CHAPTER III

# EXPERIMENTAL PROCEDURE AND WORKING FORMULAE

## INTRODUCTION:

The "particle track analysis" method using SSNTD s, described in the previous chapter has been used for the estimation of uranium and boron in plants. The various stages for experimental procedure of the work are briefly described in this chapter.

## 3.1. COLLECTION OF SAMPLES:

Twenty common medicinal plants[1-5] are collected from and around Government Ayurvedic College Campus at Jalukbari, Assam. A brief general description of these plants are given in the next chapter.

The various parts taken for the study are root, bark, leaves and in some cases, flowers, fruits and seeds. Latex, liquid exudus from the plants, is also collected in some cases. Three soil samples from different sites from the area are also taken for uranium and boron estimations.

## 3.2. PREPARATION OF SAMPLES:

The observation of tracks for uranium and boron estimations involves preparation of sample pellets which can be sent

for irradiation with thermal neutrons. Samples of two types are to be prepared. Latex is in the liquid phase, and all other samples are in the solid phase.

3.2.1. SOLID PHASE SAMPLES:

Different parts, taken as samples are cleaned and washed with distilled water and dried in an oven at a temperature of 150°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 ($1.5 \times 10^{-2}$cm. aperature) sieve to obtain a homogeneous powder. 50 mg of this ash 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 diamter 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[6] and the hand pressing machine used by Goswami et al.[7]

It may be mentioned that the uranium content of methyl cellulose is low enough that 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 venish abruptly at the boundary. The methyl cellulose shell surrounding the sample will give rise to tracks only if contamination-uranium has been introduced.[6]

3.2.2. LIQUID PHASE SAMPLES:

With latex, the samples are prepared as in the case of water samples.[8] A known volume of the latex, in the form of a (or few) drop is taken on a detector piece, and allowed to evaporate, leaving a thin film of the substance.

3.2.3. DETECTORS USED:

For Detection of fission fragment tracks from fission of $^{235}U$, lexan polycarbonate is used and for detection of alpha particles from $(n, \alpha)$ reaction of $^{10}B$ Dia-cell cellulose nitrate 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:

Packings are done for both uranium and boron estimations separately. The pellets of samples are sandwiched between two inscribed detector discs. A series of sandwiched samples are then filled into specially made aluminium cans. Two pairs of standard reference materials sandwiched between detectors are also inserted in each can at different places. The cap of the cans are tightly screwed to ensure firm contact between the pellets and the detectors.[7]

For uranium estimation, the standard reference material used is standard glass of known uranium concentration and for boron estimation the same used is orchard leaves (National Bureau of Standards U.S.A. Standard Reference Material 1571 of known boron concentration).

3.4. IRRADIATION OF SAMPLES:

The cans, thus packed, are sent for thermal neutron irradiation in the CIRUS Reactor, Bhabha 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 $(70\pm0.1)^{\circ}C$ for 20 minutes and the CN pieces with 2.5 N NaOH solution at $(55\pm0.1)^{\circ}C$ for 8 minutes with constant stirring so that the holes may be clearly observed in microscope.[7,9,10] 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 $\pm0.1^{\circ}C$.

3.6. OBSERVATION OF TRACK DENSITIES:

The fission tracks in lexan detectors and alpha particle tracks in CN detectors are counted by an optical microscope (OLYMPUS) with a magnification of 640x. When the track density is required to be determined, tracks are counted in a large number of fields of view to get adequate statistics. When the total track count is required, the whole detector is scanned. In both the cases, tracks in the periphery are left to avoid any possible contamination due to handling of samples during irradiation pecking.[6]

## 3.7. DETERMINATION OF CONCENTRATIONS:

### 3.7.1. URANIUM:

Natural uranium consists of its two main isotopes $^{238}U$ and $^{235}U$ 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 $^{235}U$ can be determined by subjecting the sample under study to fission, making use of the fact that thermal neutrons can cause fission of only $^{235}U$, so when the samples are irradiated with thermal neutrons, the $^{235}U$ isotope present in it, will undergo fission giving rise to charged fission fragments, which will escape 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.[7,9]

Fission fragments, produced by fission of $^{235}U$ are recorded in lexan, which has the capability 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$40 b)

as compared to the corresponding cross-section of 582 barns for $^{235}U$ . Thermal neutron cross-section for $^{238}U$ is also less than 0.5 mb and hence the possible contribution of fission fragments from $^{238}U$ is also insignificant.[11,12]

3.7.2. BORON:

Boron, present as trace-element in the sample, can also be estimated by using PTA.[13-14] Boron has its two isotopes $^{10}B$ and $^{11}B$ in a ratio of about 20:1 in nature. Of them, $^{10}B$ has a very large capacity of absorbing thermal neutrons. So when a sample is irradiated with thermal neutrons, $^{10}B$, present in it, upon capturing one of them, will undergo the reaction $^{10}B + {}^{1}n \rightarrow {}^{7}Li + {}^{4}He$ having a cross-section of 3838 barns.

The emerging alpha particles will penetrate into the CN detector, placed in contact with the sample, and produce radiation damage tracks, which can be revealed by chemical etching. The density of these tracks is proportional to the neutron flux and boron content of the sample and thus offers a means for determination of boron concentration in the sample.[7,9]

The above reaction is exothermic with an energy release of about 2.78 MeV and alpha particles of energy 1.78 MeV(4%) and 1.41 MeV (96%)[12].

Significant interference comes from the reaction $Li^{6}(n,\alpha)H^{3}$, since it is one of the few thermal neutron induced reactions

of a light element with a relatively large nuclear cross-section (942 barns) that produces an exothermic triton (energy 2.73 MeV, charge +ve, mass ~3 amu). It is a very penetrating particle compared to alpha particles of the same energy. Considering the relative sensitivities of various elements, those can undergo a nuclear reaction with thermal neutrons, at the same concentration Seitz et al (1973) estimated the contribution of these elements to the track density. It is shown that boron is responsible for 98% of the tracks, lithium for 1.5% and oxygen for 0.03%. Again Pilione et al (1981) have shown that alpha tracks due to boron are etched to an observable size before the triton tracks due to lithium, make a contribution to the track density.[15-17]

## 3.8. DERIVATION OF THE WORKING FORMULAE:

### 3.8.1. URANIUM ESTIMATION:

The track density $\rho$ of etched fission tracks recorded on a detector is given by[18]

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

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, $g = \frac{1}{2}$.

For fission induced in $^{235}U$ by a dose $n_1$ of thermal neutrons, the factor F becomes

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

Where $n_1$ = flux($\phi$) X irradiation time (t)

$\sigma$ = the fission cross-section of $^{235}U$ to thermal neutrons ($cm^2$)

and I = the isotopic abundance of $^{235}U$ compared to that of $^{238}U$.

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 integrating from K = 0 to K = R/2 where R is the combined range of the charged particles,

and we get

$$P = \frac{R}{2} \tag{3.4}$$

Combining equations 3.1 to 3.4, we can write down the density of induced tracks $\rho_i$ in the plastic detector as

$$\rho_i = \frac{n_1 \sigma I N_0 d C R}{4A} \tag{3.5}$$

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

$$C_w = \frac{4 \rho_i A}{n_1 \sigma I N_0 d R} \text{ gm/gm} \tag{3.6}$$

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

$$C_w(\text{Standard}) = \frac{4 \rho_s A}{n_2 \sigma I N_0 d R} \text{ gm/gm} \tag{3.7}$$

Comparing (3.6) and (3.7), we get

$$C_w(\text{Unknown}) = \frac{C_w(\text{standard}) \times \rho_i n_2}{\rho_s \times n_1} \text{ gm/gm} \tag{3.8}$$

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

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

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 $^{235}U$ in both the samples (i.e. standard and experimental) are considered to be same. The isotopic abundance of $^{235}U$ compared to that of $^{238}U$ 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.[18]

3.8.2. BORON ESTIMATION:

The number of $\alpha$ particles passing from the emitters into the external detector is given by the relation,[13]

$$\rho = \tfrac{1}{2} C \sigma \phi t \bar{R} \cos^2\theta \qquad (3.10)$$

Where $\rho$ = observed track density ($cm^{-2}$)

C = concentration of the $\alpha$ emitting element($cm^{-2}$)

$\sigma$ = thermal neutron cross-section ($cm^2$)

$\phi$ = thermal neutron flux ($cm^{-2}sec^{-1}$)

t = exposure time (sec.)

$\bar{R}$ = average range of fragments (cm )

$\cos^2\theta$ = etching efficiency factor

where $\theta$ is the cone half angle at the tip of the etched track for CN, $\cos^2\theta \simeq 1$

If the tracks are produced by only alpha particles,

$$\rho_{obs.} = \rho_\alpha = \tfrac{1}{4} C \sigma \phi t \bar{R}_\alpha \qquad (3.11)$$

assuming $\cos^2\theta = 1$

In order to estimate the boron concentration, induced $\alpha$ particle track density, $\rho_x$ of the sample is compared (as in the case of uranium) with that of simultaneously irradiated standard leaves of known boron concentration. As in the previous case, $\phi t$, the thermal neutron dose is the same in both the cases and we get

$$C_w(\text{unknown}) = \frac{C_w(\text{known}) \times \rho_x}{\rho_s} \quad \text{gm/gm} \qquad (3.12)$$

where sufixes $x$ and s refer to unknown and standard respectively.

Equations (3.9) and (3.12), both of which are same, have been adopted as the working formula for uranium and boron estimations in the present work.

3.8.3. LIQUID PHASE MATERIALS:

When the sample, covered with detector pieces, is irradiated by thermal neutrons to induce (n,f) reactions of $^{235}U$, the total number of tracks (T) produced in the detector will depend on, concentration of the element ($C_w$),

Atomic weight of the element (M),

Volume of liquid taken (V),

Geometry factor for detection of tracks in the plastic(G),

Avogadro's number (N),

Fission cross-section of the $U^{235}$ with thermal neutrons, $\sigma$

The etching efficiency fraction $E = \left[1 - \left(\frac{V_g}{V_t}\right)^2\right]$

and Thermal neutron dose ( $\phi$ ).

That is

$$T = \frac{C_w V N}{M} \phi G E \sigma \frac{I^{235}}{I^{238}} \qquad (3.13)$$

Where I refers to isotopic abundance of the two isotopes of uranium.

Thus, the concentration $C_w$, in units of weight per volume of the liquid, of uranium is given by

$$C_w = \frac{TM}{VGN\sigma E\phi} \times \frac{I^{238}}{I^{235}} \qquad (3.14)$$

Here, the thickness of the film being much less than the fission fragments, both of them are recorded as two damages. For lexan, $V_g < V_b$ , so $E \simeq I^{19}$ , substituting the value M = 238.03, $\sigma I = 4.2 \times 10^{-24}$, N = $6.02 \times 10^{23}$, G = 1 in (3.14), we get

$$C_w = \frac{94.1\ T}{V} \text{ gm/c.c.} \qquad (3.15)$$

$\phi$ is calculated with the standard glass dosimetry by using the relation

$$\phi = K\rho \qquad (3.16)$$

where K is a constant for the standard material and $\rho$ , the induced track density in the standard material.

3.9. ERRORS OF PTA:

3.9.1. 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 processing to packing. The tracks due to (b) are longer ones and can easily be recognised.

In the PTA for boron, alpha tracks are quite distinguishable from the fission tracks of uranium, present as contaminant. However, various factors may give rise to spurious back-ground tracks, which are not distinguishable (sec. 3.7.2.).

In-homogeneities 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)[9] the samples 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 ( $\rho_s$ ).

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

Uncertainity of the concentrations of uranium and boron 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.[16,20]

3.9.2.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 back-ground become 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 $\pm 3\%$ in a much laborious and expensive technique suggested by Fisher. Therefore one should count the relative track densities rather than absolute count.[21]

The overall errors are expected to lie within $\pm$10% (3% as statistical error and remaining $\pm$7% as the part formed by other errors). Proper identification and discrimination of background noise, minimisation of contamination and other factors effecting the accuracy of the method lead to minimisation of these errors.[22]

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