



APPLICATION OF NEUTRON INDUCED FISSION TECHNIQUE IN THE DETERMINATION OF URANIUM IN DRINKING WATER

*M. AKRAM, N. U. KHATTAK, A. IQBAL¹, A. A. QURESHI, KAFYATULLAH and I. E. QURESHI

Physics Research Division, PINSTECH, P. O. Nilore, Islamabad, Pakistan

¹Physics Department, University of Azad Jammu and Kashmir, Muzaffarabad, AJK

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The Neutron Induced Fission Technique has been applied to determine uranium concentration in drinking water samples collected from natural springs of Reshian area, Azad Kashmir. The technique involves the simultaneous irradiation of unknown and known specimens, fixed on a track detector, with thermal neutrons and the development of fission tracks from the nuclear reaction $^{235}\text{U} (n, f)$. The tracks produced in the detector during irradiation are then counted after chemical etching. Uranium concentrations in drinking water samples are determined by comparing its fission track density with that of a standard of known uranium concentration. Uranium concentration in water samples from the study area has been found to vary from $0.03 \pm 0.01 \mu\text{gL}^{-1}$ to $6.67 \pm 0.14 \mu\text{gL}^{-1}$ with an average values of $1.57 \pm 0.06 \mu\text{gL}^{-1}$. The concentrations of uranium in drinking water are found to be less than the maximum acceptable concentration levels of 9-30 μgL^{-1} . The drinking water from the reported area has been found to be within safe limits as far as uranium related health hazards are concerned.

Keywords: Natural water, Uranium concentrations, Fission tracks, Etching, Health hazards.

1. Introduction

Uranium is a naturally occurring lithophilic element with three primordial isotopes; ^{234}U (0.0055 %), ^{235}U (0.720 %) and ^{238}U (99.2745 %), all of which are long-lived alpha emitters. Uranium is present in almost all minerals, rocks, sand and soil [1]. The quantitative determination of uranium in water is important because of its chemical and radiotoxic effects. Uranium may enter into drinking water from naturally-occurring deposits by leaching processes taking place in the earth's crust or as a result of human activity, such as mining and milling [1-2]. Therefore, it may be hazardous if inhaled or ingested in excessive quantities. Uranium may enter into human body mainly through intake of food and drinking water. Water having uranium concentration above the proposed Maximum acceptable concentration limits of 9-30 $\mu\text{g/l}$ is not safe for drinking purposes and it leads to harmful effects on health of humans. Hence, the assessment of the risk of health hazards due to high concentration of uranium in waters is most important.

This study was undertaken to estimate uranium concentration in drinking water collected from

different natural springs of Reshian area of Azad Kashmir (Fig. 1), with the help of Neutron Induced Fission Technique [3-6] using Solid State Nuclear Track Detectors which have a wide range of applications in science and technology [7-9]. The primary purpose of this investigation was to assess whether the general public is at risk from exposure level of uranium in water that might be used for drinking. This Pakistani held-Kashmir area containing spring water is comprised of sandstone, siltstone, shale, limestone conglomerate and alluvium ranging in age from (50-0) Ma.

The mechanism of registration of fission tracks in a detector for uranium determination by this technique is shown in Fig. 2. During irradiation in a nuclear reactor, the bombardment of ^{235}U with thermal neutrons causes a fission reaction to occur. The fast-moving charged fission fragments emitted from this reaction have sufficient energy to produce etchable fission tracks in an appropriately positioned detector [3, 11]. Subsequent chemical etching of the detector enlarges the latent tracks to a size easily observable with an ordinary optical microscope. The fission fragments resulting from the (n, f) reaction entering the track-etch detectors

* Corresponding author : akram@pinstech.org.pk :

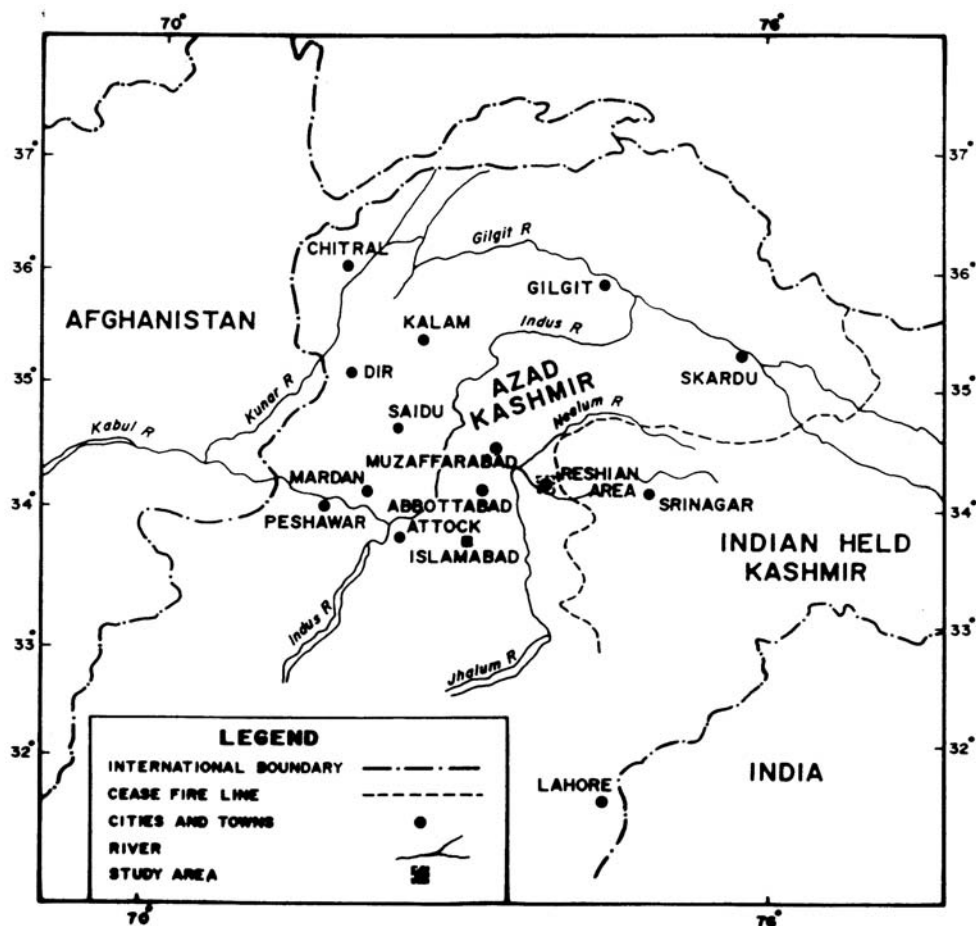


Figure 1. Map of the study area showing location of the Reshian area, Azad Kashmir

kept in contact with the unknown sample give the estimate of the fission fragment yields. A comparison of the fission-track density (tracks cm^{-2}) of these fission fragments from the sample with that of a standard sample of known concentration can directly be used for the determination of the uranium concentrations of water samples [3]. The optimum irradiation and etching conditions of Lexan detector for tracks revelation alongwith results for the estimation of uranium concentrations are described in this paper.

2. Experimentation

Thirteen drinking water samples were collected from different spring water sources of Reshian and its surrounding area in clean plastic bottles of 250 ml. Similarly standard solution of known concentration (250 mL solution containing $50 \mu\text{gL}^{-1}$ of uranium) was prepared from uranium oxide

(having certified composition of 99.94 % U_3O_8). The water samples alongwith standard solution were then transferred to glass beakers and heated continuously on a burner until 1.25 ml solutions of each water sample and standard were left behind. A drop of 0.05 mL of concentrated solution of each water sample and standard solution were taken with the help of an accurate calibrated micropipette and allowed to dry over surfaces of the detectors in dust-free environment at Nuclear Geology Laboratory, PINSTECH. The solid residue left on each detector was covered with another detector of the same size so as to sandwich the deposition layer in between two detectors. The detector used was 125 μm thick plastic polycarbonate ($\text{C}_{16} \text{H}_{18} \text{O}_3$) with trade name of Lexan® (General Electric Company, USA), the most widely used plastic material for recording fission particles. Ordinary perspex strips, obtained from local market, were used to ensure proper contact between the samples and detectors.

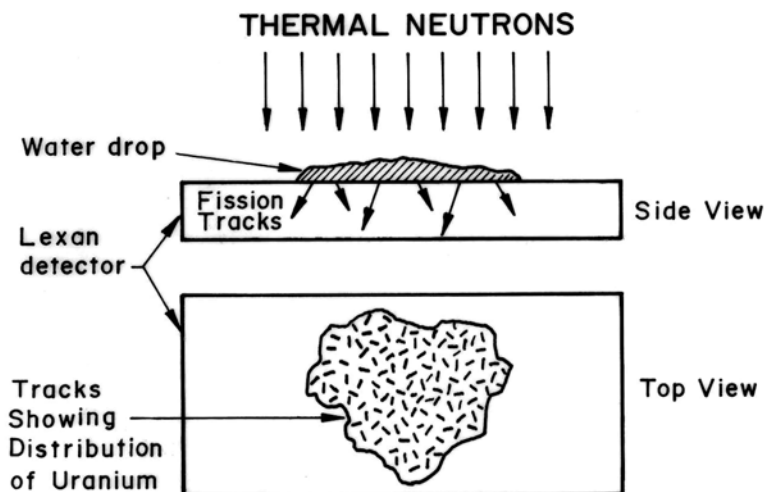


Figure 2. Schematic representation of the Neutron Induced Fission Technique used for uranium determination in drinking water.

The Lexan[®] detector assemblies along with one blank Lexan[®] detector (for background counting) were irradiated simultaneously with thermal neutrons in the Pakistan Research Reactor-I (PARR-1), PINSTECH, under a flux of the order of $10^{13} \text{ n cm}^{-2} \text{ s}^{-1}$ for various time intervals (20-200 seconds) for the optimization of irradiation time. After irradiation, the Lexan detectors were etched in 6.5M NaOH solution at $50 \pm 1^\circ \text{C}$. The experiment was repeated many times in order to achieve optimum irradiation and etching conditions for fission tracks revelation. Step-wise etching with five minutes intervals was done to visualize track development until the best tracks were obtained. An exposure time of 100 second in a thermal neutron flux of $10^{13} \text{ n cm}^{-2} \text{ s}^{-1}$ was established as an optimum exposure time, while the optimal etching time of detectors for these exposure conditions was 45 min.

The fission tracks produced in the water-drop area of the etched detectors were then counted under an optical microscope (Zeiss at 400x magnifications). In order to determine uranium concentration in water, its track density was compared with the track density obtained from the standard of known concentration. Because both the sample and the standard were irradiated and etched under similar conditions, the uranium concentration was calculated by the relation [3-5, 10]:

$$C_x = C_s [(\rho_x / \rho_s) (I_s / I_x) (R_s / R_x)]$$

Where the subscripts x and s stand for unknown and standard sample respectively; C, is the uranium concentration; ρ , the track density; I, isotopic abundance ratio of ^{235}U and ^{238}U and R, the range of the fission fragments. The quantities R_s / R_x and I_s / I_x are correction factors applied when the composition of the sample and the standard are different. Since the standard and unknown sample were same in composition, the values of R_s / R_x and I_s / I_x were taken to be unity in the present case.

3. Results and Discussion

The concentration of uranium measured in all thirteen drinking water samples collected from different springs of Reshian and its surrounding area has been summarized in Table 1. As the water samples were 200 times concentrated, therefore, the uranium concentrations in the last column of the Table 1 were divided by 200. The concentration of uranium present in the water was calculated in units of $\mu\text{g L}^{-1}$.

By making a measurement relative to the concentration of uranium in standard, many sources of uncertainty cancel, and the dominant uncertainty is the statistics of the track counting. The errors (\pm represents 1 standard deviation) in the track densities were calculated by multiplying track density by $(1/N)^{1/2}$, where N is the total number of tracks counted in that sample. The tracks of each sample were counted 2-3 times, and uranium concentrations were determined with 1 standard deviation. The systematic error due to

Table 1. Uranium concentration in water samples from different localities of Reshian area, Muzaffarabad, Azad Kashmir ($\mu\text{g/L}^{-1}$).

Sample No.	Location	Total tracks	Fields of view ^a	Average tracks	Track density ($\#/\text{cm}^2$) $\times 10^3$	U. conc. $\pm 1\sigma^b$ ($\mu\text{g/L}^{-1}$)
R-1.	Parsa Union Council	104	44	2.36	4.03 ± 0.40	0.22 ± 0.02
R-10.	Khari Pahari, Reshian	43	58	0.74	1.26 ± 0.19	0.07 ± 0.01
R-6.	Mine No. 2 (upper side)	1512	44	34.36	57.49 ± 1.50	3.18 ± 0.08
R-16.	Mine No. 1 (lower side)	2382	33	72.18	123.14 ± 2.52	6.67 ± 0.14
R-3.	Khata Panni Reshian	397	50	7.94	13.55 ± 0.68	0.73 ± 0.04
R-12.	Beside the nallah, Reshian	12	10	1.20	2.05 ± 0.59	0.11 ± 0.03
R-24.	Khari Pahari Reshian	684	32	21.38	36.47 ± 1.39	1.98 ± 0.08
R-17	Parsa Sanyare Lamnian	884	29	30.48	52.00 ± 1.75	2.82 ± 0.10
R-2.	Jay Shahadat A Y Ibn Jamal	16	44	0.36	0.62 ± 0.16	0.03 ± 0.01
B-5.	Bandbay Khun Bandi	785	45	17.44	29.76 ± 1.06	1.61 ± 0.06
T-13.	Tandali	488	31	15.74	26.86 ± 1.22	1.45 ± 0.07
S-14.	Subri-Langerpura	161	15	10.73	18.31 ± 1.44	0.99 ± 0.08
M-18.	Mahi Dhani	207	42	4.93	8.41 ± 0.58	0.46 ± 0.03
Overall Maximum						6.67 ± 0.14
Overall Minimum						0.03 ± 0.01
Overall average						1.57 ± 0.06

^a Area per field-of-view = $5.86 \times 10^{-4} \text{ cm}^2$

^b Statistical counting error = 1SD

clustering of tracks was small as the numbers of clusters found in this study were negligible. The reliability of the results obtained by this method primarily depends upon the accurate measurements of neutron flux and the number of counted fission tracks. However, the uncertainty due to the first factor is eliminated by comparing track counts for the same flux.

Influence of thorium, which can be the only other natural nuclide undergoing a fission reaction with reactor neutrons and can produce fission tracks, was not taken into consideration since the thorium content of water is usually very low ($\sim 10^{-4}$) $\mu\text{g/L}^{-1}$ compared with the uranium content. Moreover, thorium undergoes a fission reaction only with fast neutrons of energies higher than 1.2 MeV ($\sigma_f = 0.078 \text{ b}$ for fast neutrons). Since the neutrons used to irradiate the samples were thermalized, the contribution of thorium to tracks production in the detector was negligibly small.

The range of uranium concentration in drinking water samples under investigation has been found to vary from 0.03 ± 0.01 to $6.67 \pm 0.14 \mu\text{g/L}^{-1}$ with an average value of $1.57 \pm 0.06 \mu\text{g/L}^{-1}$ based on the analysis of one sample per site. This clearly

indicates that the uranium concentration in drinking water samples varies depending on the source area of water under study. The precision of the analytical procedure was found to be 4 %.

Higher uranium concentrations were observed in the spring water samples (R-6, R-16, R-17) collected from upper Reshian mine No. 2, lower Reshian mine No. 1 and Parsa Sanyaree, Laminian area, respectively, compared to all other samples in the studied areas. Uranium bearing mineral phases has been reported to occur in the graphite schists of the Salkhala Series and in the black shale of the area [11]. This might be the cause of having high concentration of uranium in these water samples.

Uranium concentration in the water sample taken from the spring near Reshian mine No. 2 (R-6) was $3.2 \mu\text{g/L}^{-1}$ while the concentration in the water sample collected from the spring near lower Reshian mine No. 1 (R-16) was $6.67 \mu\text{g/L}^{-1}$. The uranium concentration in the spring water near the lower mine was found to be two times concentration in the spring waters near upper mine. The high value of uranium concentration observed near lower Reshian mine No. 1 (R-16)

may be due to the fact that the springs near Reshian mine No. 2 is on the upper side. Spring water containing uranium near upper mine flows down towards lower mine No. 01 and mixes with lower mine spring water. Mining activities could be the reason for high uranium concentration in the spring water near lower Reshian mine (R-16), while the upper mine was not under operation.

From Table 1, it can be seen that the concentration of uranium in the water samples collected from Reshian area (sample No. 1-7) ranges from $0.07 \pm 0.01 \mu\text{gL}^{-1}$ to $6.67 \pm 0.14 \mu\text{gL}^{-1}$ with an average values of $(1.85 \pm 0.05) \mu\text{gL}^{-1}$, while uranium concentration in the water samples collected from north-west of Reshian towards Muzaffarabad (sample No. 8-13) ranges from $0.03 \pm 0.01 \mu\text{gL}^{-1}$ to $2.82 \pm 0.10 \mu\text{gL}^{-1}$ with an average of $1.23 \pm 0.06 \mu\text{gL}^{-1}$. This shows that average uranium concentration in Reshian hilly area is about 1.5 times higher than towards Muzaffarabad area, which can be attributed to be due to the presence of a known bed of uranium bearing graphitic schists in the Reshian area.

The main lithologies of the Resian area are sandstone, siltstone, shale, limestone and conglomerate. These rocks in Pakistan generally do not contain any excessive uranium content; hence no excessive contribution was expected from these rocks. However, the main contribution of uranium in drinking water appears to be from the graphitic schists and black shales in the surrounding areas from where these springs emerge. Some uranium may also have been incorporated due to the mining activity in the surrounding areas of these springs. The flowing surface water may also transport uranium from other geological formations as well encountered in its way into the spring's water.

The results of this study can be compared with the uranium concentration in water samples of the other places reported in the literature. Reported values of uranium contents are more than $30 \mu\text{g/l}$ in various U.S. community water supplies, whereas the average amount of uranium in drinking water (groundwater) is $3 \mu\text{gL}^{-1}$ [1] and $0.015\text{-}973 \mu\text{gL}^{-1}$ in domestic supplies in U.S. [12]. Uranium concentration in domestic and surface water samples were $0.67 - 20.26 \mu\text{gL}^{-1}$ in India [13]. In private groundwater supplies in Canada, uranium concentrations upto $700 \mu\text{gL}^{-1}$ have been found [14]. This comparison shows that the range of uranium concentration in drinking water as

obtained in this study lies towards the low side of the ranges reported for other locations in the world. The concentration of uranium in drinking water in the study areas was lower than provisional maximum acceptable concentration levels of $9\text{-}30 \mu\text{gL}^{-1}$ [15-16].

4. Conclusions

The range of uranium concentration in drinking water samples under investigation were found to vary from $(0.03 \pm 0.01) \mu\text{gL}^{-1}$ to $(6.67 \pm 0.14) \mu\text{gL}^{-1}$ with an average values of $(1.57 \pm 0.06) \mu\text{gL}^{-1}$. The concentration of uranium is found to be more in Reshian area than its N-W surrounding area. The observed values are within the provisional prescribed safe limits of $9\text{-}30 \mu\text{gL}^{-1}$. Consequently, the health hazards related to uranium in drinking water are negligible and use of this water for drinking and other purposes may not be a health risk. This area is, therefore, safe from the health hazards related to uranium in drinking water. However, health hazards related to other toxic and trace elements present in drinking water should also be monitored so that health hazard can be minimized.

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