



ORIGINAL ARTICLE

Microanalysis of Uranium by Using Solid State Nuclear Track Detector: Techniques, Problems, Prospects

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ABSTRACT

Uranium is the heaviest and radioactive trace element found in geological materials at ppm level. It is distributed non-uniformly and mainly occurs in a dispersed state. Uranium present in the earth is transferred to water, plants, food supplements and then to human beings. Intake of uranium and its decay series elements can be harmful to human health. When absorbed in the body it can constitute a far greater radiation health risk as the internal tissues are irradiated continuously with radiation till the isotopes are eliminated in faces or urine or else lose radioactivity by natural decay. Due to irradiation somatic mutation may occur which can alter control mechanism of cell division leading to cancer. Evaluation of the occurrence and distribution of this environmentally important trace element-can be important in assessing its mobilization to ecosystem. Although the permissible intake limit is 40 mg/d, low dose rates for a long time can be malignant as the uranium miners exposure data show the linear relationship between the incidences of lung cancer and accumulated exposure. Besides the radiological importance, uranium plays significant role in various cosmo-chronological and nucleo synthesis events. Evaluation of the occurrence and distribution of this environmentally important trace element-can be important in assessing its mobilization to ecosystem. In this paper microanalysis of uranium has been carried out by using solid state nuclear track detector. Techniques, problems and its prospects have also been discussed.

Key words: SSNTD, Uranium, Radioactive Decay, Environment

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INTRODUCTION

Uranium is the heaviest and radioactive trace element found in geological materials at ppm level. Although it is considered a rare element, it has higher abundance in earth's crust than other toxic elements such as K, Sb, Cd, Bi, and Hg etc. It is distributed non-uniformly and mainly occurs in a dispersed state. The heterogeneity is due to the geochemical processes which have recycled slowly the crustal material to and from the mantle. Ingenuous and metamorphic rocks have higher uranium concentration than sedimentary rocks. The earth's crust has 95 % ingenuous and 5 % sedimentary rocks. The amount of uranium in the outer 10 Km of the earth's crust is estimated to be about 1.3×10^{14} tons corresponding to the radioactivity of 1.7×10^{24} Bq. Uranium present in the earth is transferred to water, plants, food supplements and then to human beings. Intake of uranium and its decay series elements can be harmful to human health. When absorbed in the body it can constitute a far greater radiation health risk as the internal tissues are irradiated continuously with radiation till the isotopes are eliminated in faces or urine or else lose radioactivity by natural decay. Due to irradiation somatic mutation may occur

which can alter control mechanism of cell division leading to cancer [22]. Although the permissible intake limit is 40 mg/d, low dose rates for a long time can be malignant as the uranium miner's exposure data show the linear relationship between the incidences of lung cancer and accumulated exposure [21]. Besides the radiological importance, uranium plays significant role in various cosmo chronological and nucleo synthesis events. Evaluation of the occurrence and distribution of this environmentally important trace element-can be important in assessing its mobilization to ecosystem. Uranium is transferred to water by its leaching action from bed rocks [2]. It can also be introduced into water supplies by human activity in its mining and milling. Radiotoxic uranium and its salts are found in many valence states, the most common being hexavalent and tetravalent [5]. Water soluble hexavalent compounds of uranium can injure kidney preventing the normal waste product elimination resulting in renal disfunction. It can also combine with the active sites (phosphate groups), on the surface cells blocking normal metabolic processes for cell's survival [24]. The unstable tetravalent form of natural uranium is oxidised to the more toxic hexavalent form.

TECHNIQUES

Several methods such as Neutron activation analyses, radiometric method, Mass spectroscopy, Fission track method etc. are being used for trace uranium determination. Solid state nuclear track detection provides a cheap, rapid, simple and sensitive technique and has the potential capability of micro mapping at even sub-ppb level of fissionable impurities. First suggested by Price and Walker in 1963, the technique has been extensively used for the microanalyses of uranium. The relative abundance of the uranium isotopes can be assumed to be constant in natural uranium. Thus an estimation of U-235 content in the sample instead of abundant U-238 can yield the uranium concentration in it. U-235 is fissionable with thermal neutrons having a high fission cross section of 580 barns. Although fission can also be initiated by fast neutrons in U-238, the method is not suitable due to the presence of Th-232 in samples which is also fissionable with fast neutrons [6]. Also the difficulty in the availability of fast neutrons poses a problem.

Samples in which uranium concentration is to be measured and a standard material of known uranium concentration are kept in close contact with a fission track detector (various Makrofol, Lexan and Melinex-O etc.) and are irradiated with thermal neutrons. The tracks formed as a result of latent damage in the detector by the fission fragments due to (n, f) reaction on U-235 is visualised by the preferential chemical etching. By comparing the track densities in the detectors, uranium concentration in the sample can be determined. Any Th-232 in the samples contributes very little to the total damage from (n, f) reactions due to very low cross section for thermal neutrons 2.5 μb as compared to 580 b for U-235. Two methods dry method and Vet method were applied for microanalyses of uranium in a large no. of samples of different materials.

1. DRY EXTERNAL DETECTOR METHOD

(a) For Solid Phase Materials:

The external detector method was suggested by Fleischer et al, in 1875 [9] for bulk determination of trace elements. Accurately weighed sample powder of material was homogeneously mixed with methyl cellulose powder used as a binding material free from uranium in the ratio 1:2 by weight. From 200 mg of this mixture a thin pellet of about 1mm thickness and 1.3 cm in diameter was made by using a hydraulic pellet making machine applying a pressure of about 50 MPa. Pellets of standard glass of known uranium concentration [1 & 18] were also made in an exactly similar manner. Each of these pellets was sandwiched between a pair of plastic track detectors (Makrofol KG, Lexan, Melinex-o). All the pellets of the samples and the standard glass were enclosed in aluminium 'Can'

and irradiated with thermal neutrons from a reactor at BAEC, Trombay. The neutron dose used for irradiation was 15about 10 (nvt). After irradiation, the detector disks having latent fission tracks were separated from the pallets and chemically etched under appropriate etching conditions to reveal the fission tracks. The tracks were scanned using an optical microscope at a magnification of 400X. In the detectors in contact with coal and fly-ash samples, a typical distribution of tracks with clusters of tracks (in the form sunbursts) in the background of uniform track density was observed [12]. Thus the areas of uniform and non-uniform tracks were mapped out separately. In the case of clusters, the track density was obtained by measuring the actual area occupied by each cluster. Uranium concentration was calculated using the expression [13-14].

$$U_x = U_s (T_x / T_s) (I_s / I_x)$$

Where the subscripts x and s stand for the sample and standard respectively, T is the fission track density and I, the isotopic abundance ratio of U-235 and U-238 (taken as unity, assuming that the isotopic abundance ratio is the same for the samples and standard substance). For coal and fly-ash samples uranium concentrations were calculated separately for uniform and non-uniform track distribution

(b) For Liquid Phase Materials:

All samples of water etc. were slightly acidified immediately after collection by mixing a small amount (~2%) of concentrated nitric acid. This minimized the loss of uranium by absorption in the container for storing time up to 30 days. A known volume (0.06 ml) of liquid sample was placed on a clean circular disc of plastic detector using a micropipette already rinsed with double distilled water and test liquid. The water droplet on the detector discs were allowed to evaporate in still air in an oven at 70°C. The evaporated droplet will leave a thin film of non-volatile constituents present in the water sample including uranium. The left-out non-volatile constituent of the sample appeared like an almost circular patch on the detector disc. It was covered with another similar circular detector disc. Each pair of detector discs was made to be in intimate contact with the dried specimen to form a pellet. The pellets were encapsulated in cylindrical aluminum capsule. A standard glass piece of known uranium concentration serving as a thermal neutron flux dosimeter was also placed in the aluminium capsule. A blank pellet of detector disc³ was also kept in the capsule to take care of contamination due to uranium present in the distilled water and the detector itself. The capsules were irradiated with thermal neutrons with a flux of about 10^{15} nvt in the thermal column of the reactor at BARC Bombay.

The pattern of the tracks formed in the detector was almost circular with high track abundance along the rim of the droplet site and uniform in the inner region [3]. Therefore, regions of crowded and uniform tracks were napped out separately. Tracks along the rim are found to have almost constant track density and can be represented in terms of constant number of tracks per unit length (N_1) along the rim having a width δ . In the inner region tracks per unit area (N_a) were determined. A minimum of 500 tracks were counted to obtained N_1 and N_a . The total number of tracks on a detector disc can be expressed as [7 & 15].

$$N = 2 \Pi R N_1 + \Pi (R-\delta)^2 N_a$$

Where R is the radius of the droplet. The neutron flounce ' Φ ' was obtained from the irradiated standard glass. In some cases where the droplet is not exactly circular i.e. having slight ellipticity and flattening of the track pattern, straight forward variation in the eq. (2) can be made. There may arise some problems due to overcrowding of tracks and non-uniformity in their spatial distribution. When the track density is too high to count along the rim, a separate irradiation with a lower neutron flux is required. N_1 is resolvable in most of the parts but not in whole of the rim indicating that N_1 varies circumferentially. For such type of pattern either a full counting of the tracks or series of

equally spaced measurements along the rim are to be done. Also the indication of particulate concentration of uranium is found by cluster of tracks. Such type of track patterns of liquid drop residue may be due to the suspended solids contained in the liquid. Uranium concentration, C , in liquid samples is calculated from the expression [9].

$$C = N.M / V G N_A E \sigma_f \Phi I$$

Where M is the atomic weight of fission isotope (U-235); V the volume of the drop; G the geometry factor for the detection of tracks in the detector taken as unity since the non-volatile residue has a thickness much less than the range of fission fragments [4]. N_A the Avogadro's number; σ_f is the fission cross section for fissile isotope (580 barns); E the etching efficiency of the detector and I the isotopic abundance ratio of U-238 ($= 7.26 \times 10^{-3}$). For the usual combination of uranium isotopes it can be calculated that 1 mg of uranium has an activity of 24.79 Bq. Using the relation, the resulting activity of uranium in samples can be calculated.

(2) WET INTERNAL METHOD

Heterogeneity of track distribution in the track detectors has been frequently observed in the dry external detector method used for liquid samples. Also the deposited film area of evaporated deposits of solutions on the track detector is very large and therefore, the microscopic counting of all the tracks and especially in the heterogeneous region is quite difficult. In the wet method [10-11], the detector piece is immersed in the solution containing fissile material and after irradiation with thermal neutrons, homogeneous track distribution in the detector can be obtained, since the distribution of fissionable material in a solution is very uniform and directly proportional to the fissile material per unit volume. The uniformity and linearity has been found to be maintained in "the range of 8×10^{-3} g/ml to 4×10^{-2} g/ml of uranium and there is no self absorption within this range. Thus, the wet method using fission track registration technique may be a suitable method for the trace analyses of fissile material in a sample. The track density, T , recorded on the detector is given by [19]

$$T = K_{\text{wet}} n \sigma_f \Phi t$$

Where n is the number of fissile atoms per unit volume of the liquid sample, σ_f the fission cross section, Φ the neutron flux, t the time of irradiation and K_{wet} , the constant of proportionality which gives a measure of the track wet registration efficiency in the sample and has the dimensions of length. If C is the uranium concentration in the solution and X the percentage of U-235 ($\sim 0.71\%$), above eq. can be written as

$$T = K_{\text{wet}} C N_A X \sigma_f \Phi t / A$$

Where N_A is the Avogadro's number and A is the atomic weight of the uranium. The value of K_{wet} depends on the track registration characteristics (critical angle, threshold energy for track formation etc.) of the detector itself as well as on the range and energy loss characteristics of the fission fragments in the aqueous medium through which it travels. For standard and sample eq. (5) can be written as

$$T_S = K_{\text{wet}} C_S N_A X \sigma_f \Phi t / A \quad (6)$$

$$T_X = K_{\text{wet}} C_X N_A X \sigma_f \Phi t / A \quad (7)$$

Where C_S and C_X are the concentration (weight/volume) of total uranium. From eqs. (6&7) one gets,

$$C_X = (T_X/T_S) C_S \quad (8)$$

The method was used for the uranium analyses in tubers, the details of which are given elsewhere [16]. The calibration lines obtained from least square fit of the track density Vs uranium concentrations of 20 standard solutions (5 to $100 \mu\text{g/l}$) for Laxan and Makrofol-KG show clearly that the track density is directly proportional to the uranium

concentration of the solutions. Also there is no self absorption as the linearly is maintained [17].

RESULTS AND DISCUSSION

Investigation were initiated to study uranium concentration in various materials of interest. These include microanalyses of uranium in soil, plants, building materials, water samples tubers, tobacco, tea leaves, coal, dry milk, drugs etc. collected from various sources and regions of the country. A large number of water samples (drinking, domestic, ground and surface etc.), covering a wide area of several states in India were investigated. The results indicate that the uranium Concentration is correlated with the uranium content in the rocks, soil, sand and minerals with which water comes in contact. The most likely explanation for certain specific large values may be the uranium deposited as thin surface layers in bed rock fissures. When they are dissolved, the deposits of this type may release high concentrations of uranium into the water. Uranium content widely depends on the type and site of the surface. In surface waters, the use of phosphate fertilizers in the soil in its neighborhood may have led to increased uranium concentration in the rivers. In spring and lake water it is derived by its leaching action while passing through the rocks. The natural sources may contaminate both ground and surface water with occurrence being more common in ground water sources. Source wise uranium is found to be more abundant in sea water followed by well, river and tap water. Use of earthen materials like coal industry and power generation can enhance the levels of uranium in the natural water bodies in the adjoining areas. In building materials, yellow brick is found to have lowest level of uranium while cement has the highest no. noticeable area-wise distribution of uranium has been obtained in the case of soil samples. Low level of uranium in soils from Antarctica region may be attributed to absence of human intervention with the lithosphere in the region. Here the redistribution of the radionuclides takes place only by the natural geochemical and geophysical processes. Industrial activity can enhance the level of uranium in soil in the adjoining areas of activity as evident from the measurements at Fertilizer and chemicals, Travancore, Alwaye, Kerala and the coal fired thermal power project, Kasimpur (U.P.). Indian coal has higher levels of uranium as compared with coal samples from many other countries. Microscopically the distribution of uranium is not uniform in coal and fly-ash. Coal combustion enhances the concentrations of uranium in fly-ash. Enhancement is 3 times in uniform distribution but 1.5 times in non-uniform distribution. Due to enhanced level of uranium in fly-ash, the use of fly-ash as building material and its spread out may modify the radioactivity in the environment. Each plant has its own rate of assimilation of uranium from the growing medium and depends on elemental contents of soil and water, thus uranium concentration in plants should vary from plant to plant and place to place.

In general, cigarettes were found to have higher uranium content than beedies by a factor of about 1.4 on the average. Pan masala shows the highest level of uranium. Pan masala and chewing tobacco remain in contact with the tongue, inner cheek palate and gum for long time. Therefore, the presence of uranium in these materials may be of considerable importance similar to occurrence of Po-210 in cigarette smoke [20] which is found to play an important role in the etiology of lung cancer [23]. Fission track registration technique is highly sensitive, reliable and has the capability for micro-mapping even at sub ppb levels of fissionable elements. In dry fission track method, the track detector is in contact giving rise to a 2 π or nearly 2 geometry. In the pellet in contact with the detector in 2 geometry, the registration efficiency is very high. The Preparation of uniform thin pellet requires the expertise and efforts and slight non-uniformity in it will give rise to non-uniform distribution of tracks in the detector. For accurate evaluation of data, the entire area of the detector should be scanned for obtaining the total number of tracks. Uniform distribution of tracks enables the evaluation of track density by scanning a small representative area. The important advantage of the wet method is the uniform distribution of tracks observed in the detector. However, the care should be taken to

avoid undisclosed particles suspended in the solution so that cluster of tracks may not appear. The method can be used for the measurement in the sample having very low uranium concentration.

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REFERENCES

1. Azam A. and Prasad R. (1989b): J. Radioanal. and Nucl. Chem.133199.
2. Bansal V. (1989a): Ph.D. Thesis, Aligarh Muslim University, Aligarh.
3. Bansal V. (1989a): Ph.D. Thesis, Aligarh Muslim University, Aligarh.
4. Bansal V., Rawat A., Jojo P.J. and Prasad R (1992): Health Phys.62, 257.
5. Cothorn C.R.and Lappenbusch W.L. (1983): Health Phys. 45, 89.
6. Durrani S.A. and Bull R.K. (1987): Solid State Nuclear Track Detection, Principles, Methods and Applications, Pergamon Press, Oxford.
7. Fleischer R.L. and Delany A.C. (1976): GEC Report No. 76 CRDO 11, New York, pp. 124.
8. Fleischer R.L. and Lovett D.L. (1968): Geochim. Cosmo chim Acta 32, 1126.
9. Fleischer R.L., Price P.B. and Walker R.M. (1975): Nuclear Tracks in Solids and Applications, University of California Press, Berkeley pp. 490.
10. Iyer R.H., Sampatkumar R. and Chaudhari R.K. (1974): Nucl. Instr. and Meth. 115, 23.
11. Iyer R.H., Sampatkunmar R. and Chaudhari R.K. (1973): Nucl. Instr. and Meth. 109, 453.
12. Jojo P.J. and Prasad R. (1993a): Proc. of VIIth Int. Conference. on Particle Tracks in Solids, Jodhpur (India) 93.
13. Jojo P.J., Rawat A. and Prasad R. (1994b): Nucl. Geophys. 8, 55.
14. Jojo P.J., Rawat A., Kumar A. and Prasad R. (1993b): Nuc1.Geophys. 7,445.
15. Jojo P.J., Rawat A., Kumar A. and Prasad R. (1994a): J.Radioanal. Nucl.Chem. 178,245.
16. Kumar A. (1994a): Ph.D. Thesis, Aligarh Musiim University, Aligarh.
17. Kumar A., Jojo P.J., Rawat A. and Prasad R (1994b): J.Trace and Microprobe Technique 11, No.4.
18. Kumar S. and Srivastava D.S. (1984): Ind. J. Pure & Appl. Phys. 22, 15.
19. Kumar S. and Srivastava D.S. (1994): Ind. J. Pure & Appl. Phys. 22, 15.
20. Laiet N., Sharma P.K., Nagpaul K.K., Behera D. and Malik S.K. (1986): Nucl. Tracks 12,781.
21. Morgan K.Z. (1973): CRC Hand book of Chemistry and Physics, CRC Press Cleveland (U.K).
22. Power C.B. (1981): Cell Biology, 2nd Edn. Himalaya Publishing House, India. Price P.B. and Walker R.M. (1963); Appl. Phys. Lett 2, 23.
23. Sheikh G.N., Subba Ramu S. Sadasivan S. and Mishra U.C. (1988): Lung India 6.199.
24. Steere M. V. (1970): Hand book of laboratory safety The Chemical Rubber Company, U.S.A.