

RANGE AND DEPTH DOSE DISTRIBUTION OF LOW ENERGY CHARGED PARTICLES IN DOSIMETER GLASSES

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Abstract—A new method for the direct determination of particle ranges and depth dose distributions in silver-activated phosphate glasses is based on the successive removal of extremely thin surface layers from the exposed glass by chemical etching ("peeling") and measurement of the residual radiophotoluminescence between successive etchings. Glass composition, etching chemicals and etching speed can be varied within wide limits. The experimental technique, using Yokota-type dosimeter glasses and 28% NaOH at 60°C (etching speed 0.12 μ /min) is briefly described. As an example of the practical application of the method, measurements using several types of radiation sources (aqueous solutions of ^3H , ^{63}Ni and ^{35}S , solutions and thin and thick solid sources of ^{239}Pu , ^{237}Np and ^{235}U , monoenergetic protons, deuterons and He^+ ions in a wide energy range) have been made.

Accuracy, possibilities and limitations of the method are briefly discussed. Possible sources of error are: discoloration of the glass because of very high surface doses; uncertainties in the determination of the etching speed; etching speed along charged particle tracks higher than the bulk etch rate for ions of very high LET.

1. INTRODUCTION

The calculation of the range of low energy electrons and ions in solids is difficult, mainly because charge exchange effects become important and elastic scattering causes an effective range which is smaller than the sum of the total distances measured along the path of the particle. Different theoretical approaches to convert the path length calculated by the Bohr-Nielson equation^(1, 2) into the experimentally measured projected range^(3, 4) may lead to different results, and the agreement with experimental values has to be proved.⁽⁵⁾

During recent years, several more or less indirect methods for the measurement of average and maximum ranges have been used, for instance by determination of changes in the refraction index of quartz surface layers caused by ion bombardment;⁽⁶⁾ by elastic proton scattering at the embedded heavy ions in low Z target materials;⁽⁷⁾ by determination of the line-shape of the emitted α particles;⁽⁸⁾ and by

luminescence excitation⁽⁹⁾ or deterioration⁽¹⁰⁾ measurements in thin fluorescent layers.

More direct methods have also been applied, such as the use of stacks of thin metal foils,⁽¹¹⁾ and the chemical determination of ranges of radioactive ions in some metals (Al, W) by chemical removal of uniform thin surface layers and measurement of the residual radioactivity of the target material.⁽¹²⁻¹⁶⁾ In a similar study, the range of ^{134}Cs ions in germanium was determined by etching of the Ge crystal.⁽¹⁷⁾ It was found, however, that approximately 25% of the ^{134}Cs could not be removed by prolonged etching because of either an abnormal range of the ions or non-uniform etching of the crystal.

It would, however, be desirable to apply this relatively simple, fast and direct "peeling" method for range studies without the drawbacks of the earlier methods, in particular:

- (a) Without restriction to range measurements of radioactive ions. This is, for instance, possible by using irreversible changes in the physical properties of the target material, which are proportional to the energy transferred, localized to

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- the point of energy transfer and can easily be measured.
- (b) By the use of a target material whose composition can be varied within wide limits.
 - (c) Whose homogenous structure guarantees a uniform etching process.

All these specifications can be met within certain limitations by the use of silver-activated phosphate glasses as target material. Ionizing radiation causes the formation of quasi-permanent luminescence centers (radiophotoluminescence, RPL) in these glasses, which can be detected by simple fluorimetry. It is known that phosphate glasses are etched very uniformly and the glass composition can be varied within certain limits. Glasses containing Ag concentrations between approx. 0.1 and 10% and phosphates of Li, K, Na, Ba, Mg, Al, BeO, B₂O₃, etc., have been described.⁽¹⁸⁾

2. EXPERIMENTAL PROCEDURE

In general, a range determination by this method consists of the following procedure:

I. The clean dosimeter glass with known background (predose) luminescence is first exposed to the ionizing radiation. In case of monodirectional electrons or positive ions from accelerators, or α or β particles from radioactive planes, glass plates or blocks with a plane surface are most suitable. If the glass is exposed by submersion in a uniform medium, such as an α or β active solution, other shapes, including glass rods or spheres, may also be used.

II. By waiting a prescribed period of time or by heat treating the glass (in most dosimeter glasses about 15 min at 150°C), the optimal stable RPL is obtained,⁽¹⁸⁾ which is measured in a proper glass dosimeter reader (365 m μ excitation, RPL maximum around 620 m μ). The predose effect is subtracted from the reading. The RPL should be intense enough to allow measurements down to at least about 1% of the initial radiation effect. On the other hand, it should not be too intense. Local doses higher than several thousand rads produce a non-linear response because of glass discoloration (for details on this effect, see ref. 19). Discoloration does not affect the results of range measurements, but can cause errors in depth dose determinations.

III. The glass is etched for a known time in a solution of known composition and temperature. Etching conditions depend strongly on the expected range and the glass composition. The thickness of the removed surface layer is determined from earlier calibration data and will depend on the expected range of the ionizing particle and the glass composition.

IV. After rinsing and drying, the RPL measurement is repeated and the predose subtracted. This cycle of etching and reading is repeated until prolonged etching causes no further reduction in the RPL reading.

V. The logarithm of the residual RPL in percent of the original RPL is plotted as a function of the thickness or weight of glass removed by the etching process. The resulting graph then corresponds to the well-known intensity over absorber thickness plots for α , β , and X radiation. In other cases, other graphic representations may have advantages. By differentiation information on the depth dose distribution can be obtained.

There are several methods by which the etching speed can be determined, such as:

- (a) Measuring the weight reduction of a dosimeter glass block after prolonged etching.
- (b) Direct measurement of the change in glass thickness by the use of a thickness gauge after prolonged etching.
- (c) Microscopic methods, either directly, or by measurement of the diameter of etch pits which result after bombardment of the glass with particles of very high LET such as fission fragments, followed by prolonged etching. If an etch pit is large enough, half of the diameter increase per unit of time corresponds within a few percent to the bulk etch speed of the glass surface. If the etching solution is sufficiently agitated, the thickness of the removed glass layer will be proportional to the etch time. Etching during ultrasonic agitation guarantees fast replacement of the exhausted etching agent at the glass surface by a new etching agent.

All chemicals that attack the glass surface at a sufficient speed can be used. In cases of the usual dosimeter glasses this may be inorganic

acids such as HF, bases such as NaOH, or organic complex formers. The etching speed depends mainly on

- (a) the glass composition,
- (b) the etching agent,
- (c) the concentration of the etching chemicals, and
- (d) the etching temperature.

The etching kinetics of some dosimeter glasses

compared with glasses kept at room temperature have been found.

3. RESULTS

As an example for the effect of monoenergetic charged particles, the residual RPL in a glass containing 53.5% O, 33.3% P, 4.6% Al, 4.2% Ag, 3.6% Li and 0.8% B (Toshiba, Tokyo) has been measured as a function of the

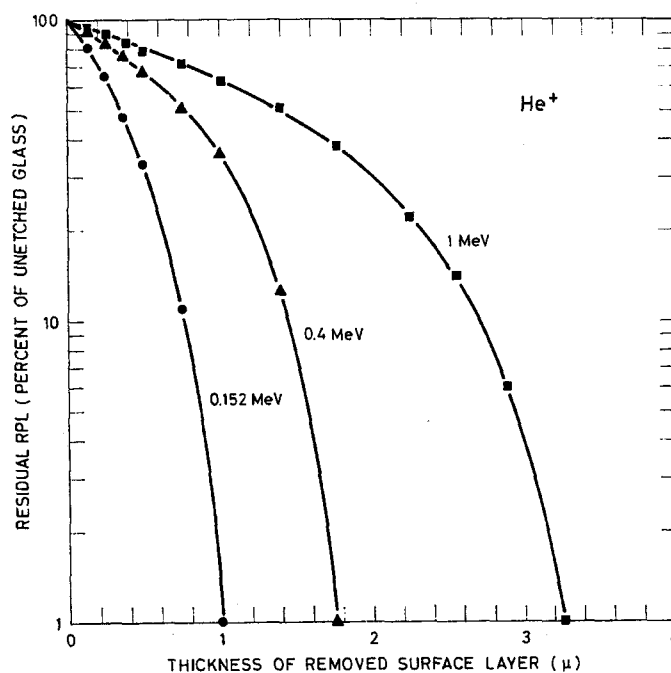


FIG. 1. Residual RPL of a dosimeter glass irradiated with He^+ ions of different energies as a function of the thickness of the removed surface layer, normalized for reading of the unetched glass = 100.

have been described in detail in another publication.⁽²⁰⁾ Using the Yokota-type dosimeter-glass⁽²¹⁾ and 28% NaOH at 60°C, for instance, an etching speed of about 0.12 μ/min has been obtained. Even extended etching of unexposed glasses or glasses irradiated with penetrating radiation does not affect the RPL reading. In order to investigate the possible effect of RPL center diffusion in the glass on the measured ranges, glasses have been kept at 200°C for several hours. No differences in the results

thickness of the removed surface layer for He^+ ions of 152 keV, 0.4 MeV and 1 MeV. The same glass type has been used in all experiments, the fluorimetric evaluation has been done in a Toshiba FGD-3B reader. Similar results have been obtained with protons and deuterons in the 0.15 to 1.8 MeV range. The obtained ranges were slightly higher than the reported ranges for these particles in aluminium above 1 MeV. The exposures have been made by placing the glass inside the accelerator tube

in the target area of Van de Graaff and Cockroft-Walton type accelerators. Even at very small beam currents, exposure times of less than a second were sufficient. At higher beam currents or exposure times, a yellowish discoloration of the glass surface was visible to the naked eye. In this case the usual slope of the curves (Fig. 1) was changed. Because of the superimposed effects of ultraviolet light absorption in the uppermost discolored layer (absorption maximum around $320\text{ m}\mu$) and the total RPL

bremsstrahlung. After exposure to $^{90}\text{Sr}/^{90}\text{Y}$, as it was expected, no decrease in the RPL intensity has been observed by the removal of more than 20μ .

In glasses exposed to mixed α , β and γ emitters the background may be quite high. But also in such cases an α range determination is possible (Fig. 3). The differences obtained for glasses exposed to a ^{235}U foil of 30μ thickness and a ^{237}Np layer of 0.34μ thickness electro-deposited on steel is caused in part by the

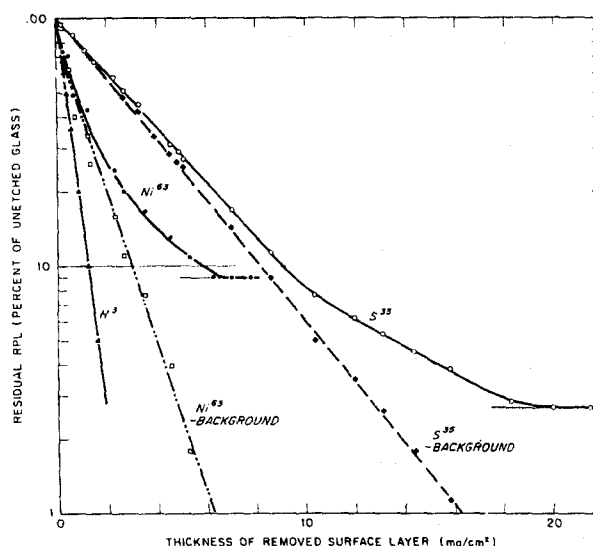


FIG. 2. Residual RPL of a dosimeter glass exposed to solutions of different β emitters as a function of the thickness of the removed surface layer, normalized for reading of the unetched glass = 100.

decrease during etching, the measured RPL decrease was at first slower—in some cases even an increase was observed—and then, close to the maximum range, faster than given in Fig. 1.

In another experiment, glass blocks have been immersed in aqueous solutions of soft β emitters. In Fig. 2, the results for ^3H (maximum β energy 18 keV), ^{63}Ni (67 keV) and ^{35}S (167 keV) containing solutions before and after subtraction of the background is given. It can be seen that even in the case of ^3H the range can easily be measured. The background may be caused by γ -emitting contaminations and

difference in α energies, the ^{237}Np α energies being somewhat higher, in part by the different energy distributions because of different source thickness.

The effect of source geometry can also be seen in Fig. 4, where glasses have been exposed to ^{239}Pu in aqueous solution, in a thin and in a thick solid source. The background effect is higher in the glasses exposed to the solution and the thick source because of the higher contribution of penetrating γ radiation to the glass surface dose. After subtracting the background effect, however, the data for the thin source and the solution agree.

4. DISCUSSION

Also numerous other measurements using the described technique have been made in order to establish the usability of the method. The minimum and maximum ranges measured and the accuracy obtained in the experiments described do not represent its limitations. More accurate determination of the etching speed and more constant etching conditions should, for instance, result in an accuracy and reproducibility of better than $\pm 5\%$.

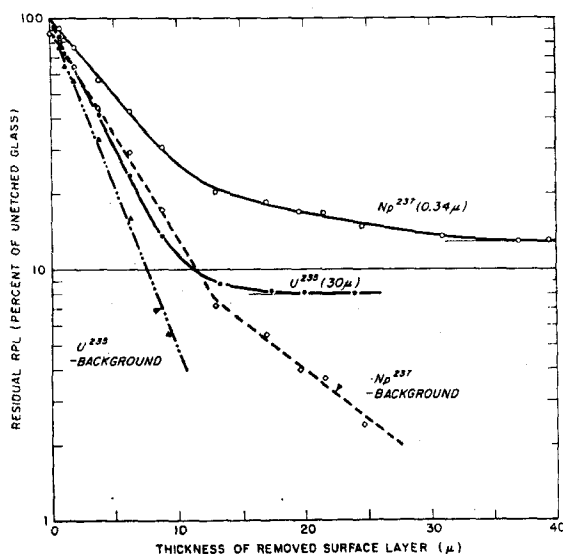


FIG. 3. Residual RPL of a dosimeter glass exposed to a thin ^{237}Np and a thick ^{238}U foil as a function of the thickness of the removed surface layer, normalized for reading of the unetched glass = 100.

The lower limit for range measurements is not given by the etching technique because the thickness of the removed layers can be made extremely small by reduction of etching time and/or concentration and temperature of the etching solution. It will be limited by the fact that the average volume dose of the glass block or plate will become smaller with decreasing range of the particles and discoloration may already occur in a thin surface layer while the total RPL of the glass is still too low for accurate measurements. If stack arrangements of exposed glass plates are used, the limit may be

extended because of the increased percentage of the exposed glass volume. For the same reason, glass powders could be submersed in radioactive solutions. There is no upper limit, but at ranges exceeding several tens or hundreds of microns, other even simpler direct methods may be superior.

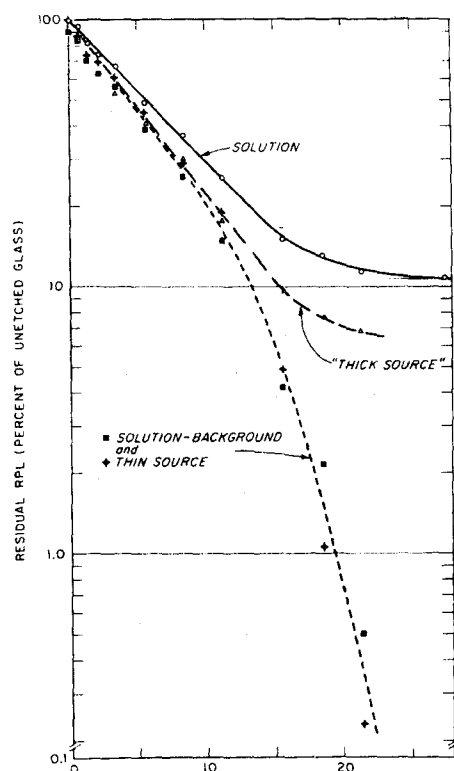


FIG. 4. Residual RPL of a dosimeter glass exposed to a solution, a thin (evaporated solution) and a thick source (metal sheet) of ^{239}Pu as a function of the thickness of the removed surface layer, normalized for reading of the unetched glass = 100.

When the ionization density (LET) along the track of the particle exceeds a critical value, there will be radiation damage produced in the glass. Preferential etching will take place along the particle track. In this case an etch pit is formed which can be seen in an optical microscope and the surface removal method can no longer be applied. The LET limit depends on

glass composition and etching conditions and is not known for the glasses under study, but fission fragments are certainly above and α particles below this limit.

If the decrease in RPL intensity between successive steps of etching is used for depth dose distribution measurements, the LET dependence of the RPL has to be taken into account. It has been demonstrated⁽²²⁾ that the dose response of RPL glasses begins to decrease at about 1 keV/ μ , corresponding to a proton energy of 100 MeV, and is at 100 keV/ μ (~ 200 keV protons) less than 20% of the low LET sensitivity. It is, however, possible to deduct at least approximate depth dose distributions from the experimental data.

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REFERENCES

1. N. BOHR. Kgl. Danske Videnskab. Selskab., Mat.-fys. Medd. **18**, 8 (1948).
2. K. O. NIELSEN. In Electromagnet. Enrichm. of Isotop. and Mass Spectrometry, M. L. Smith (ed.), Acad. Press, New York, 1956, p. 68.
3. J. LINDHARD and M. SCHARF. *Phys. Rev.* **124**, 128 (1961).
4. O. S. OEN, O. K. HOLMES and M. T. ROBINSON. *J. Appl. Phys.* **34**, 302 (1963).
5. L. MORBITZER and A. SCHARMANN. *Z. Phys.* **185**, 488 (1965).
6. R. L. HINES and R. ARNDT. *Phys. Rev.* **119**, 623 and **120**, 1626 (1960).
7. D. POWERS and W. WHALING. *Phys. Rev.* **126**, 61 (1962).
8. B. DOMEIJ *et al.* *Arkiv for Fys.* **24**, 399 (1963).
9. C. FELDMANN. *Phys. Rev.* **117**, 455 (1960).
10. L. MORBITZER and A. SCHARMANN. *Z. Phys.* **177**, 174 and **181**, 67 (1964).
11. O. SELIG and R. SIZMANN. *Nukleonik* **8**, 303 (1966).
12. V. A. J. VAN LINT, R. SCHMITT and C. S. SUFFREDIN. *Phys. Rev.* **121**, 1457 (1961).
13. J. A. DAVIES *et al.* *Can. J. Chem.* **38**, 1526 and 1535 (1960) and **39**, 601 (1961).
14. J. A. DAVIES *et al.* *Arkiv for Fys.* **24**, 377 (1963).
15. I. BERGSTROM *et al.* *Arkiv for Fys.* **24**, 389 (1963).
16. J. UHLER *et al.* *Arkiv for Fys.* **24**, 413 (1963).
17. M. M. MREDOV and M. M. OKUNEVA. *Dokl. Akad. Nauk SSR*, **113**, 795 (1957).
18. K. BECKER. *Symp. on Solid-State and Chemical Dosimetry*, IAEA Vienna, 1966, and IAEA Atomic Energy Rev. **5**, 43 (1967).
19. K. BECKER. *Health Phys.* **11**, 523 (1965).
20. K. BECKER, USNRDL-Tr 904 (1965) and *Health Phys.* **12**, 769 (1965).
21. R. YOKOTA, S. NAKAJIMA and E. SAKAI. *Health Phys.* **5**, 219 (1961).
22. E. TOCHILIN *et al.*, Unpublished.