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Application of SSNTDs for microanalysis, prospecting and earthquake prediction

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Introduction

Solid State Nuclear Tracks Detectors (SSNTDs) have been exploited for microanalysis due to their unique capabilities for measuring the concentration and spatial distribution of various elements. This field was opened with the measurement of uranium in minerals via the detection of fission fragments in samples irradiated with thermal neutrons (Price and Walker, 1963). Element mapping or "micromapping" is one of the most unusual and important features of track detectors. In addition to fission fragments, lighter fragments such as alpha particles and protons have been increasingly used in microanalysis. "Lexan prints" (Kleeman and Lovering, 1967) and "alpha-autoradiographs" (Coppens, 1977) give information about U/Th concentrations with or without neutron irradiation.

Of late, fission track dating has been exploited for prospecting of oil, natural gas and geothermal sources of energy using apatite and volcanic glasses (obsidians) as geological thermometers. Radon monitoring using Plastic track recorders has opened up new visitas in the field of U/Th exploration and earthquake prediction (Fleischer, 1981).

Uranium/thorium determinations

(a) Internal detector method :

This method is applicable where the substance can itself register tracks. After exposure to specified thermal neutron dose from a reactor, the sample is either cleaved or polished to expose an internal surface. After suitable chemical etching, the tracks are counted under a microscope. Surface track density is given by (Fleischer *et al.*, 1975).

$$T_i = N_u \sigma I \phi R_o n \qquad ...(1)$$

Where Nu is the number of U atoms/cm^3 in the sample, σ, the fission cross-section, I, isotopic ratio, θ is thermal neutron flux determined by using a standard glass dosimeter, Ro, is the mean range of fission fragment tracks and n, is the etching efficiency of the detector. The concentration of uranium in the sample is expressed by :

$$C_u = \frac{N_u}{N} = \frac{2T_i}{\sigma I \phi R_{on}} \qquad ...(2)$$

In practice, it is possible to detect U concentrations in materials as low as 10^{-15} atom fraction provided high thermel neutron doses ($\sim 10^{20}$n/cm^2) are available from the reactor. This is considered to be the most sensitive technique for U microanalysis.

(b) External director method :

It is conveniently used for those samples which fail to register tracks. The detector is placed against the polished surface of the mineral and irradiated. In most of the cases, homogenised fission track technique (Fisher, 1970) is used where pellets covered with external detector are irradiated alongwith a pellet of standard

glass dosimeter. The U concentration expressed in weight fraction in the unknown sample is given by :

$$C_X\ (U) = \frac{I_s\ T_X\ R_X}{I_X\ T_s\ R_X}\ C_s\ (U) \ldots (3)$$

Where the subscripts x and s refer to the unknown and the standard, I is isotopic ratio and T is the surface density of induced fission tracks and R is the effective range of fission fragment tracks.

External detectors generally used are muscovite mica, quartz and lexan plastics. The detector must be thin with a uranium concentration much lower than that of the standard glass or the sample for which U is to be determined. Whatever the detector, it is mandatory that all preprations be made in a dust-free environment.

Uranium Mapping : Thin plastic film (Makrofol polycarbonate, 8 cm thick) is pressed against the surface of interest by thermal pressure casting step and the assembly is irradiated in the thermal column of a reactor. The detector film is etched and studied under a microscope. The fission map or 'Lexan print' reveals the distribution of uranium in various mineral phases of the rock specimen. Although B and Li play a role in giving prints in terrestrial rocks, good prints may also be obtained from fission tracks alone.

(c) **Thorium mapping** :

In natural samples, Thorium is more abundant than Uranium by a factor of nearly four. It is necessary to perform two irradiations, one with thermal neutrons to determine the concentration of 235 U, the other with high energy particles that fission both Uranium and Thorium. Substraction method is used for Th estimation in the samples.

In the double beam irradiation technique an unknown and a standard sample of known U concentration are first irradiate in a flux of thermal neutrons and next irradiated with 30 MeV alpha-particles in a cyclotron (Hair et al., 1971). Th/U ratio of an unknown sample is given by the expression :

$$\frac{(C_{Th})_X}{(C_U)_X} = \frac{T_{X\,(\alpha)}}{T_{s\,(\alpha)}} \times \frac{T_{s\,(n)}}{T_{X\,(n)}} \text{-I} \quad \ldots (4)$$

Where the symbols have their usual meanings. Thorium can also be evaluated by irradiating the unknown sample in a fast neutron reactor, by wrapping the specimen in a Cd foil. The accuracy of the method is comparable with (n, r) analysis.

Alpha-Autoradiography : Determination of U/Th ratios in geological samples is possible by using alpha autoradiography. Coppens (1977) used nuclear emulsions for recording of alpha tracks and used the expression :

$$10^6 N = (9.20\ U + 2.80\ Th)\ k \quad \ldots\ldots (5)$$

Where N is α ract production rate and k is the co-efficient of absorption. A modified relation has been established in our laboratory (N.P. Singh et al., 1985) substituting cellulsose nitrate (LR-115 type 2) plastic track detector for nuclear emulsions and assuming radioactive equilibrium in the geological sample. The modified relation is :

$$10^6 N = (6.77\ U + 1.60\ Th)\ k \quad \ldots\ldots (6)$$

$$\text{Where } k = \frac{0.85}{\Sigma \frac{CS}{A}} \quad \ldots\ldots (7)$$

C is the concentration of an element of atomic weight A and S is the stopping power of alpha particles in that element.

Total track production rate is recorded by placing the plastic detector against the polished surface of the sample. U conc is determined independently by fission track technique and Th is estimated by α autoradiography using relation (6) hence this is termed as F/α metion (Fisher, 1677). Our analysis of phosphoritc samples from Mussorie area shows that U/Th results are comparable to those of gammaray ray spectrometry (N.P. Singh et al., 1985).

Boron/lithium determinations

Boron is a fairly ubiquitous element and has been measured in a variety or substances including water, minerals and rocks, glass, natural and man-made diamonds, steel alloys and a variety of biological material (Fleischer et al., 1975). The partitioning of boron in silicate melts as a function of temperature and pressure has also been studied by Seitz (1973).

Trace amounts of boron and lithium can be determined uniquely by recording their thermal neutron-induced reaction products in plastic track detectors. When several elements undergoing (n, α) reaction are present, it becomes difficult to use the track technique to identify the specific reaction. However, Pilione and Carpenter (1981) have developed the multiple discriminating detector technique for determination of in-situ distribution and abundance of lithium and boron in heterogeneous samples.

The element lithium which undergoes the reaction ^{6}Li (n, α)^{3}H is unique, since it produces a triton particle which is far more penetrating than alpha particle of the same energy. By inserting an absorber material between the sample and the plastic detector during thermal neutron irradiation, alpha particles from ^{10}B and ^{17}O are absorbed, and the triton track density is given by (Carpenter and Pilione, 1980).

$$T \propto \frac{CIF\sigma}{M} \qquad \ldots\ldots(8)$$

Where C is lithium concentration (ppm), I is isotopic abundance of ^{6}Li (%), F is neutron fluence (ϕt), σ is thermal neutron cross section, and M is at wt. of lithium. Similarly, quantitative boron determination has been made in Si samples by using ^{10}B (n, α) ^{7}Li reaction by Freyer et al. (1980). The track density is given by relationship :

$$T = 2.4 \times 10^{-26} \times \varphi th \times CB \qquad \ldots\ldots(9)$$

Where ϕth is thermal neutron flux (n/cm^2) and CB is conc. of boron (atomsicm^3). The practical sensitivity for boron detection is limited to 1ppm due to the presence of other competing (n, a) reactions, either in the specimen sample or in the detector itself.

Uranium/thorium prospecting

Radon isotopes are the daughter products of radium in the naturally occurring uranium and thorium series. These isotopes can diffuse through the soil and enter the atmosphere provided the permeability of the overlying rock and soil is sufficiently high. Because of their short half lives and being natural alpha emitters, these isotopes can easily be detected by α - sensitive plastic detectors.

The measurement of radon in the soil gas is a well established method of prospecting for uranium (Gingrich, 1975). In U-exploration surveys, there used to be difficulties in interpretation of anomalies in thoriferrous areas as thoron signal could not be discriminated properly from radon signal.

This difficulty has largely been overcome by the use of Radon. Thoron Discriminator fabricated by Atomic Minerals Division, Hyderabad (Ghosh and Soundararan 1984) by using special membranes to exclude aerosols and short-lived thoron isotope (Ward *et al.*, 1977).

The question of how far from uranium are elevated levels of radon can be recognised is critical. Even if the are is 10^4 times enriched in radom-emitting power relative to its surroundings, the excess signal only exceeds the background noise to a distance of lim from the ore, for an assumed typical value for of 1.2m based on the simple diffusion model (Fleischer and Mogro-Campero, 1981). This distance is many times greater than that of other exploration methods that utilize radioactivity (less than 30 cm for gamma rays from ^{238}U decay chain). But radon assisted finds of uranium at distances upto 200m are explained not on the basis of diffusion alone but due to vertical flow of gas in the ground. The added advantage of the track technique is that it recognizes radon anomalies which are generally missed by prompt-reading counters because it integrates signals over a time period that is much longer than that of the barometric fluctuations.

Search for geothermal sources

A geothermic field is usually associated with a volcanic region and, therefore, with an abundance of volcanic glasses such as obsidians. Fission track dating of volcanic glasses is a well-established techniques (Virk, 1985).

The magmatic chambers constitute an excellent source of heat for a geothermical system. These chambers can be geologically identified from the surface by its recent volcanic products. Therefore, the fission track age of the volcanic units is of great interest for a location of worthwhile thermal energy field. However, delimit the thermal area, the geological survey should emphasize the the tectonic and stratigraphic settings of the area, recent faulting and the distribution and age of young volcanic rocks. Preliminary surveys using SSNTD technique have been undertaken in countries like Mexico, Japan and Italy where large number of active volcanoes already exist.

Prospection for oil and natural gas

Naeser *et al.* (1976) pointed out that track annealing characteristics of apatite make it extremely sensitive to thermal perturbations, suggesting that fission track dating of apatite could be developed into a geothermic exploration technique. The advantage of apatite as a geological thermometer is that it can give information not only on maximum palaeo-temperatures but also on their variation through time. Therefore, the study of fission track dating of apatite is of great practical significance to prospecting for oil and natural gas in sedimentary basins where apatite is a common detrital mineral. Exploration work has been undertaken in China (Feng Shi, 1985) and Australia (Gleadow and Lovering, 1983) using SSNTD technique.

Earthquake prediction

Although radon by itself has rarely been used to predict an earthquake correctly, it has been correlated in many cases with earthquake activity (Gasli earthquake in Soviet Union May 17, 1976 and the Haichang earthquake in China, Feb. 4 1975 ;

both of magnitude 7.3) and is one of several useful precursors for earthquake prediction. Radon response to distant earthquakes (from 1 km to 1800 cm) is explained by dislocation model (Fleischer, 1981).

Sadovsky *et al.* (1972) observed a long term increase in radon followed by a short term transient in water from a deep well just prior to a major Tashkent earthquake. Since then many studies of radon in earthquake zones using SSNTD technique have been reported (Fleischer and Mogro-Campero, 1981 ; Scidel and Monnin, 1981, Siedel *et al.*, 1984 and Segovia *et al.*, 1985).

How does an impending earthquake lead to altered radon in the ground. The charges are generally ascribed to effects of the stress and strain build-up, which act either by altering the amounts released into pore spaces or by redistributing the free gas within the pore spaces. Multiple possibilities exist in each of these two categories.

Nuclear track detection system is the same in both U-exploration and earthquake prediction. However, in the search for uranium the usual objective is to collect many simultaneous measurements over an area but in earthquake prediction many sequential measurements are made at each site of interest.

Radon measurements using cellulose nitrate (LR-115) have been carried out since 1981, in and around active volcanaoes in Mexico and Costa Rica (Seidel *et al.*, 1984). Surveys of radon outgassing in connection with geothermal faults have been performed in Mexico (Cruz *et al* , 1985). Our own measurements of radon activity before and after slight earthquake tremors recorded at Amritsar show a great promise for SSNTD technique in earthquake prediction in the country.

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