	.018

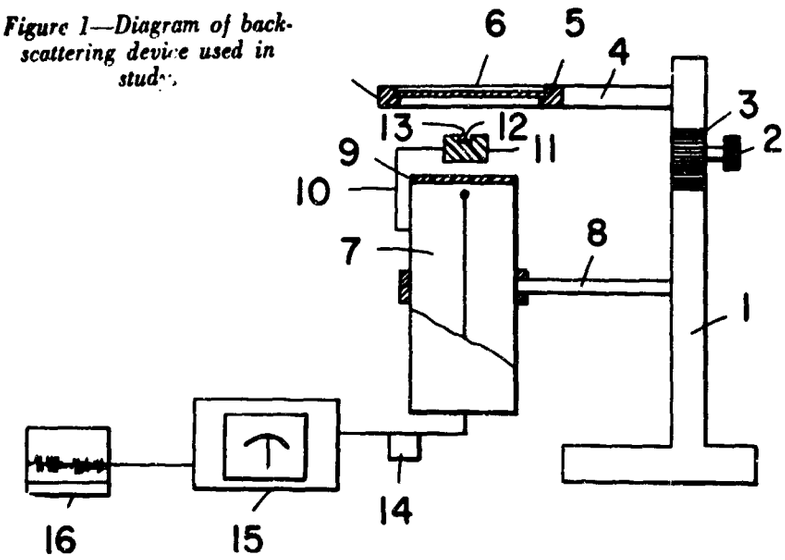
* d = days
y = years

The penetration of beta particles through matter is a complex phenomenon due to the many ways that interactions occur.[8, 9] The actual path of the beta particle in a substance cannot be predicted. When beta particles strike a surface, they are absorbed or scattered by the atoms in the material. Some of the beta particles return in the general direction they came from (this is backscattering); however, the intensity of the scattered beta radiation will be largest in the forward direction from the source.

The backscattering is not a reflection in the optical sense, since the reflection does not occur in the surface layer but is a volume phenomenon. Backscattering increases with increasing thickness of the target until saturation occurs. Saturation occurs when the reflecting layer is about one-third of the beta-particle range in the material.[10] Further increase in thickness does not add to the reflected intensity. In backscattering, the beta particles must not only penetrate the entire thickness of material at saturation thickness, but also return through the same thickness to reach and actuate the radiation detector.

There is no difference in backscat-

Figure 1—Diagram of backscattering device used in study.



1. Metal stand. 2. Knurled knob for adjusting sample height. 3. Gears. 4. Arm. 5. Plastic sample holder (see Figure 2). 6. Specimen to be measured. 7. Radiation counter. 8. Support arm. 9. Window of counter. 10. Plate bracket for radioactive source. 11. Plastic source container and shield. 12. Radiation source. 13. Plastic film shield. 14. Preamplifier. 15. Scaler. 16. Recorder.

tering from a substance in liquid or solid state despite density differences. When the radioactive source consists of materials of high Z number, the effect of scattering in the source itself must be considered.

The range of beta particles in air is several meters, depending on the energy of the particle. However, range is usually expressed in equivalent thickness or mg per cm^2 of an absorber. For low Z elements or materials (aluminum, mica) the absorption thickness is almost independent of the nature of the absorber. It has been evident for some time that the backscattered intensity is dependent on the nature of the scattering material, i.e., the saturation level increases with increasing Z. In addition Müller[3] showed that the backscattering of beta particles is a discontinuous function of Z but linear in Z within each period of the periodic system.

Müller found that backscattering could be calculated for any compound using an average atomic number ($\overline{Z}$), or % backscattering. For example, $\overline{Z}$ for iron is 23.387 and for iron oxide (Fe_2O_3) it is 16.393. The % backscattering for rusted iron is approximately 30% less than that of iron.

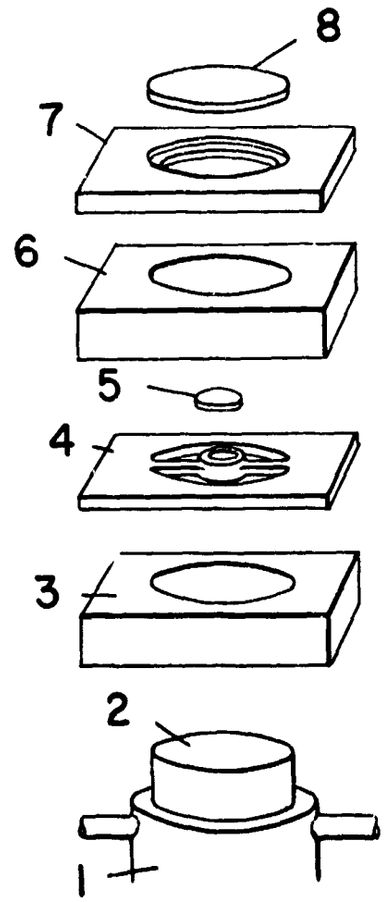
Beta Backscattering Gauge

A 1.9 mg/cm^2 mica end window (Geiger-Müller) counter connected to a preamplifier and a scaler was used. Figure 1 is a diagram of the backscattering instrument used. An exploded view of the methyl methacrylate plastic assembly for the radioactive source, sample holder and a proportional gas flow counter assembly is shown in Figure 2. The size of the samples used ranged from ½ to 1½ in. in diameter. Larger samples can be measured by increasing the diameter of the radioactive source. The optimum distance between the target and detector was found to be 14-15 mm (Table II), and the source-to-target distance ranged from 7.9 to 8.9 mm. Figure 3 shows the effect of varying source-to-target distances on the backscattering of Pm-147 beta particles from a 530 mg/cm^2 aluminum target.

The source holder consisted of a 5/16-in. diameter methyl methacrylate disk with a ⅛-in. diameter and ⅛-in. deep cavity for adding the radioactive material. The radioactivity of the promethium-147 used ranged from 0.3 to 5.0 microcuries. Promethium-147 has a half-life of 2.6 years and a maximum energy of 0.223 Mev. The cavity was covered with a cellophane window approximately 7.5 mg/cm^2.

Effect of Thickness of Mylar Films on Copper on Backscattering. A thin film or coating of a product with an average atomic number less than the base metal shows less backscattering than that obtained with the uncoated metal. In order to illustrate this, various thicknesses of Mylar film, which has a $\overline{Z}$ of 6.3 and is therefore comparable to many expected corrosion products, were placed over an uncorroded mechanically buffed copper disk and the amount of backscattering measured. Table III shows that the decrease in the amount of beta backscattering radiation obtained is a function of the thickness of the Mylar film used.

Figure 2—Exploded view of methyl methacrylate plastic assembly for the radioactive source, sample holder, and counter.



1. Methane gas proportional counter. 2. Aluminized polyester film window. 3. Plastic spacer for counter-to-specimen distance. 4. Plastic radioactive source holder. 5. Radioactive source with shield. 6. Plastic spacer for source-to-target distance. 7. Plastic sample holder. 8. Target material to be measured.

TABLE II

Effect of varying target-to-detector and source-to-target distances on backscattering of Pm-147 beta particles from a 530 mg/cm² aluminum target[a]

Distance of Target to Detector (mm)	Distance of Source to Target (mm)	Net Counts/Min[b]
0.05	2.9	500
10.05	3.9	1100
11.05	4.9	9500
12.05	5.9	19000
13.05	6.9	23000
14.05[c]	7.9	24000
15.05[c]	8.9	24000
16.05	9.9	22000
17.05	10.9	21000
18.05	11.9	19000

[a]Distance from source to detector (fixed at 6.15 mm), measured with a Gaertner Telescope Cathetometer.
[b]Background of source was 2300 c/m.
[c]Optimum distances.

TABLE III

Effect of various thicknesses of Mylar film on beta-backscattering from a mechanically buffed copper disk.

Thickness of Mylar, mils	mg/cm²	Net c/m × 10^{-3}	% Decrease in backscattering
0	0	33.8	....
.25	.88	29.0	14.2
.35	1.24	25.5	24.6
.50	1.77	24.2	28.4

[a]Total count was 100,000; BG = 2800 c/m.

TABLE IV

Effect of surface finish on backscattering of Pm-147 beta particles from one inch copper disks

Surface	Net c/m × 10^{-3}
Etched ($2H_2SO_4$, $1HNO_3$, $5H_2O$)	33.6
Sandblasted	31.1
Electropolishing (H_4PO_4)	33.8
Mechanical Buffing (FeO)	35.8
Control (not cleaned)	33.7

[a]Total count was 100,000; BG = 2800 c/m.

Effect of Cleaning of Surface and Surface Finish on Backscattering. An experiment was performed to determine whether any differences in backscattering would be obtained from copper disks with surfaces cleaned by a number of mechanical and electrochemical methods. Table IV shows that no appreciable differences in the amount of backscattering were obtained from the metal disks cleaned by three different methods. However, the samples sandblasted had 7.7 per cent less backscattering than the untreated control. This decrease in backscattering is attributed to the presence of low Z atoms of silicon and oxygen from the sandblasting on the surface of the copper, and to the presence of pits.

The sandblasting produces pits or cavities in the metal surface. It is known that as the distance of the target (pitted copper surface) to the detector increases the amount of measured backscattering decreases. Also, if the surface of a copper specimen had slight eruptions, scales, or protuberances, the amount of measured backscattering would increase. Backscattering radiation may, therefore, be used to measure the surface condition of a metal (cavities, crevices, voids, cracks, flaking, and erosion).

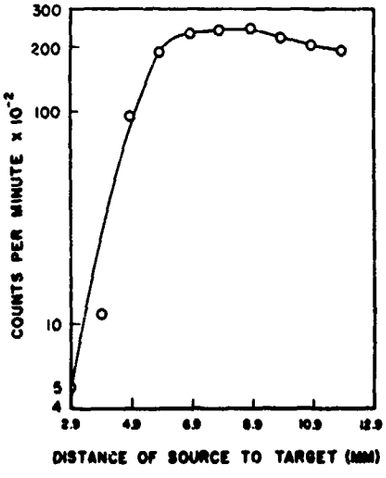


Figure 3—Effect of varying source-to-target distances of backscattering of Pm-147 beta particles from a 530 mg/cm² aluminum target.

Effect of Surface Corrosion and Oxidation on Backscattering. A number of experiments were done with the backscattering device to determine whether backscattering radiation from promethium-147 beta particles can be used to show the presence of corrosion on metal specimens. Table V lists the metals used, the surface treatments applied, and the amount of backscattering radiation obtained. The results showed definite decreases in the amount of backscattering from corroded and oxidized metal surfaces. A more detailed study on the use of radiation backscattering and radiation-induced X rays for measuring metal corrosion is described in another article.[11]

Radiation-Induced X Rays

The second method for measuring surface composition utilizes radiation-induced X rays in the specimen. When radiation (alpha particles, betas, protons, electrons, and gammas) interact with the orbital electrons of a material, X rays characteristic of the material are produced. The principle of the method is that on a clean metal surface (such as copper) a maximum number of copper Kα X rays will be induced in a certain volume of the specimen by the radiation. However, if the same metal surface is then oxidized, the volume increases due to the migration of oxygen atoms into the metal. The same volume would

then have fewer metal atoms then before, resulting in fewer copper Kα X rays produced per unit volume.

A number of radiation sources may be used such as polonium-210, americium-241, strontium-90, promethium-147, sulfur-35, thallium-204, carbon-14, hydrogen-3, cesium-137, and cobalt-60.

Beta-Particle-Induced X-Ray Instrumentation. The metal specimens to be examined are placed in a methyl methacrylate sample holder similar in design to that used for the beta backscattering radiation work. A carbon-14 radiation source having an activity of several millicuries was used. The scintillation counter had a 5 mil beryllium window and a 2 mm thick NaI (Tl) crystal coupled to a photomultiplier tube. The pulse-height analyzer was operated at 956 volts with a 5-volt window and a time constant of 50 seconds. A 1000 channel analyzer was used. Suitable single- or multichannel analyzers with high sensitivity to low-energy X rays of one to 100 Kev are also suitable. The multichannel pulse-height analyzer is connected to a scaler, rate meter or recorder.

The radiation detectors used are thin beryllium window scintillation counters and thin window or windowless proportional counters. The source, target, and detector set-up may be operated in air or in an atmosphere of hydrogen or helium for highest sensitivity.

Beta-Particle-Induced Cu Kα X Rays. Salt spray corroded and uncorroded copper specimens were measured for the intensity of the copper Kα X rays (8.047 Kev). It was found that the count rate for the corroded copper was 14 per cent less than that obtained with the uncorroded copper metal. Additional experimental work will be reported in a future article.

If the composition of a brass specimen is desired, the multichannel analyser would be set to scan for the zinc Kα X ray (8.638 Kev) and then for the copper Kα X ray. The ratio of the intensities or count rates of the two Kα X rays would show the percentage composition of zinc and brass in the specimen.

Every element has a characteristic Kα X ray of known energy. If an unknown material is to be analyzed, the entire energy spectrum is scanned. The presence or absence and the amount of any element is determined by the peak height at a particular energy level.

TABLE V

Effect of surface corrosion or oxidation of metals on backscattering of Pm-147 beta particles

Metal	Surface Treatment	Net counts/ min × 10^{-3}	% Decrease in backscattering
Copper disk (1″ diam)	Uncorroded	33.6	....
	Oxide by heating in air	31.7	5.7
	Oxide by heating in water	33.3	0.9
	Sulfide	33.0	1.8
	Nitrate by nitric acid reaction	24.7	26.5
	Chloride by hydrochloric acid reaction	29.0	13.7
	Salt spray exposure	25.9	22.9
Steel disk (11/16″ diam)	Uncorroded	17.0	....
	Oxide by heating in flame	16.2	4.7
	Salt spray exposure	13.5	20.6
Niobium foil	Unoxidized	23.9	....
	Oxide (thin coating)	23.0	3.7
	Oxide (thick coating)	21.9	8.4

Acknowledgments

Appreciation is expressed to L. Celli and T. Mackus for sample preparations, and to Dr. W. McNeill for review of the manuscript.

References

1. Symposium on Low Energy X-Ray and Gamma Sources and Applications, held in Chicago, Ill. Oct. 20-21, 1964. (In Press)
2. "The Use of Isotopes: Industrial Use," *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy*, vol. 19, 1958, United Nations, Geneva, Switzerland.
3. R. H. Müller, "Interaction of Beta Particles With Matter," *Analytical Chemistry*, vol. 29, 1957, p. 969.
4. R. H. Müller, "Interaction of Beta Particles With Matter," *Industrial and Engineering Chemistry*, vol. 50, 1958, p. 205.
5. D. C. Müller, "Interaction of Beta Particles With Organic Compounds," *Analytical Chemistry*, vol. 29, 1957, p. 975.
6. P. R. Gray, D. H. Clarey, and W. H. Beamer, "Interaction of Beta Particles With Matter; Analysis of Hydrocarbons by Beta-Ray Backscattering," *Analytical Chemistry*, vol. 31, 1959, p. 2065.
7. P. C. Aebersold and E. E. Fowler, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy*, vol. 19, 1958, p. 76, United Nations, Geneva, Switzerland.
8. R. C. Goettelman, "Measurement of Satellite Erosion Rates by Backscattering of Beta Rays," *Jet Propulsion*, vol. 28, 1958, p. 753.
9. P. Connor, W. H. Hardwick, and B. J. Laundy, "Determination of Particle Size by Beta Backscattering," *Journal Applied Chemistry*, vol. 9, 1959, p. 525.
10. G. Friedlander, J. W. Kennedy, and J. M. Miller, *Nuclear and Radiochemistry*, J. Wiley & Sons, New York (1964).
11. S. Berk, "Radiation Backscattering and Radiation-Induced X-Rays for Measuring Metal Corrosion," *Materials Protection*, vol. 4, no. 11, 1965, p. 39.

Unclassified

Security Classification

DOCUMENT CONTROL DATA · R&D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY (Corporate author) FRANKFORD ARSENAL, Philadelphia, Pa. 19137 SMUFA L3300	2a. REPORT SECURITY CLASSIFICATION Unclassified 2b. GROUP NA

3. REPORT TITLE

RADIATION BACKSCATTERING AND RADIATION-INDUCED X-RAYS FOR MEASURING SURFACE COMPOSITION AND STRUCTURE

4. DESCRIPTIVE NOTES (Type of report and inclusive dates)

Technical Research Article

5. AUTHOR(S) (Last name, first name, initial)

BERK, Sigmund (NMI)

6. REPORT DATE	7a. TOTAL NO. OF PAGES	7b. NO. OF REFS
June 1966	Four	11

8a. CONTRACT OR GRANT NO. AMCMS Code 5025.11.294 b. PROJECT NO. DA 1C024401A328 c. d.	9a. ORIGINATOR'S REPORT NUMBER(S) Frankford Arsenal Report A66-14 9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report) None

10. AVAILABILITY/LIMITATION NOTICES

Distribution unlimited.

11. SUPPLEMENTARY NOTES	12. SPONSORING MILITARY ACTIVITY
	AMSMU-RE-R

13. ABSTRACT

The use of promethium-147 beta backscattering to measure the thickness of plastic coatings on copper was determined. Backscattering radiation was also used to determine the structure and surface composition of copper specimens. A preliminary study of the use of beta-particle backscattering and beta-particle-induced X-rays for measuring corrosion is described.

DD FORM 1 JAN 64 1473

UNCLASSIFIED

Security Classification

14. KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Metals Corrosion Measuring Metal Corrosion Radiation Backscattering Radiation-induced X-ray Salt Spray Corrosion Oxidation of Metals Beta-particle Backscattering Promethium-147						

INSTRUCTIONS

1. ORIGINATING ACTIVITY: Enter the name and address of the contractor, subcontractor, grantee, Department of Defense activity or other organization *(corporate author)* issuing the report.

2a. REPORT SECURITY CLASSIFICATION: Enter the overall security classification of the report. Indicate whether "Restricted Data" is included. Marking is to be in accordance with appropriate security regulations.

2b. GROUP: Automatic downgrading is specified in DoD Directive 5200.10 and Armed Forces Industrial Manual. Enter the group number. Also, when applicable, show that optional markings have been used for Group 3 and Group 4 as authorized.

3. REPORT TITLE: Enter the complete report title in all capital letters. Titles in all cases should be unclassified. If a meaningful title cannot be selected without classification, show title classification in all capitals in parenthesis immediately following the title.

4. DESCRIPTIVE NOTES: If appropriate, enter the type of report, e.g., interim, progress, summary, annual, or final. Give the inclusive dates when a specific reporting period is covered.

5. AUTHOR(S): Enter the name(s) of author(s) as shown on or in the report. Enter last name, first name, middle initial. If military, show rank and branch of service. The name of the principal author is an absolute minimum requirement.

6. REPORT DATE: Enter the date of the report as day, month, year; or month, year. If more than one date appears on the report, use date of publication.

7a. TOTAL NUMBER OF PAGES: The total page count should follow normal pagination procedures, i.e., enter the number of pages containing information.

7b. NUMBER OF REFERENCES: Enter the total number of references cited in the report.

8a. CONTRACT OR GRANT NUMBER: If appropriate, enter the applicable number of the contract or grant under which the report was written.

8b, 8c, & 8d. PROJECT NUMBER: Enter the appropriate military department identification, such as project number, subproject number, system numbers, task number, etc.

9a. ORIGINATOR'S REPORT NUMBER(S): Enter the official report number by which the document will be identified and controlled by the originating activity. This number must be unique to this report.

9b. OTHER REPORT NUMBER(S): If the report has been assigned any other report numbers *(either by the originator or by the sponsor)*, also enter this number(s).

10. AVAILABILITY/LIMITATION NOTICES: Enter any limitations on further dissemination of the report, other than those imposed by security classification, using standard statements such as:

(1) "Qualified requesters may obtain copies of this report from DDC."

(2) "Foreign announcement and dissemination of this report by DDC is not authorized."

(3) "U. S. Government agencies may obtain copies of this report directly from DDC. Other qualified DDC users shall request through _____."

(4) "U. S. military agencies may obtain copies of this report directly from DDC. Other qualified users shall request through _____."

(5) "All distribution of this report is controlled. Qualified DDC users shall request through _____."

If the report has been furnished to the Office of Technical Services, Department of Commerce, for sale to the public, indicate this fact and enter the price, if known.

11. SUPPLEMENTARY NOTES: Use for additional explanatory notes.

12. SPONSORING MILITARY ACTIVITY: Enter the name of the departmental project office or laboratory sponsoring *(paying for)* the research and development. Include address.

13. ABSTRACT: Enter an abstract giving a brief and factual summary of the document indicative of the report, even though it may also appear elsewhere in the body of the technical report. If additional space is required, a continuation sheet shall be attached.

It is highly desirable that the abstract of classified reports be unclassified. Each paragraph of the abstract shall end with an indication of the military security classification of the information in the paragraph, represented as *(TS)*, *(S)*, *(C)*, or *(U)*.

There is no limitation on the length of the abstract. However, the suggested length is from 150 to 225 words.

14. KEY WORDS: Key words are technically meaningful terms or short phrases that characterize a report and may be used as index entries for cataloging the report. Key words must be selected so that no security classification is required. Identifiers, such as equipment model designation, trade name, military project code name, geographic location, may be used as key words but will be followed by an indication of technical context. The assignment of links, rules, and weights is optional.