CHAPTER - III

EXPERIMENTAL PROCEDURE AND WORKING FORMULA

INTRODUCTION :

The "Particle track analysis" method using SSNTDS, described in the previous chapter has been used for the estimation of uranium in plants. The various stages of the experimental procedure are briefly described below.

3.1. COLLECTION OF SAMPLES :

Thirty eight common plants belonging to cereals, pulses, underground vegetables, betel leaves, its chewable ingredients and samples of lime are collected from Garal, Dalibari, Jalukbari and Maligaon from Kamrup district of Assam ; Nongpoh of Meghalaya. A brief description of the different categories of selected plant samples are given in the next chapter.

The various parts taken for the study are root, stem, leaf, grain, nut and fruit. Twenty soil samples from the respective plant habitat are also taken for uranium estimation.

3.2. PREPARATION OF SAMPLES :

The observation of tracks for uranium estimation involves preparation of sample pellets to be sent for irradiation with thermal neutrons. It is described below.

3.2.1. PREPARATION OF THE PELLETS :

Different parts, taken as samples are cleaned and washed with distilled water and dried in an oven at a temperature of $150^{o}C$ for 24 hours. The dried samples are then crushed and

fused in contamination free silica crucibles in a furnace at a temperature of about 700°C for 2 hours. The powdered ash is then sieved through a 100 mesh (1.5×10^{-2}cm. aperture) sieve to obtain a homogenous powder. For lime, dried limestones are crushed and then sieved to get the fine powder. 50 mg of this powder is mixed thoroughly with 100 mg of methyl cellulose which acts as a binder. This mixture is then pressed into flat pellets of fixed sizes (about 1.3 cm diameter and 1 mm thick) with smooth cohesive surfaces by a Hydraulic Rock Crusher (Make CUTROCK LONDON). The pellet making action of this machine is similar to the gig used by Fisher[1] and the hand pressing machine used by Goswami et al.[2]

It may be mentioned that the uranium content of methyl cellulose is low enough and hence it gives essentially Zero tracks under suitable irradiation condition. Also any contamination can be recognised in the detectors - the tracks associated with uranium within the samples vanish abruptly at the boundary.

3.2.2. DETECTOR USED :

For detection of fission fragment tracks from fission of U-235 lexan polycarbonate is used. Detector pieces are cut into discs of same sizes as the pellets and washed with distilled water and dried in a dust-free atmosphere.

3.3. IRRADIATION PACKING :

The pellets of samples are sandwitched between two inscribed detector discs. A series of sandwitched samples are then

filled into specially made aluminium cans. A pair of standard reference material sandwitched between detectors are also inserted in the can at different places. The caps of the cans are tightly screwed to ensure firm contact between the pellets and the detectors.[2] The standard reference material used is standard glass of known uranium concentration.

3.4. IRRADIATION OF SAMPLES :

The can, thus packed, are sent for thermal neutron irradiation in the CIRUS Reactor, Bhaba Atomic Research Centre, Bombay to a dose of about 10^{15} nvt. so that an optimum track density, observable under an optical microscope, is obtained.

3.5. ETCHING OF IRRADIATED DETECTORS :

After irradiation, the detector pieces are taken out. The lexan pieces are etched with 6N NaoH solution at a temperature of $(70 \pm 0.1)^{\circ}C$ for 20 minutes with constant stirring so that the holes may be clearly observed in the microscope.[2,3,4] The constant temperature bath used for etching the detectors is provided with a thermostat which can maintain the constancy of temperature with an accuracy of $\pm 0.1^{\circ}C$.

3.6. OBSERVATION OF TRACK DENSITIES :

The fission tracks in lexan detectors are counted by an optical microscope (olympus) with a magnification of 640 x. When the track density is required to be determined, tracks are counted in large number of fields of view to get adequate statistics. Tracks in the periphery are left to avoid any possible

contamination due to handling of samples during irradiation packing.[1]

3.7. DETERMINATION OF URANIUM CONCENTRATION :

Natural uranium consists of its two main isotopes U-238 and U-235 in a ratio of about 140 : 1, so that the knowledge of either of them is sufficient to know the uranium content of a material. The fraction U-235 can be determined by subjecting the sample under study to fission, making use of the fact that thermal neutrons can cause fission of only U-235 so when the samples are irradiated with thermal neutrons, the U-235 isotopes present in it, will undergo fission giving rise to charged fission fragments, which will escape from the sample and impinge on the detector in contact with it. As a result, the fission fragments will leave radiation damage tracks in the detector, which, on etching, will show the nuclear tracks. The number of these tracks is directly proportional to the neutron flux and uranium content of the sample. Hence the knowledge of track density (with the same neutron flux) gives a means for determining uranium concentration in the sample.[2,3]

Fission fragments, produced by fission of U-235 are recorded in lexan, which has the capacity of discriminating against low energy charged particles and light weight recoil nuclei. Contribution of fission fragments due to possible thorium concentration in a sample is insignificant, if it is irradiated by thermal neutrons. Because the cross section of fission of thorium for thermal neutrons is extremely low ($\sim$40b)

as compared to the corresponding cross-section of 582 barns for U-235. Thermal neutrons cross-section for U-238 is also less than 0.5 mb and hence the possible contribution of fission fragments from U-238 is also insignificant.[5,6]

3.8. DERIVATION OF THE WORKING FORMULA :

The track density ρ of etched fission tracks recorded on a detector is given by[7]

$$\rho = F.N.P.g \qquad (3.1)$$

where F = the fraction of uranium atoms undergoing fission.

N = the number of uranium atoms per unit volume of the host material.

P = the probability of a fission fragment originating within one track range, R, of the surface and crossing it to leave an etchable track. R is the range of the particle over which its rate of primary ionisation is above the critical rate for the recording medium (Fig. 2.1). As noted in the figure, this is essentially the entire range for a fission fragment.

g = a geometrical factor which depends on whether tracks cross the surface from one or both the sides. For uranium fission, two fragments are produced in each event. Only one of them can produce tracks in an external detector. Hence in this case $g = 1/2$.

For fission induced in U-235 by a dose n_1 of thermal neutrons, the factor F becomes

$$F = n_1 \sigma I \qquad (3.2)$$

where n_1 = flux (ϕ) x irradiation time (t)

σ = the fission cross-section of U-235 to thermal neutrons (Cm^2)

and I = the isotopic abundance of U-235 compared to that of U-238.

The value of N is given by

$$N = \frac{N_o dc}{A} \qquad (3.3)$$

where N_o = Avogadro's number

d = the density of the host phase ($gm\ cm^{-3}$)

C = weight concentration of uranium in the host phase (gm/gm)

A = the atomic weight of uranium (gm.)

The factor P may be found by intergrating from K = 0 to K = R/2 where R is the combined range of the charged particles, and we get

$$P = \frac{R}{2} \qquad (3.4)$$

combining equations 3.1 to 3.4, we can write down the density of induced tracks ρ_i in the plastic detector as

$$\rho_i = \frac{n_1 \sigma I N_o dCR}{4\ A} \qquad (3.5)$$

The concentration of uranium in the sample can thus be expressed as

$$C_W = \frac{4\ \rho_i\ A}{n_1 \sigma I N_o dR}\ gm/gm \qquad (3.6)$$

In order to estimate the uranium concentration, induced fission track density, ρ_i of the sample is compared with that (ρ_s) of the standard glass of known uranium concentration.

$$C_w(\text{standard}) = \frac{4\ \rho_s A}{n_2 \sigma I N_o d R}\ \text{gm/gm} \qquad (3.7)$$

comparing (3.6) and (3.7), we get

$$C_w(\text{unknown}) = \frac{C_w(\text{standard}) \times \rho_i n_2}{\rho_s \times n_1} \qquad (3.8)$$

As the irradiation is with the same dose of thermal neutrons in both the cases,

$$n_1 = n_2 = n$$

Hence equation (3.8) reduces to

$$C_w(\text{unknown}) = \frac{C_w(\text{Known}) \times \rho_i}{\rho_s}\ \text{gm/gm} \qquad (3.9)$$

This expression assumes that the value of the product d x R is the same for the unknown and standard samples. Further, the relative abundance of U-235 in both the samples (i.e. standard and experimental) are considered to be same. The isotopic abundance of U-235 compared to that of U-238 is constant in nature (0.72% is the accepted value). Except in a single highly unusual case of deviation from this value (0.44%), no variation has been observed. Hence the results do not threaten the assumption of constant isotopic abundance.[7]

3.9. EXPERIMENTAL ERROR :

The detector and the sample may be contaminated during handling or processing. In the absence of external contamination of the given sample, contaminant tracks may be produced due to contaminations (a) on the surface of the detectors or (b) within the detector itself (rarely).

In the PTA for uranium, tracks due to (a) can be easily confused with true tracks. This gives rise to clusters or sunbursts of tracks, which may also be formed due to the presence of uraniferous grains in the sample. On the peripheries of the detectors it is most likely that the contamination takes place, right from the processing to packing. The tracks due to (b) are longer ones and can easily be recognised.

Inhomogeneities in thermal neutron flux yields different track densities in different samples with the same concentration of isotopes of interest. Since the thermal neutron flux in the reactor may be inhomogeneous (upto 10% per $cm)^3$ the sample must be irradiated in a small capsule (3.5 cm in the present case) over which the flux is almost uniform. Also more than one standard sample, located at different places in the sample assembly, should be used and the average track density, thus calculated, is used as standard track density (ρ_s)

Direct interference may occur due to possible presence of Pu-239, Th-232 in the samples, producing fission tracks. These tracks are shown to be negligibly small in number.

Uncertainity of the concentrations of uranium in the standard materials may also contribute to experimental error.

To avoid variability in track counting, the relative track densities, rather than absolute numbers are counted so that errors in track recognition criteria are cancelled out.[8,9]

3.10. STATISTICAL ERROR :

The PTA is based on the counting of discrete events (tracks), the density of which may vary from unusually low values to high values ($10^7/Cm^2$). While scanning, large counting rates, though time consuming, have advantage as problems of statistical fluctuations and background becomes serious. Since track events are emerging basically out of nuclear reactions, which are random in nature, a series of repeated counts taken under most stable conditions, will fall on a Gaussian distribution curve based on N, the average number of counts alone. The standard deviation S is given approximately by $\sqrt{n}$ and is referred to as the standard counting error.

Due to individual criteria for the acceptance or recognition of tracks, different laboratories may differ by as much as 20% in track counting. This can be reduced to ±3% in a much laborious and expensive technique suggested by Fisher. Therefore, one should count the relative track densities rather than absolute count.[10]

The overall errors are expected to lie within ± 10% (3% as statistical error and remaining ± 7% as the part formed by other errors). Proper identification and discrimination of background noise, minimisation of contamination and other factors affecting the accuracy of the method lead to minimisation of these errors.[11]

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