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Authors

Lamb, James F.
Lee, Diana M.
Markowitz, Samuel S.

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LAWRENCE RADIATION LABORATORY
UNIVERSITY of CALIFORNIA BERKELEY

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James F. Lamb, Diana M. Lee, and Samuel S. Markowitz

Lawrence Radiation Laboratory
University of California
Berkeley, California 94720

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ABSTRACT

^{3}He activation analysis has been used for oxygen surface profile analysis of high-purity silicon. Theoretical maximum recoil ranges of ^{18}F produced by ^{3}He activation of ^{16}O were calculated for the target matrices C, Al, Ti, Cu, Ag, Te, Gd, Ta, and Au based upon total momentum transfer. The incident ^{3}He ion energies were 1-35 MeV. The predicted maximum recoil ranges in aluminum were verified experimentally at 5, 10, 20, and 25 MeV using thin Al catcher foils. The experimentally measured distribution of induced ^{18}F activity in high-purity silicon obtained using 8-MeV ^{3}He ions was then compared with prediction based upon the theoretical curves and with results in the literature from similar experiments using ^{4}He activation. An oxygen concentration of about 1 ppm in bulk silicon was found, and the maximum recoil range was determined to be about 0.5 mg/cm^{2}.

INTRODUCTION

The use of ^{3}He as the activating particle for charged particle activation analysis has been developed (1) because the low binding energy of the ^{3}He nucleus enables nuclear reactions to proceed with minimum energy bombardment, because the two-proton, one-neutron configuration of ^{3}He leads to a wealth of possible reaction products, and because most of the reactions induced give rise to neutron deficient products whose decay characteristics are favorable for detection. Considerable effort has been devoted to proving the practicality of the system (2-5) and to determining its applicability to various analytical problems (5,6).

In this report we present a method for predicting and interpreting results of surface profile analysis by charged particle activation. The ability to preferentially activate sample constituents which lie within the first few mg/cm^2 of surface of a sample and to determine these constituents with high sensitivity is a feature almost unique to charged particle activation, and because of the advantages mentioned above, ^{3}He is almost uniquely suited as the incident particle.

DISCUSSION

The energy with which the product of a nuclear reaction recoils from the target depends upon the reaction mechanism. The maximum recoil energy will be obtained in <u>compound nucleus</u> reactions in which the incident particle and the target nucleus are fused into an intermediate product having the total system linear momentum. The compound nucleus then de-excites, predominantly by particle emission which is symmetric in the forward and backward directions.

The averaged result is almost total linear momentum transfer to the recoiling product. Direct reaction mechanisms involve the transfer of nucleons between the incident particle and the target nucleus without formation of a reaction intermediate, with the result that much of the system linear momentum may remain with the emitted particles. ^{3}He induced reactions among the light elements have been shown to proceed by both mechanisms.

For the purpose of surface profile analysis by ^{3}He activation, it will be practical to assume complete linear momentum transfer and to calculate the recoil energy of the reaction product from (7)

$$E_r = \left[\frac{A_i A_p}{(A_i + A_t)^2} \right] E_i$$

where E_r is the recoil energy, and A_i, A_p, and A_t are the mass numbers of the incident particle, product nucleus, and target nucleus, respectively. E_i is the incident beam energy.

Using this calculated value of the maximum recoil energy, it is possible to estimate the maximum recoil range for the reaction products in various target and absorbing media. Steward (8) has developed a Fortran IV computer code for calculating the total pathlength ranges of any heavy ion in nongaseous stopping media which incorporates available experimental range data with various theoretical treatments, each within its range of validity.

The maximum recoil energy of ^{18}F produced as the sum of the reactions $^{16}O(^{3}He,p)^{18}F + ^{16}(^{3}He,n)^{18}Ne \rightarrow ^{18}F$ is

$$E_r = \left[\frac{(3)(18)}{(3+16)^2} \right] E_i = 0.15\ E_i \quad .$$

The values of E_r obtained for $1 < E_i < 35$ were used with Steward's computer code to calculate the maximum ranges of recoil ^{18}F in various stopping media shown in Figure 1.

EXPERIMENTAL

1. Test with Mylar and Aluminium. Before applying the theory to analysis of a sample, we wished to verify the theoretical estimates with an experimental test. Targets were prepared by decking together 5 upstream aluminum recoil catcher foils, a mylar (33% O, 66% C, 1% H) target foil, and 14 downstream aluminum recoil catcher foils. The aluminum foils, each 100 $\mu g/cm^2$ thick, and the mylar foil, 1 mg/cm^2 thick, were placed in individual 0.005-inch thick aluminum envelopes having 1-inch diameter center holes. The entire target decks were compressed to a total thickness of about 1 cm to minimize scattering losses. Target decks were bombarded for 20-minutes each at beam currents of about 0.05 μA at energies of 5, 10, 20, and 25-MeV at the Berkeley Heavy Ion Linear Accelerator. The ^{18}F activity of each foil was determined by counting the 511-keV annihilation radiation resulting from the 110-min positron decay using a NaI(Tl) detector coupled to a multi-channel analyzer. No chemical separations were employed.

The resulting activities from a typical bombardment at 10-MeV are shown in Figure 2. Foils 4, 3, 2, and 1 to the left of the mylar foil, designated M, are upstream aluminum recoil catcher foils. The approximately constant activity in downstream foils 8 through 13 may be interpreted as the background resulting from activation of the natural oxide coating on each aluminum catcher foil. The upstream catcher foils farthest from the mylar have lower background activities because of uncompensated recoil losses. The results for the forward direction recoils from all four bombardments are shown together in Figure 3 with the data recalculated in terms of the percent of the total forward recoils which pass through each catcher foil. The background activity from the aluminum surface oxide has been subtracted from each curve. The maximum recoil range may be estimated from the figure as the ordinate intercept. Since each catcher foil is 100 $\mu g/cm^2$ thick, the ordinate scale corresponds to an aluminum thickness of 0 - 1 mg/cm^2.

These experimental maximum ranges compare quite well with the theoretical maxima estimated from Figure 1. The distribution of recoils in the catcher foils is of significance only relatively among the different bombardments because the large thickness of the mylar compared to the range of the ^{18}F causes distortion in all curves in favor of lower energy recoils.

2. Surface Oxygen Profile in Silicon. The utility of the recoil-range calculations for surface analysis is demonstrated by surface profile analysis of high-purity silicon. The oxide coating on a freshly prepared silicon surface is known to be restricted to a very thin (10 Å) layer (9). Saito, et al. (10) obtained a surface profile analysis of high purity silicon using ^4He activation. Because the energy thresholds for the (α,pn), (α,d), and $(\alpha,2n)$ reactions are all above 20-MeV, a correspondingly high-energy beam must be used for ^4He-induced reactions. This results in recoil penetration by the

reaction products to greater depths than those obtained with use of ^{3}He as the activating particle at much lower energies. Saito, et al. found recoil ^{18}F at depths of about 10 mg/cm^2 in silicon following ^{4}He activation at energies up to 40-MeV. Most of the activity was contained within the first 5 mg/cm^2.

From Figure 1 we may estimate a maximum recoil range in silicon for the ^{3}He-induced ^{18}F, (the ^{18}F ranges in Si may be approximated by the Al ranges without appreciable error because the ranges for Si are within a few percent of those for Al at the energies encountered here). At a ^{3}He energy of 8-MeV this range is approximately 0.5 mg/cm^2. The predicted result was tested by irradiating a disc of high purity zone-refined silicon which had been prepared for fabrication of a semiconductor particle detector. The surface was ground flat on a glass plate using 600-mesh silicon carbide powder and then washed with water and alcohol immediately before irradiation. The surface was protected from down-streaming contamination in the accelerator by a thin gold cover foil during irradiation. The target was bombarded for 45-min at 8-MeV at a beam current of 0.1 μA. Decay of the activity induced in the silicon was followed by counting the 511-keV annihilation radiation until all short lived activities had decayed to a negligible level, leaving only the 110-min activity, ^{18}F. A thin layer of the irradiated surface was then removed by surface grinding on the glass plate, its thickness determined by weight difference, and the activity remaining on the silicon disc redetermined. This process was repeated, using fresh grinding compound each time, until the activity per mg of Si removed remained constant.

Saito, et al. obtained an oxygen content of the order of 1 ppm in bulk high purity Si. We, therefore, have calculated our data in terms of ppm oxygen for convenient comparison.

The effective range of the ^{3}He ion in Si at 8-MeV is about 12.6-mg/cm^2. The beam energy, after traversing the first 2.5 mg/cm^2 of surface (roughly the total Si removed by grinding), would be degraded from 8-MeV to about 7.2-MeV. The difference in the average thick-target cross section for the ^{16}O + ^{3}He → ^{18}F reaction at these incident energies, 8- and 7.2-MeV, amounts to less than about 10% of the 8-MeV average thick target cross section. We, therefore, have used the activity induced in a thick Ta_2O_5 comparison standard, also bombarded at 8-MeV, to calculate each oxygen content. The ppm oxygen shown in Figure 4 were calculated as the mg oxygen per mg of silicon remaining after each surface grinding; the effective thickness of the Si was taken as that thickness (in mg/cm^2) reached by the ^{3}He beam down to $\approx$ 2 MeV (where no further nuclear reaction takes place on ^{16}O).

RESULTS AND CONCLUSIONS

By very simple calculation, a predicted maximum recoil energy has been determined for recoiling ^{18}F induced in oxygen by ^{3}He bombardment. This predicted energy, used with available range-energy calculations, yields predicted ^{18}F recoil ranges which, in Al and Si, agree very well with experimental data. The method provides a means of estimating the amount of material which must be removed from the surface of an irradiated sample in order to obtain data for bulk material only. Furthermore, the presence of induced ^{18}F activity significantly higher than that in the bulk material at depths below the calculated maxima may be interpreted in terms of the thickness of the oxide layer. With use of an accurate mechanical grinder capable of removing a known thickness of surface material, a very accurate surface profile of any suitable sample matrix could be rapidly determined. The method is not limited to either oxygen as the target nucleus, aluminum or silicon as the target matrix, or even to ^{3}He as the incident particle, though the last is recommended because ^{3}He reactions may be induced at low bombarding energies resulting in smaller momentum transfer and smaller recoil distortion of the surface profile.

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FIGURE CAPTIONS

Fig. 1. Range of ^{18}F in various stopping media. The upper scale is the ^{3}He energy corresponding to the total momentum transfer ^{18}F energy shown on the lower scale as calculated by Eq. (1).

Fig. 2. Distribution of recoiling ^{18}F in 100-μg/cm^{2} Al catcher foils. M designates mylar target. ^{3}He energy = 10-MeV.

Fig. 3. Percent of recoil ^{18}F passing through each catcher as a function of the number of 100-μg/cm^{2} Al catcher foils. ▼ = 5-MeV, ◆ = 10-MeV, ■ = 20-MeV, ▲ = 25-MeV.

Fig. 4. Surface oxygen profile of high-purity silicon determined by ^{3}He activation.

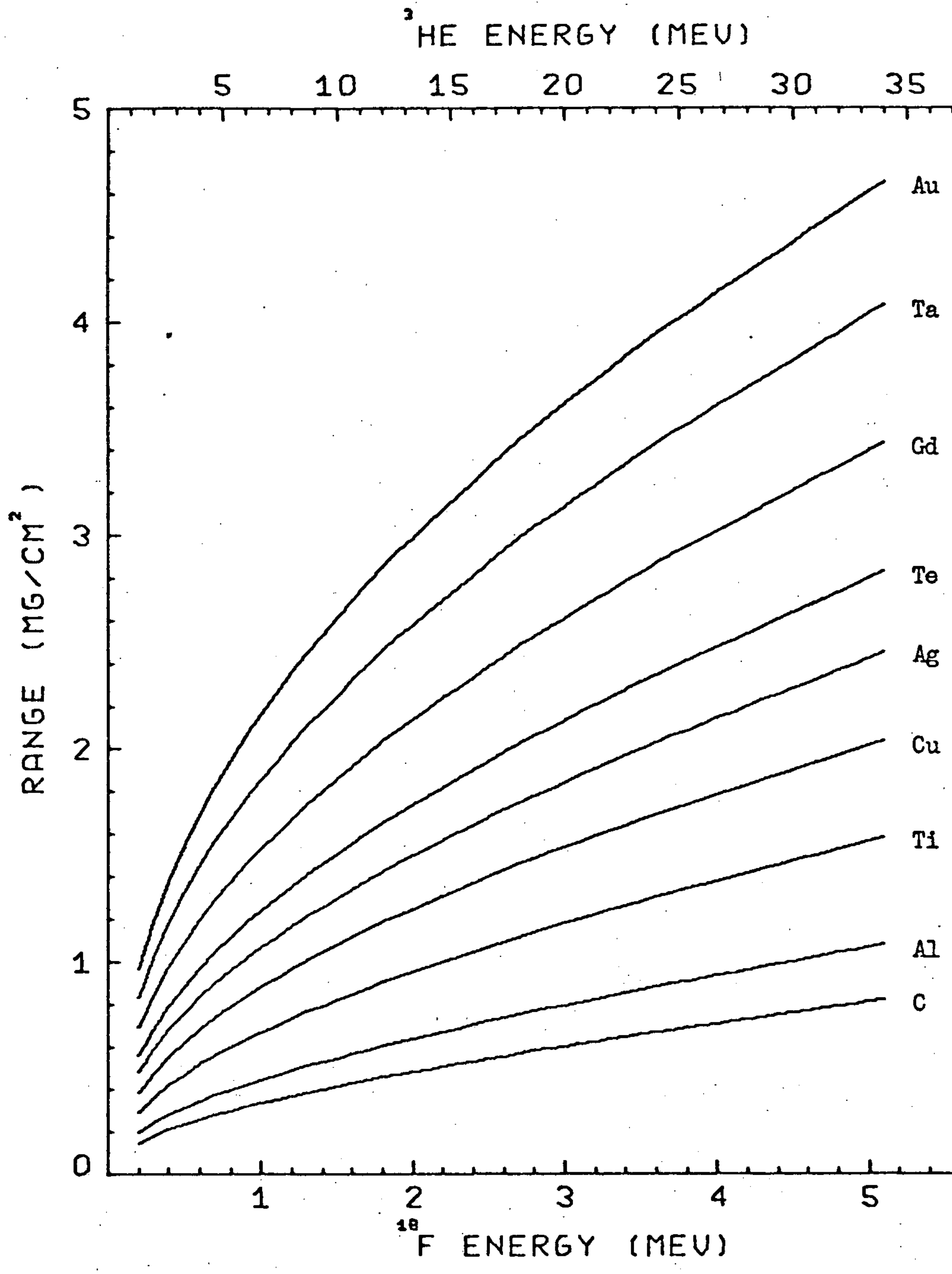

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Fig. 1.

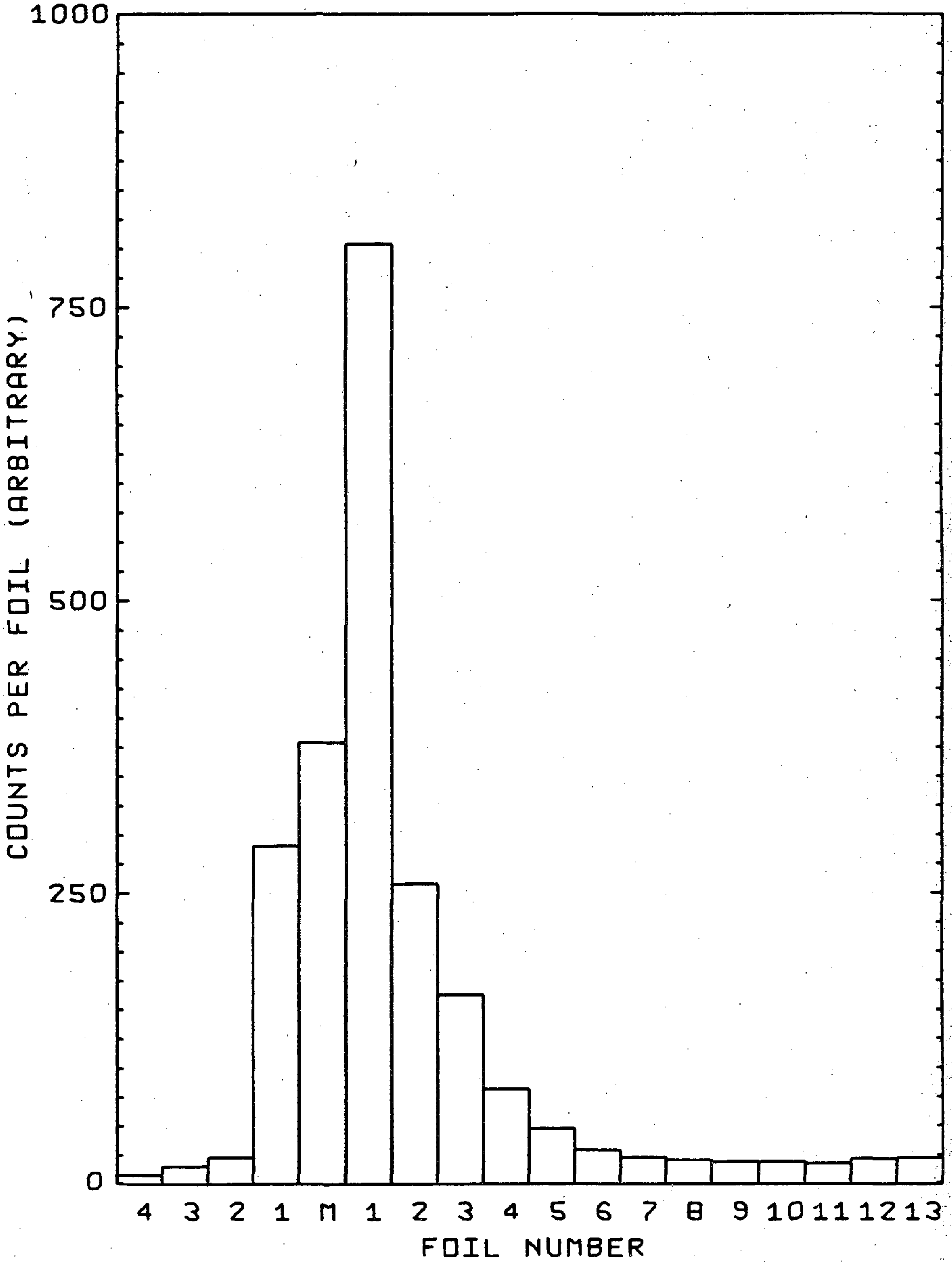

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Fig. 2.

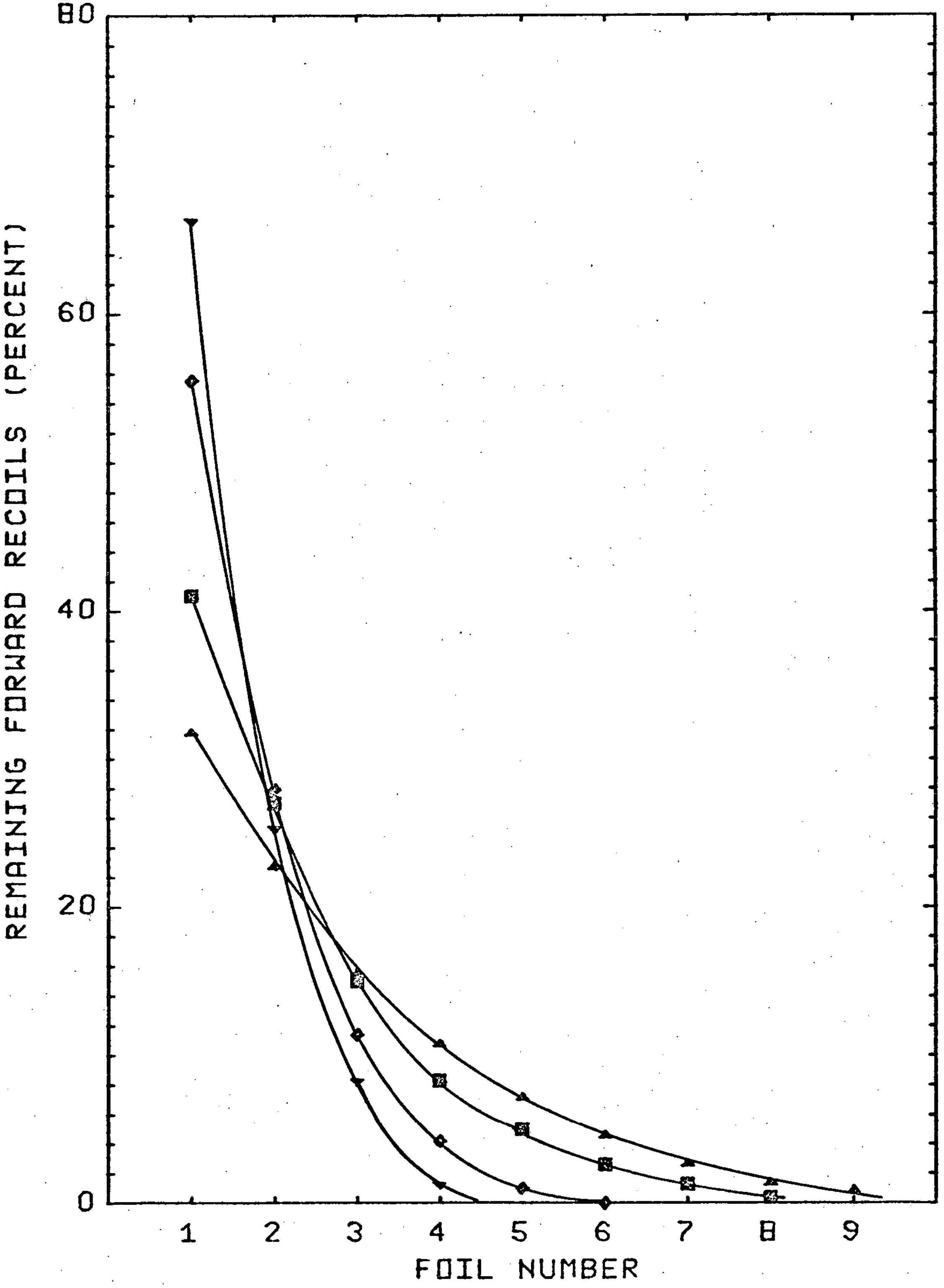

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Fig. 3.

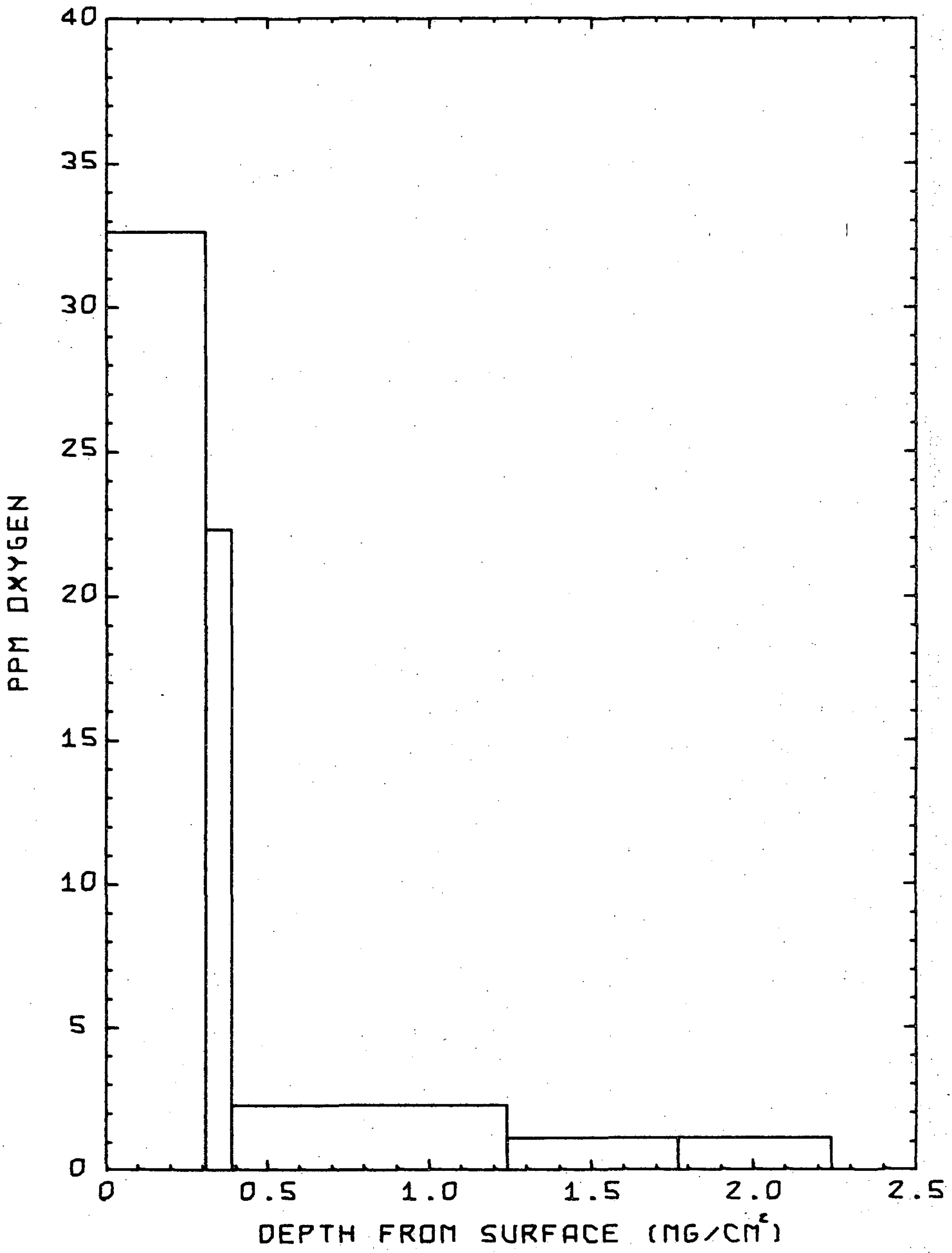

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Fig. 4.

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LAWRENCE RADIATION LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720