DETERMINATION OF THE ^{235}U CONTENT IN ENRICHED SAMPLES BY THE FISSION TRACK REGISTRATION TECHNIQUE

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ABSTRACT

The content of ^{235}U in enriched samples was determined using the solid state registration technique with automatic counting Uranium solutions in diluted nitric acid have been irradiated using the thermalized neutrons of a ^{252}Cf source Enrichments around 20% can be measured with 2% precision

1 – INTRODUCTION

This paper deals with an investigation of the content of ^{235}U in enriched samples using the solid state registration technique

The registration of fission fragments in insulating materials[4] such as mica glass and certain synthetic plastics (Makrofol Lexan etc) has been used during the last ten years for a number of applications in nuclear technology[5 7]

In our laboratory the fission track method has been developed for the analysis of ^{235}U content by irradiating samples of uranium with the thermalized neutrons of a ^{252}Cf source[1] The uranium was in solution form with Makrofol strips immersed in it (wet method)[6]

The same technique described in this paper can also be applied for analysis of uranium in sea water and also for the determination of the fissile material content in solutions including highly radioactive fuel reprocessing wastes without resorting to chemical separation of the fission products or any other purification

2 – EXPERIMENTAL PROCEDURE

The method is based on the principle that only ^{235}U is fissionable with thermal neutrons so if a standard uranium sample with known isotopic abundance and a sample whose enrichment we want to know are irradiated with thermal neutrons it is possible to calculate the ^{235}U content of the latter by comparing the total number of fission tracks registered in a Makrofol foil The Makrofol is much more sensitive to the damage produced by the fission fragments than to any other radiation So there is no background radiation to be considered

The total number of tracks registered T is proportional to the number of fissile atoms (in this case proportional to ^{235}U atoms since the neutrons are thermalized) to the fission cross section σ_f to the thermal flux ϕ and to the irradiation time t

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The proportionality constant K is identified as the absolute track registration efficiency in the solution if the geometrical conditions are maintained in the irradiations

Consequently $\epsilon = (T/T_s)(C_s/C)\,\epsilon_s$ where T and T_s are the track densities of the sample and standard C and C_s are the concentration of uranium in the sample and in the standard and ϵ_s is the enrichment of the standard

The uranium solutions (in diluted nitric acid) have been irradiated in cylindrical lucite boxes ($\phi = 5$ cm $h = 5$ cm) with the Makrofol strips of 1.8 cm x 2.5 cm and 10 μm immersed in them

The neutrons used were provided by a 352 μg source of ^{252}Cf and were thermalized with water (figure 1) The thermal flux at the best irradiation position previously determined was measured with gold foils and was found to be 2.0×10^6 n/cm^2 s The cadmium ratio measured for natural uranium was about 5 The fast fissions of ^{238}U produced by non thermalized neutrons have been eliminated by making irradiations with and without a cadmium foil of 1 mm thickness

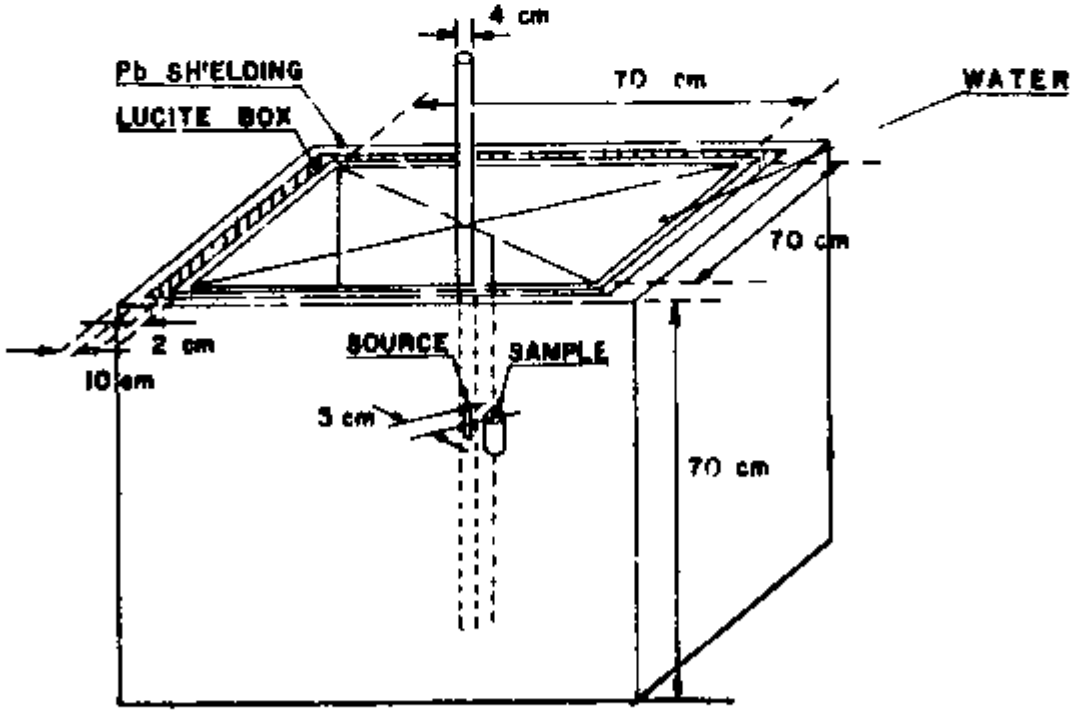

Figure 1 — Experimental arrangement of ^{252}Cf source used for the irradiation

The irradiation time was about 1 h and the concentration of the solutions was chosen in order to get convenient statistics for the number of tracks. The irradiation time with the cadmium cover was about 4 h.

After the irradiation the Makrofol strips were rinsed with water and diluted nitric acid for a few minutes and etched in KOH (35%). At the beginning the temperature of the etching solutions was 60°C and the etching time was 20 min. As it was difficult to maintain the temperature of the etching bath[6] with variations less than 0.4°C the reproducibility of data obtained with the same etching conditions was around 2%. For this reason we have changed the etching bath temperature to room temperature and the time to 24 h. In all the results a confidence level of 95% was used for the calculation of the variances.

The etching solution was stirred all the time for uniformity of the attack. The Makrofol was then dried and scanned.

The total number of tracks was counted in an automatic discharge chamber[2]. Initially 1300 V were applied in order to get all the holes in the Makrofol passed through. Then a count in a scaler was made applying 550 V in a predetermined area (2.54 cm^2). In figure 2 we can see the holes produced in a mylar foil where it can be seen that the effect of spurious discharges can be neglected. For scanning conditions a reproducibility around 0.2% was found.

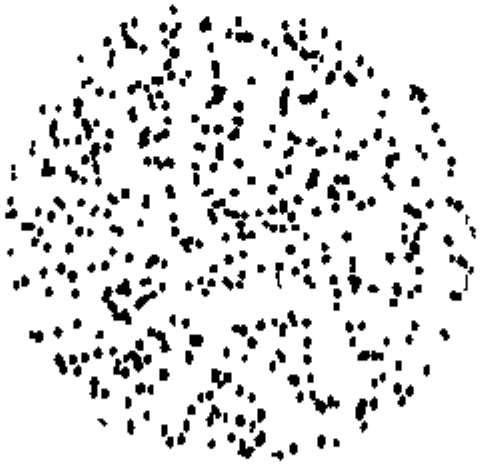

Figure 2 — Holes produced in a mylar foil after the discharge in the irradiated Makrofol.

3 – RESULTS AND DISCUSSION

In order to check the effects of self absorption in the solution when the uranium concentration changes we have performed several irradiations with different concentrations of natural uranium solutions in diluted nitric acid The concentrations changed from 1 mg/ml to 20 mg/ml This range was selected because the enriched solutions had concentrations more or less in this interval The irradiation time has changed from 20 min to 1 h

The track density obtained was plotted against uranium concentration and the data have been statistically treated(8) in order to draw the best straight line through the data points (shown in figure 3)

We have found in the literature[3] a discussion about the non proportionality between track densities and thermal neutron fluence when uranyl acetate solution is used The explanation given was the formation of radiocolloid and pseudo radiocolloid which manifests itself in a solid state track detector as a cluster

In our case using uranyl nitrate and maintaining the pH constant during all the irradiations at a value of 1 4 (the radiocolloid formation is very dependent on the solution pH) we have not seen any cluster after scanning several foils of Makrofol under the microscope

Moreover the straight line obtained (figure 3) also shows that there is a good linearity when the uranyl nitrate is used Several irradiations with the same uranium solution have been performed and the reproducibility obtained was 1 5%

Uranium solutions with concentrations from 1 to 40 mg/ml and known ^{235}U content (from 0 4% to 19 98%) have been irradiated in order to obtain a calibration curve

The results obtained for the several enrichments are shown in figure 4

Only the 0 4% 3 396% and 19 98% enrichment samples have certificates of the ^{235}U content The others have been prepared mixing the enriched solutions with natural uranium solutions Of course the calibration curve is affected by the precision of the standards used and a better one can be obtained depending on the standard solutions

Using this calibration curve the samples can have the enrichment determined with a precision of 2 7 5% for enrichments between 20% and 2% For better results at low enrichments another calibration curve must be made

The errors are due to various operations such as weighing during the preparation of the solutions, reproducibility of the irradiation conditions correction for fast fissions and statistical fluctuations in the number of tracks measured

When the uranium is irradiated as a solution the fissile atoms will be uniformly distributed giving a uniform track density that will reduce the scanning time since it is not necessary to scan the whole Makrofol foil

As Makrofol does not contain any uranium the interference of its background is eliminated and the technique described can be used even for extremely low uranium concentrations This can be an important fact when controlling the manufacturing of fuel elements because just a small amount of material has to be used for tests Of course for very low concentrations the irradiation time will increase and a stronger source of neutrons will be needed

The ^{252}Cf source is much more convenient than a reactor for this kind of measurement because the power fluctuations in a reactor will be a significant source of error to be corrected However for extremely low concentrations of uranium the reactor ought to be used

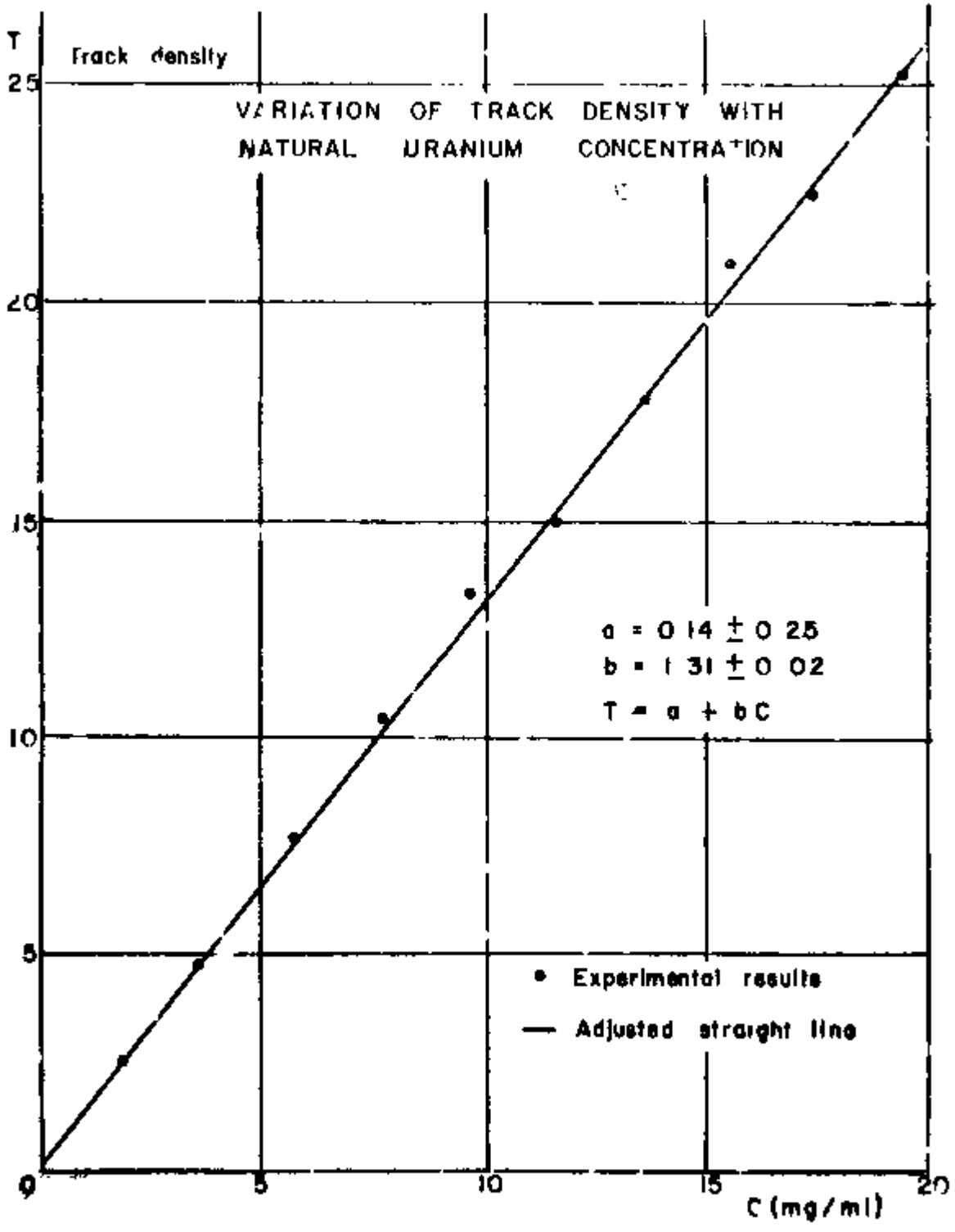

Figure 3 — Variation of track density number of tracks per unit area, (per irradiation time) with natural uranium concentration. The errors shown in the experimental points are the statistical ones

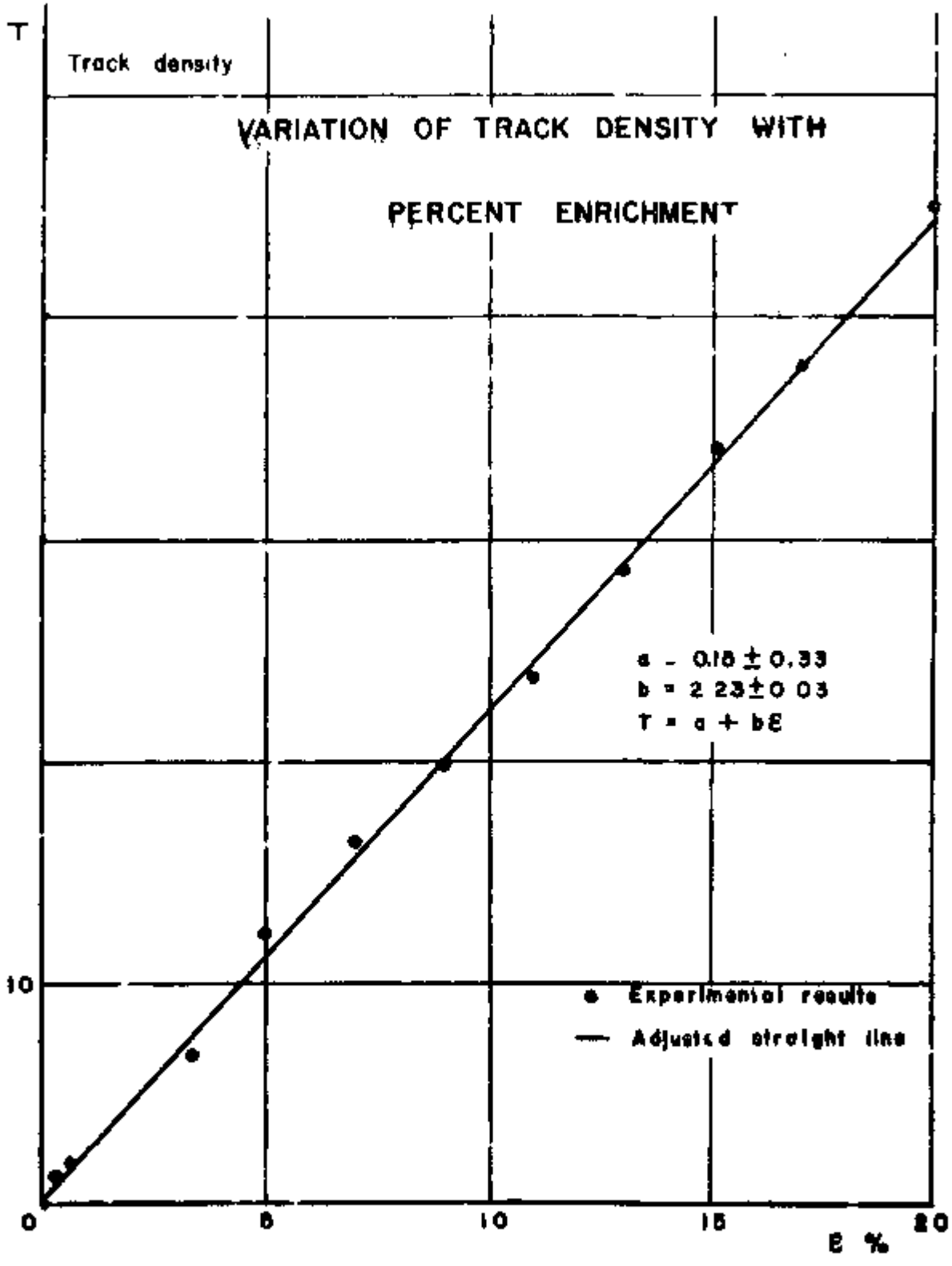

Figure 4 — Variation of track density (number of tracks per unit area per uranium solution concentration) with percent enrichment. The errors shown in the experimental points are the statistical ones plus the errors in the concentration of the solutions.

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RESUMO

O conteudo de ^{235}U em amostras de uranio enriquecido foi determinado usando se a técnica do registro de traços de fragmentos de fissão com contagem automatica Soluções de nitrato de uranilo foram irradiadas com nêutrons termalizados provenientes de uma fonte de ^{252}Cf Enriquecimentos em torno de 20% podem ser medidos com precisão de 2%

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