

## SHORT COMMUNICATION

# DETERMINATION OF URANIUM CONTENT OF TUBERS BY THE FISSION-TRACK REGISTRATION TECHNIQUE

O. Y. MAFRA and M. E. COUTO

Institutot Militar de Engenharia, Seção de Energia Nuclear, CEP-22290, Rio de Janeiro (RJ), Brasil  
and

L. P. GERALDO

Instituto de Energia Atômica (IEA), Área de Física Nuclear, CP-11049, Pinheiros, São Paulo (SP), Brasil

(Received 7 August 1980; in revised form 12 November 1980)

**Abstract**—The fission-track registration technique using the “wet method”, with Makrofol® Kg as the detector, was applied to the determination of uranium concentration in tubers. The raw samples (carrots) were ashed, dissolved and then irradiated in a thermal neutron flux. The uranium concentration was measured in samples from crops in the neighbourhood of the Cercado uranium mine in Poços de Caldas, Minas Gerais, and in carrots from the Rio de Janeiro local vegetable market. In the ashes of the Poços de Caldas carrots  $2.8 \pm 0.1 \text{ mg U kg}^{-1}$  was found; and in the Rio de Janeiro samples the concentration found was  $0.42 \pm 0.01 \text{ mg U kg}^{-1}$ .

### 1. INTRODUCTION

The determination of uranium concentration in tubers is of considerable interest for environmental control. Since these root vegetables are high mineral absorbers (Kaindl *et al.*, 1961), they can absorb the uranium from the soil to a high degree.

In this paper the fission-track registration technique was applied to the problem and the “wet method” (Iyer *et al.*, 1973, 1974) used. The great advantage of using solutions instead of the “dry method” is the homogeneous track distribution achieved, thus providing a more accurate measure of the average bulk uranium content.

The detector used was the Makrofol kg-10  $\mu\text{m}$  (Bayer) and counting was made in an automatic chamber (Cross and Tommasino, 1970). The

detectors were immersed in carrot solution and the whole assembly was irradiated in polystyrene containers.

The samples measured came from agricultural regions near the Poços de Caldas uranium mines and from the Rio de Janeiro food market.

### 2. EXPERIMENTAL

#### 2.1 Sample preparation

The samples were dried in an oven for 24 h at 100°C. The dry product was then subjected to gradual ashing, raising the furnace temperature at intervals during 16 h, to 485°C (HASL-300, 1972). The ashes thus obtained were dissolved in  $\text{HNO}_3$  1:1 (v/v). Subsequently, the residue was treated with

® Makrofol is a registered trade mark of BAYER AG of Leverkusen, W. Germany.

concentrated  $\text{HNO}_3$  and the solution evaporated to dryness. This procedure was repeated until a white residue was obtained. This residue was finally dissolved with diluted  $\text{HNO}_3$  (0.3 M) and the solution filtered (Whatman 40) to eliminate the undissolved residue.

### 2.2 Sample irradiation

For high uranium concentrations ( $\sim$  ppm), the irradiations were performed in the IEN Argonauta reactor, Rio de Janeiro ( $1.24 \times 10^9 \text{ n cm}^{-2} \text{ s}^{-1}$ ). For lower uranium concentrations ( $\sim$  ppb) the IEA R-1 reactor, São Paulo, was used ( $7.2 \times 10^{12} \text{ n cm}^{-2} \text{ s}^{-1}$ ).

Polystyrene containers of 50 ml were used for the irradiation of the solutions with the Makrofol detectors immersed in them. This arrangement was used in order to give a large contact area between the solutions and the detectors, and a better geometrical reproducibility (Geraldo *et al.*, 1979).

The thermal neutron fluence was monitored by the same fission-track registration technique. Each sample was always irradiated together with a monitor solution of known uranium content.

### 2.3 Etching of the detectors

For the detectors irradiated at the Argonauta reactor, the etching conditions were: KOH 35% ( $D_{20}^{20} = 1.336$ ) solution for 15 min at 60 °C (Bertine *et al.*, 1970). For the detectors irradiated at the IEA R-1 reactor, the integrated neutron flux was greater than  $10^{16} \text{ n cm}^{-2}$ ; therefore softer etching conditions are necessary in order to prevent the detector becoming brittle. The etching conditions in this case were: KOH 24% ( $D_{20}^{20} = 1.223$ ) solution, for 12 min at 60 °C (Geraldo *et al.*, 1979).

### 2.4 Track counting

An automatic discharge chamber was used for track counting. The working voltage was 600 V and the counting area was fixed at 2.54 cm<sup>2</sup> (Mafra *et al.*, 1977). Each detector (2.0 × 0.22 cm) provided six counting areas, thus improving the statistical accuracy. The reproducibility of the measurements was within 0.1%, and the reproducibility found for the whole technique was within 1.0%.

## 3. RESULTS AND DISCUSSION

In order to verify whether the carrot ashes were modifying agents of the irradiated aqueous solution, irradiations were performed using solutions of carrots obtained from the local (Rio de Janeiro) food market and from farms located in the neighbourhood of uranium mines, artificially contaminated with 100 ppm of uranium. These results were checked against those obtained with pure uranyl nitrate solutions with the same concentration.

In Table 1 it can be observed that the carrot ashes do not act as a modifying agent of the irradiation medium. It can also be seen that the irradiations at the Argonauta reactor give better reproducibility of results (within 1%) than those previously obtained at the IEA R-1 reactor (Geraldo *et al.*, 1979). A calibration curve (Fig. 1) using

Table 1. Verification of the influence of carrot ashes as a modifying agent in the irradiated solutions through which the fission fragments travel

Weight of carrot ashes (g)	Location	Total track number
0	Standard solution	10593
0.0	Standard solution	11508
0.5	São Beneditos Farm (Poços de Caldas)	10879
0.5	Rio de Janeiro Market	10520
1.0	São Beneditos Farm (Poços de Caldas)	10843
1.0	Rio de Janeiro Market	10861
2.0	Rio de Janeiro Market	10823
3.0	Rio de Janeiro Market	10349

Mean value  $\bar{X} = 10797$  tracks.

Standard deviation  $S = 346$ .

Standard deviation of the mean  $S_m = 122$ .

Student  $t$  factor for 5% level of significance and seven degrees of freedom, namely  $t_{7;0.05} = 2.36$ .

Reproducibility  $\frac{t \cdot S_m}{\bar{X}} = 2.7\%$ .

The uranium concentration in all solutions was 100 ppm, and the etching conditions were: 8.3 M KOH bath for 15 min at 60 °C.

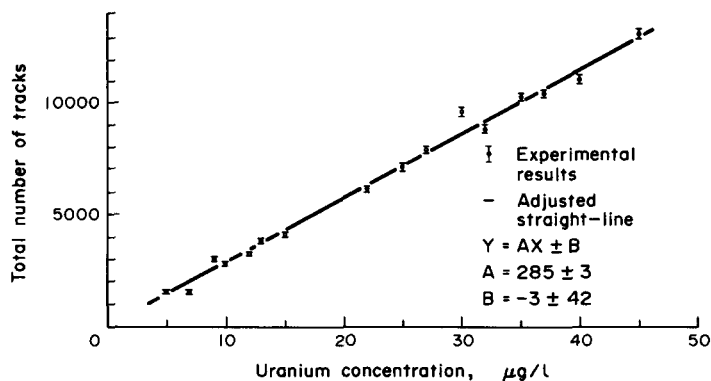


FIG. 1. Total track number (over the  $15.2\text{cm}^2$  detector area) versus uranium concentration in uranyl nitrate solutions for 12 min etching time in 24 % KOH. Thermal neutron flux  $7.2 \times 10^{12} \text{ n cm}^{-2} \text{ s}^{-1}$ .

uranyl nitrate solutions with concentrations in the range of  $5 \mu\text{g l}^{-1}$  to  $45 \mu\text{g l}^{-1}$  was constructed. The irradiation time was in the range of 15–30 min at the IEA R-1 reactor. The straight line was adjusted by the least-squares method; and with this calibration curve, uranium concentrations can be determined with a total error in the range of 1.5 % to 4.0 %.

The results obtained for carrot ashes from the local (Rio de Janeiro) market and from Poços de Caldas can be observed in Table 2.

#### 4. CONCLUSIONS

The viability of the nuclear-track registration technique for tubers was confirmed for the

Table 2. Uranium concentration in carrots from Poços de Caldas mines region and from the Rio de Janeiro area

Location	Concentration of U ( $\text{mg kg}^{-1}$ ashes)
Rio de Janeiro	
1	$0.43 \pm 0.01$
2	$0.40 \pm 0.01$
Mean value:	$0.42 \pm 0.01$
Poços de Caldas	
1	$2.72 \pm 0.04$
2	$2.87 \pm 0.05$
3	$2.75 \pm 0.05$
Mean value:	$2.78 \pm 0.06$

particular case of carrots. An extension to other tubers can easily be made, since the only parameter to be modified would be the influence of ashes in the irradiation medium, which implies the existence of absorbers or scattering elements.

As the ashes correspond on the average to 1 % of the raw product (HASL-300, 1972), it can be estimated that the uranium concentrations in the fresh carrots analysed were  $28 \mu\text{g U kg}^{-1}$  and  $4 \mu\text{g U kg}^{-1}$ , respectively, for the mines region and for Rio de Janeiro area.

These results were expected, since the uranium concentration in the water in the vicinity of the Cercado uranium mine (Poços de Caldas) is  $425 \pm 22 \mu\text{g U l}^{-1}$  (Geraldo *et al.*, 1979).

Little data can be found in the literature for the absorption factor, and this factor is strongly dependent on the kind of soil, season of the year, maturity grade and water content in minerals. According to Kovalevski (1969), this biological absorption coefficient ranges from 0.2 to 0.02, and decreases for increasing uranium concentrations.

Although the uranium concentration found in the carrots near uranium occurrences is higher than that found in the market food stuffs from other regions, it is still much lower than the limit of 150 mg U per day established by the ICRP for an individual ingestion of even 1 kg per day.

*Acknowledgements*—The authors wish to thank L. R. Pinto and E. A. Santos for their great help. The staffs of the IEN and IEA reactors are also thanked for their help in the sample irradiations.

## REFERENCES

- Bertine K. K., Cham L. H. and Turekian K. K. (1970) Uranium determination in deep sea sediments and natural waters using fission tracks. *Geochim. Cosmochim. Acta* **34**, 641–648.
- Cross W. G. and Tommasino L. (1970) A rapid reading technique for nuclear particle damage tracks in thin foils. *Rad. Effects* **5**, 86–89.
- Geraldo L. P., Cesar M. F., Mafra O. Y. and Tanaka E. M. (1979) Determination of uranium concentration in water samples by the fission track registration technique. *J. Radional. Chem.* **49**, 115–126.
- HASL-300 (1972) Procedures Manual. Health and Safety Laboratory, USAEC, pp. Sr-01-03, U-03-01.
- Iyer R. H., Sager M. L., Sampathkumar R. and Choudhuri N. K. (1973) Application of the fission track registration technique in the estimation of fissile materials:  $^{235}\text{U}$ -content in natural and depleted uranium samples and total uranium in solution. *Nucl. Instr. and Meth.* **109**, 453–459.
- Iyer R. H., Sager M. L., Sampathkumar R. and Choudhuri N. K. (1974) Fission track registration in solid state track detectors immersed in fissile—material solutions. *Nucl. Instr. and Meth.* **115**, 23–27.
- Kaindl K. and Linser H. (1961) Radiation in agricultural research and practice. IAEA, Report No. 10, pp. 25–37.
- Kovalevskii A. L. (1969) Absorption of natural radioactive elements by plants. *Chem. Abstr.* **70**, 65343d.
- Mafra O. Y., Cesar M. F., Geraldo L. P., Tanaka E. M. and Renner C. (1977) Determination of the  $^{235}\text{U}$  content in enriched samples by the fission track registration technique. *Nucl. Instr. and Meth.* **143**, 117–119.