1. INTRODUCTION

1.1 General Introduction

Uranium is a natural and commonly occurring radioactive element which is commonly found in rocks, soil surface and underground water, air, plants and animals. It is the heaviest element with atomic number of 92. The name derives from the planet Uranus, which in roman mythology is "Father Heaven". The electronic configuration of uranium is [Rn] $5f^3$ $6d^1$ $7s^2$. The details of the properties are given Appendix I (1). The common ionic species are U (III), U (IV), U (V) and U (VI). Hexavalent uranium, in the form of uranyl ion (UO_2^{2+}), is the most stable oxidation state (2).

Elemental uranium consists of three naturally occurring isotopes in the proportion 99.28% U-238, 0.72% U-235 and 0.0057% U- 234 respectively. Eleven other transient species have been identified but their short half lives preclude natural occurrence. The decay schemes of U- 235 and U-238 include the decay modes of alpha, beta, gamma, thermal energy emission and spontaneous fission; all but the later terminate in stable lead products. Some nuclear properties of naturally occurring isotopes are presented in Table 1.1(3).

Table 1.1: Properties of naturally occurring uranium (3)

Isotopes	Relative abundance by weight	Half Life (years)	Specific activity ($Bqmg^{-1}$)
U-238	99.28%	4510000000	12.4
U-235	0.72%	710000000	80
U-234	0.0057%	247000	231000

1.2 Historic and Present Uses of Uranium

For many years, uranium was used primarily as a colorant in ceramic glazes, producing colors that ranged from orange-red to lemon yellow. It was also used for tinting in early photography. Its radioactive properties were not recognized until 1896, and its potential for use as an energy source was not realized until the middle of the 20th century. Its primary use is as fuel in nuclear power reactors to generate electricity. It is also used in weapons applications and in small nuclear reactors to produce isotopes for medical and industrial purposes around the world (4).

1.3 Natural Abundance

Uranium is the 48th most abundant element in the earth crust. It is more abundant than some well known elements such as Ag, Hg, Cd and I (5). Small amounts of uranium are found almost everywhere in soil, rock, and water. However, concentrated deposits of uranium ores are found in just a few places, usually in hard rock or sandstone. These deposits are normally covered over with earth and vegetation. It is a reactive metal so; it is not present as free uranium in the environment. Uranium can combine with other elements in the environment to form uranium compounds. Uranium in the environment is mainly found as uranium oxide, typically as UO_2 and sometimes as UO_3. The average concentration of natural uranium in soil is about 2 ppm. Concentration of uranium in granite ranges from 2ppm to 20 ppm. Uranium in higher concentrations (50-1000 ppm of soil) can be found in soil associated with phosphate deposits. In air uranium exists as dust. Very small, dust-like particles of uranium in the air are deposited onto surface water, plant surfaces and soils (3). Uranium is also found in phosphate rock, lignite, monazite sands and can be recovered commercially from these (1). Approximate ranges of uranium concentration in various materials are presented in Table 1.2.

Table 1.2: Abundance of Uranium in Natural Materials (6).

Materials	**Reported Concentration Range (ppm)**
Cosmos	0.01-0.1
Meteorites:	
Stony achondrites	$0.15\text{-}14.5 \times 10^{-2}$
Stony chondrites	$0.62\text{-}2.8 \times 10^{-2}$
Irons	$<0.003\text{-}1.7 \times 10^{-2}$
Tektites	1.2 – 3.0
Igneous Rocks:	
Ultramific (dunties and peridotities)	0.001-0.8
Mafic (gabbro and diabase)	0.3-3.4
Intermediate (diorite and quartz diorite)	0.1-11.0
Sialic (granite, syenite and monzonite)	0.15-21
Pegmatities	1.0-4.0
Sedimentary Rocks:	
Black Shales	3.0-250.0
Red, green, and, gray shales	1.2-12.0
Orthoquarzites	0.2-0.6
Limestone and dolomite	0.1-9.0
Bentonite	1.0-21.0

Bauxite	3.0-27.0
Halite	0.01-0.02
Anhydrite	0.25-0.43
Metamorphic Rocks:	
Marble	0.11-0.24
Slate	1.2-6.1
Phyllite	1.0-2.7
Schist	1.8-2.9
Gneiss	4.5-15.0
Amphibolite	2.6-4.1
Granulite	3.2-7.0

1.4 Mineralogy

The main ore minerals of the uranium are the oxides uraninite (pitchblende), coffinite, brannerite and davidite. More than 100 secondary (supergene) uranyl (UO_2^+) minerals are known, of which the most common are gummite, schroekingerite, zippeite, autunite, carnotite, tyuyamunite and uranophane.(7).

The classification of uranium bearing mineral are based on chemical scheme. Uranium, ideally UO_2 is the most common mineral and found in pegmites and granites. Uranium minerals like carnotite, tyuyamunite, coffine, maonazite, limonite, zircon, magnenite and uraninite are detected in different parts of Nepal (8). High radioactivity has been observed at the field location like Jagat area and Chunikhel of Northern Shivapuri Hills and Bramkhel of Central Kathmandu Valley (9). Table 1.3 summarizes data of more important uranium minerals.

Table 1.3 Properties and amount of Uranium in Some Minerals (7, 10).

Mineral	Color	Chemical Composition	% Uranium	Hardness	Specific gravity
Uraninite (pitchblende)	Black-greenish	$UO_2 + (UO_3,Th,Pb)$	45-85	5-6	7-10
Coffinite	Black	$U(SiO_4)_{1-x}.(OH)_{4x}$	-	5-6	5.1
Euxenite-polycrase	Dark brown	$(Y, Ca,Ce,U,Th)(Nb,Ta,Ti)_2O_6$	0.6-9	5.5-6.5	4.3-4.9
Brannerite	Black	$(Y,Ca,Fe,U,Th)_3(Ti,Si)_5O_{16}$	27-43	4.5	4.5
Samarskite	Black	$(Y,Ca,Fe,U,Th)(Nb,Ta)_2O_6$	-	5-6	5.7
Davidite	Black	$(Fe,Cu,U)(Ti,Fe,V,Cr)_3(O,OH)_7$	4	6	4.3-4.9
Carnotite	Yellow to greenish yellow	$K_2(UO_2)_2(VO_4)_{2.x}H_2O$	52-55	soft	4.7
Autunite	Yellow to pale green	$Ca(UO_2)_2(PO_4)_2.xH_2O$	45-48	2-2.5	3.0-3.2
Tyuyamunite	Yellow	$Ca(UO_2)_2(VO4)_2.xH_2O$	50-56	2	3.7-4.3
Uranophane	Greenish-yellow	$Ca(UO_2)_2Si_2O_7.xH_2O$	55	2.5	3.7-3.9
Torbernite	Green	$Cu(UO_2)_2(PO_4)_2.xH_2O$	47-50	2.5	3.2
Thucholite	Black	Th,U,C,H,O	Variable	3.5-4	1.8

1.5 Natural Radioactivity in Building Materials

Radionuclides are found in air, water and soil. They are even found in us, being that we are products of our environment. Every day, we inhale and ingest radionuclides present in air, food and water. Natural radioactivity is common in the rocks and soil that makes up our planet, in water and oceans, and in our building materials. Among the building materials marble and granite are also radioactive. Marble and granite both are natural stones. Both stone are quarried from earth. Granite is a highly dense material composed deep inside the earth's core while marble is formed from sediments under the seabed. Both solidify into stone after millions of years but the mineral composition of the two stones makes marble and granite different (11, 12). The chemical composition of granite is

SiO_2: 73.4%,Fe_2O_3: 0.84%,Al_2O_3: 13.54%,TiO_2: 0.15%,CaO : 1.15%,MgO : 0.15%,P_2O_3: 0.05%,K_2O : 4.85%,Na : 0.38%,Mn : 0.01%.

The chemical composition of marble is

SiO_2: 3.55%,FeO : 0.29%,Fe_2O_3: 0.04%,Al_2O_3: 0.11%,CaO : 51.69%,MgO : 2.18%,K_2O : 0.26%,Na_2O : 0.92%,Li_2O : 0.47%,P_2O_3: 1.63%

Listed below are a few of common building materials and their estimated levels of uranium, thorium and potassium (13).

Table 1.4: Estimates of concentrations of uranium, thorium and potassium in building materials(13).

Material	**Uranium**		Thorium		Potassium	
	ppm	**mBq/g (pCi/g)**	ppm	mBq/g (pCi/g)	ppm	mBq/g (pCi/g)
Granite	**4.7**	**63 (1.7)**	2	8 (0.22)	4.0	1184 (32)
Sandstone	**0.45**	**6 (0.2)**	1.7	7 (0.19)	1.4	414 (11.2)
Cement	**3.4**	**46 (1.2)**	5.1	21 (0.57)	0.8	237 (6.4)
Limestone concrete	**2.3**	**31 (0.8)**	2.1	8.5 (0.23)	0.3	89 (2.4)
Sandstone concrete	**0.8**	**11 (0.3)**	2.1	8.5 (0.23)	1.3	385 (10.4)

Dry wallboard	**1.0**	**14 (0.4)**	3	12 (0.32)	0.3	89 (2.4)
By-product gypsum	**13.7**	**186 (5.0)**	16.1	66 (1.78)	0.02	5.9 (0.2)
Natural gypsum	**1.1**	**15 (0.4)**	1.8	7.4 (0.2)	0.5	148 (4)
Wood	**-**	**-**	-	-	11.3	3330 (90)
Clay Brick	**8.2**	**111 (3)**	10.8	44 (1.2)	2.3	666 (18)

1.6 Geochemistry

The geochemistry of uranium is primarily a study of the migration and distribution of uranium under the influence of physio-chemical forces. The geochemical behavior of the elements has been broadly discussed by Goldsmidt, who classified the elements under four headings: Siderophile (association with metallic iron); Calcophile (affinity for sulfur); Lithophile (affinity for oxygen); Atomophile (affinity for elemental state). The primary dispersion of uranium is largely a function of its chemical behavior, ionic potential and affinity for the major elements like oxygen and metallic iron. However, the secondary dispersion of uranium in the zone of weathering is largely controlled by E_h (redox potential) and pH (hydrogen ion concentration) condition of the geochemical environment.

The pH is an extremely important in determining the mobility of uranium in the weathering cycle. Uranium is soluble in alkaline solution at pH range 7.5. It gets precipitated in acidic environment at pH range 6.5 and below. Redox potential of an environment contributes at pH ranges 6.5 and below. Redox potential of an environment contributes significantly in the fixation and distribution of this element. Uranium leached out from its source rocks is transported in surface water as dissolved uranyl dicarbonate complex (pH= 7.5 – 8.5). Uranium is soluble in the presence of CO_2 and at low temperature and pressure either in the IV or VI valence state, depending on the pH and E_h of the solution. Tetravalent uranium can be transported in solutions with pH values less than 3. Transportation of hexavalent state is possible in slightly acidic or alkaline solutions at E_h values ranging +0.3 to -0.4 volts. The U^{4+} ions are the most abundant species in nature and U^{6+} ions are responsible for their behavior in magmetic or postmagmetic environments (14).

1.7 Uranium Cycle

Igneous rocks of acidic composition with high silica content represent the original source of uranium with the richest concentration of the element occurring in hydrothermal and supergene deposits. In igneous rocks, uranium is present in the tetravalent form, which in the late *magmetic* stage possibly gets oxidized to the $(UO_2)^{2+}$ form. In later form, uranium is highly soluble and so it is carried away in solution, leaving behind Th $^{4+}$. The transformation of U^{4+} to U^{6+} imparts mobility to uranium where as thorium whose valency does not change is left behind in the relatively high temperature *pegmatitic* phase **(14)**. Thus, the difference in their geochemical behavior makes uranium and thorium part company in the late *magmatic* stage. The migratory uranyl ion is again reduced to its tetravalent state as,

$$3H_2O + 2Fe^{2+} + UO_2^{2+} \longrightarrow Fe_2O_3 + UO_2 + 6H^+$$

The main genetic type of uranium deposits and the cycle of formation of each type indicating the source of uranium solution of disintegration, precipitation are illustrated in the Figure 1.1 given below:-

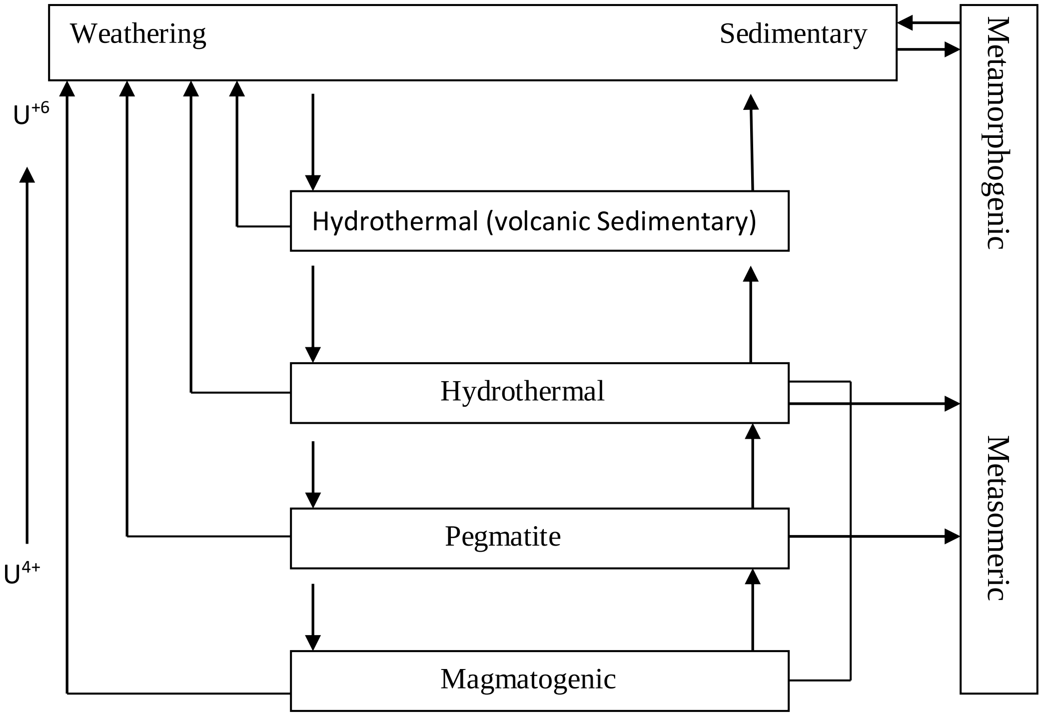

Fig. 1.1: The Uranium Cycle, (Virnave, 1988) **(14)**.

1.8 Public Health Statement of Uranium

Uranium is a chemical substance that is radioactive. Scientists have never detected harmful radiation effects from low levels of natural uranium, although some may be possible. Many people as well as animals have developed signs of kidney disease after intake of large amounts of uranium. There is also a chance of getting cancer from any radioactive material like uranium. Uranium can decay into other radionuclide, which can cause cancer if we are exposed to enough of them for a long enough periods. The chance of getting cancer is greater if we are exposed to enriched uranium because it is more radioactive than natural uranium. The Committee of the Biological Effects of Ionizing Radiation (BEIR IV) reported that eating food or drinking water having normal amount of uranium will most likely not cause cancer or other health problems in most people. The Committee used data from animal studies to estimate that a small number of people who steadily eat food or drink water that has larger than normal quantities of uranium in it could get a kind of bone cancer called sarcoma. The Committee reported calculations showing that if people steadily eat food or drink water containing about 1pCi of uranium everyday of their lives, bone sarcomas would be expected to occur in about 1 to 2 of every million people after 70 years, based on the radiation dose alone.

International organizations like the International Commission on Radiological Protection (ICRP) and the National Council on Radiation Protection and Measurements (NCRP) provide on recommendations for protecting people from materials, like uranium, that give off ionizing radiation. The federal government considers these recommendations and develops regulations and guidelines to protect public health. EPA has set a goal of no uranium in drinking water. EPA has also decided that any accidental uranium waste containing 0.1 curies of radioactivity must be cleaned up (15).

1.9 Behavior of uranium in the body

Uranium is introduced into the body mainly through ingestion of food and water and inhalation of air. The size of the uranium aerosols and the solubility of the uranium compound in the lungs and gut influence the transport of uranium inside the body. Coarse particles are caught in the upper part of the respiratory system from where they are exhaled or transferred to the throat and then swallowed. Fine particles reach the lower part of the lungs. If the uranium compounds are not easily soluble, the uranium aerosols will tend to remain in the lungs for a longer period of time, and deliver most of the radiation dose to the lungs. They will gradually dissolve and be transported into the blood stream. For more soluble compounds, uranium is absorbed more quickly from the lungs into the blood stream. About 10% of it will initially concentrate in the kidneys. Most of the uranium ingested is excreted in faeces within a few days and never reaches the blood stream. Most of the uranium in the blood stream is excreted through urine in a few days, but a small fraction remains in the kidneys, bones and other soft tissue (3).

1.10 Quantitative Analysis

Various procedures are available for the quantitative determination of uranium. Trace amounts of the uranium may be analyzed by neutron activation analysis (Yaprak et al., 1993) **(16)**; Inductively coupled plasma- spectrometry (Sasmaz et al., 2008)**(17)**; Fluorometric and Colorimetric (Adams and Maeck, 1954)**(18)**; Flow injection analysis (Hirano et al.,2003)**(19)**; Spectrophotometric analysis (Khan et al., 2005**(20)**; Ramakrishna and Murthy, 1980**(21)**; Leonardo et al., 1999 **(22)**; Tomitsugu Taketatsu, 1984**(23)**; Oscar and Michael, 1957**(24)**).

1.10.1 Spectrophotometric Method

Concentration of uranium in the various environmental samples can be determined by various methods like Spectrophotometric, Fluorometric, Colorimetric, flow injection analysis, neutron activation analysis and so on. Among them, spectrophotometric methods (Ramakrishna and Murthy, 1980**(21)**, Khan et al., 2005**(20)**, Leonardo et al., 1999 **(22)**, Tomitsugu Taketatsu, 1984**(23)**, Oscar and Michael, 1957**(24)**) are the one, which are used to measure the uranium in the environmental samples owing to their simplicity regarding instrumentation and handling. By measuring the absorbance of the colored complex formed by the combination of uranium solution and spectrophotometric reagents, the concentration of uranium is determined with reference to an analytical curve prepared from the standards.

1.10.2 Spectrophotometer

The basic parts of the spectrophotometer are a light source, a holder for the sample, a diffraction grating or monochromator to separate the different wavelength of light and a detector. The radiation source is often a Tungsten filament, a deuterium arc lamp is continuous over the ultraviolet region (190-400 nm)**(25)**, and more recent use are light emitting diodes (LED) and Xenon Arc Lamps for the visible wavelengths. The detector is typically a photodiode. Photodiodes are used with monochromators, which filter the light so that only light of a single wavelength reaches the detector.

1.10.3 Theoretical basis of Spectrophotometric Analysis

Beer-Lambert Law states that "when a beam of monochromatic radiation is passed through a solution of an absorbing substance, the rate of decrease of intensity of radiation with thickness of absorbing solution is proportional to the intensity of incident radiation as well as to the concentration of the solution".

Beer-Lambert law, also known as Beer's law or the Lambert-Beer Law, provides the theoretical basis of spectrophotometric analysis **(26)**. Mathematically, they are combinely expressed as,

$A=a\ b\ c$……………………………………………………………(1)

Where, A=absorbance or optical density

a= constant called absorptivity

b=length of the absorbing homogenous medium

c= concentration of the absorbing species

A is also defined as

$A=\log I_0/ I$………………………………………………(2)

where, I_0 = intensity of the incident light

I = intensity of the transmitted light

The unit of “a” will depend on how the concentration is expressed. If the concentration “c” is expressed in $molL^{-1}$ and “b” in centimeter then “a” is given by the symbol ε and is called molar extinction coefficient or molar absorptivity. In other words, if the concentration is expressed in mole fraction i.e. a dimensionless fraction, the molar absorptivity ε takes the same dimension as the absorption coefficient. However, if the concentration is expressed is moles per unit volume, the molar absorptivity ε is used in the $L\ mol^{-1}$.

Equation (1) can be written as

$A=\varepsilon bc$…………………………………………………… (3)

The sensitivity of spectrophotometric method depends on the value of ε. The higher its value, the more the sensitive is the method.

The absorbance, A also depends upon the wavelength of the incident light. For a particular absorbing species, there is a particular wavelength for which the absorbance is maximum, called λ_{max} and it is a characteristic of the colored absorbing species.

The Beer - Lambert law describes the linear relationship between absorbance and concentration(27).

However, there are the restrictions to the law and the linearity of the Beer- Lambert Law is limited by the chemical and the instrumental factors. Causes of the non - linearity includes,

- Deviations in molar absorptivity coefficients at high concentration due to the electrostatic interactions between the molecules.
- Scattering of the light due to the particulates in the samples and changes in the refractive index at high analyte concentration.
- Very large and complex molecules.

However in practice, provided that the steps are taken to ensure that the concentration is measured in the linear part of the calibration functions, the Beer – Lambert law applies.

The applicability of the Beer - Lambert law may be tested for the any particular system if one measures the absorbance for each of a series of solution of known concentration of the absorbing species. If the plot of the experimental data in terms of absorbance versus concentration ‘C’ yields a straight line passing through the origin, the Beer - Lambert law is valid. Beer - Lambert law is applicable only for the low concentration of the solution. At higher concentration of the solution, the Beer – Lambert is not applicable. By measuring the absorbance of a series of solution containing known concentration of absorbing species, the suitable calibration curve can be drawn. With the help of this calibration curve, it is possible to determine the concentration of absorbing species.

1.11 Literature Survey

The research related to determination of uranium from environmental samples has been a widespread interest in the field of analytical chemistry. Now a day, a various methods have been used for the determination of uranium in environmental samples which includes neutron activation analysis, fluorometric and flow injection analysis, gravimetric, coulometric and spectrophotometric.

In 1954 Adams and Maeck(18) used fluorometer and colorimeter for micro determination of uranium in rocks and minerals. Samples containing less than 10 ppm of uranium were best analyzed by fluorometric and more than 10 ppm by colorimeter analysis.

In 1957 Nietzel et al. (24) determined the uranium with thiocyanate in butyl cellosolve-methyl isobutyl ketone in water medium by using the spectrophotometer. This method for the determination of uranium in ores and leach liquors consists of separation of uranium from interfering ions by extraction into methyl isobutyl ketone using aluminium nitrate as salting agent. The optimum range of uranium concentration is from 0.4 to 2.0mg of uranium oxide (U_3O_8) in the sample aliquot.

In 1986 Kanai, et al. **(28)** used the fluorimetry and extractive spectrophotometry to measure the concentration of uranium in 36 geological samples. This method was based on the complexation of uranium with Arsenazo (III) after solvent extraction with TOPO (tri-n-octyl phosphine oxide) and used to detect the ultra trace amount of uranium.

In 1997 Saron and Baishya**(29)** used the spectrophotometric method to determine the uranium in soil, stream sediments and rock samples using 5-(2'-carboxyphenyl)azo-8-quinolinol in the non toxic surfactants of Triton X-100. This method is sensitive over the range 0-3.3 μgml^{-1} of uranium.

In 1999 Tiexeira et al. (22) used spectrophotometric method to determine uranium using 2-(2-Thiazolylazo) p-cresol in presence of surfactants. This method allowed the determination of uranium in the range from 0.30 to $12.0 \mu gml^{-1}$. This is a pH sensitive method. A maximum and constant absorbance was observed at pH ranging 6.0 to 7.0.

In 2002 Ali et al. (30) used the counter current extraction process for recovery of Uranium (IV) from phosphoric acid using Octa-Phenyl Acid Phosphate (OPAP) extractant. It was based in the bench scale results; a continuous counter current extraction flow sheet was developed and tested using a 16 stage horizontal mixer settler.

In 2003 Hirano, et al. (19) studied simultaneous determination of uranium and thorium by flow injection analysis using selective masking. The injected sample solution was first mixed with a reagent solution containing Chromazurol S (CAS) and Cetyltrimethyl Ammonium Chloride (CTMAC) and the absorbance of Uranium - and Thorium – CAS complexes were measured in the first flow cell at 620 nm. The calibration graphs were linear at least over the ranges 0.1 to 10 $mgdm^{-3}$ for Uranium and 0.08 to $8mgdm^{-3}$ for Thorium.

In 2003 Yordanova, et al. (31) studied the uranium and plutonium content in soils and plants from the water catchment basin in the Rila mountains by using alpha spectrometry. Results of the radiochemical determination of ^{234}U, ^{235}U, ^{238}U and Pu activities in 544 samples of soil, plant and water from the lake region were reported. The ^{234}U and ^{238}U activities determined in the soil and sediment samples vary from 40 to 2390 Bq/kg dry weight.

In 2003, Kaphle and Khan (8) carried out radiometric survey in different parts of Nepal. They were able to trace some radioactive bodies. The radioactive minerals were reported in Makanwanpur, Baitadi, Rasuwa, Thumki-Jagat area and Kakani-Panchamane area of Kathmandu valley.

In 2004, Shrestha(9) used spectrophotometer for chemical analysis to estimate radioisotopes like uranium and Multi Channel Analyzer was used for physical analysis to determine terrestrial radioisotopes like Uranium, thorium, Potassium and artificial radioisotopes of caesium. High radioactivity at the field locations like Jagat and Chunikhel areas of Northen Shiva Puri Hill and Bhramkhel of the central Kathmandu valley was reported.

In 2005 Khan, et al. (20) used the spectrophotometric method to determine the uranium concentration using Arsenazo (III) as chromogenic reagent in perchloric acid. This method was found to be accurate with 96-98.6% recovery rate. This method had been successfully applied to the standard reference materials and ore samples.

In 2009 Lutfullah, et al. (32) used optimized and validated spectrometric method for the quantitative analysis of uranyl ion in the soil sample. This method was based on the chelation of uranyl ion with piroxicam to produce yellow complex in 1, 4-dioxin-water medium and effective in the concentration range of 6.75×10^{-2} to 9.45×10^{-1} $\mu g ml^{-1}$.

In 2000 Sumithrarachchi, et al. (33) evaluated the gamma radiation exposure in a room from construction material. The result showed that the annual effective dose from construction materials was directly proportional to the activity concentration of the radionuclides present in the construction materials.

In 2002 Vosniakos, et al. (34) measured natural radioactivity concentration of building materials in Greece. Natural radionuclides (^{238}U, ^{232}Th and ^{40}K) concentrations in specially prepared samples were measured utilizing the direct gamma ray counting method.

In 2003 Papachristodoulou et al. (35) used HPGe γ – ray spectrometry to assess the isotopic composition of uranium in soils. The uranium activity in the samples varied from 48 to 112 $Bqkg^{-1}$, whereas the activity ratio of $^{238}U/^{235}U$ averaged 23.1±4.3.

In 2003 Ahmad et al. (36) determined the uranium occurrences in Malakand granite and granitic gneisses. The collected data revealed that high chemical uranium values were due to uraninite, which occurs as inclusion in biotite along joint planes parallel to regional foliation.

In 2003 Menon et al. (37) used γ- ray spectrometry for the estimation of abundances of heat – producing elements, K, U and Th in some of the granites and gneisses of the Bundelkhand and Bastar terrains *in situ*.

In 2005 Al-Jarallah et al. (38) used active radon gas analyzer with an emanation container for the measurement of radon exhalation for a total of 205 selected samples of construction materials used in Saudi Arabia. The ^{226}Ra content of the granites varied from below the minimum detection limit up to 297 Bqkg^{-1} with an average of 83±73(1σ) Bqkg^{-1}.

In 2006 Salama et al. (39) used CR – 39 and LR – 115 – II solid – state nuclear track detectors (SSNTDs) to determine the thorium to uranium ratios in different soil samples.

In 2007 Saleh et al.(40) used high resolution γ-ray spectrometry with an accumulating time for about 80000 second each to measure γ – ray spectra and from measured γ – ray spectra, activity concentrations were determined for marble samples ^{238}U(0.71-44.1 Bq/kg), while for granite ^{238}U(0.03-314.4 Bq/kg).

In 2009 Gupta et al. (41) used alpha sensitive LR – 115 type II plastic track detectors to estimate alpha radioactivity from radon emanated from some building construction materials viz; stones, soil, cement and fly ash.

In 2009 Mehra et al. (42) measured natural radioactivity in Brick Samples using gamma – ray spectrometry.

Thus it is interesting to carry radiochemical studies on different construction materials.

2. OBJECTIVE OF THE WORK

The objective of this work is to determine the amount of uranium by chemical method and gross radioactivity by physical method in different marble and granite samples. The specific objectives of the study are as follows:

1. To optimize the quantitative extraction of uranium from different marble and granite samples collected from local market.

2. To determine the gross radioactivity of the marble and granite samples.

3. To estimate the amount of uranium present in the marble and granite samples.

3. EXPERIMENTATION

3.1 Preparation of Reagents

All the reagents used were of Analytical Grade. These reagents were used without any further purification; the details are given in Appendix II.

a) Preparation of Stock Uranium Solution (1000 µgml^{-1})

2.109 g of pure uranium nitrate ($UO_2(NO_3)_2.6H_2O$) was weighed out accurately and dissolved in 10 percent nitric acid made in double distilled water. Final volume was made to 1000 ml in volumetric flask and used as stock solution in which the uranium content was as follows:-

1ml of uranyl nitrate solution = 1000 µg of uranium.

b) Preparation of Working Uranium Solution (10 µgml^{-1})

1ml of the stock uranium solution was diluted to 100ml of a volumetric flask using double distilled water.

1ml of uranyl nitrate solution = 10 µg of uranium

c) Preparation of Arsenazo III Solution

0.02g of Arsenazo (III) was accurately weighed and it was dissolved in double distilled water and final volume of the resulting solution was made up to mark in 100ml volumetric flask. Thus obtained solution contained the Arsenazo (III) as follows:

1ml of Arsenazo (III) solution = 0.2 mg of Arsenazo (III)

d) Preparation of Hydrochloric Acid (0.1M)

2.08ml of concentrated hydrochloric acid was diluted up to the mark in 250ml volumetric flask.

e) Preparation of Aluminium Nitrate

Saturated solution of $Al(NO_3)_3.9H_2O$ was prepared in 100 ml beaker using distilled water. For the analytical works, fresh solution was prepared every day.

3.2 Experimental Instruments

a) Electronic Balance (GT210V) was used for weighing.

b) pH strips were used to measure the pH.

c) Spectrophotometer (WPA – Linton, Cambridge Type S – 104) with 10 mm glass cells were used to estimate the uranium content in samples.

d) Multichannel Analyzer (MCA) coupled with NaI (Tl) crystal detectors were used to measure the gross radioactivity of the construction materials. This instrument is exclusively designed for gamma ray analysis with the crystal of NaI (Tl) sized 3”×3”. The model of this instrument is named ORTEC – NORLAND’S – 5600 MCA, which has been designed to fulfill the needs for precision, reliability and versatility in radiation analysis.

3.3 Procedure

3.3.1 Collection of Samples

Different types of granite and marble samples were collected in plastic bags from local market.

3.3.2 Absorption Spectra of Uranium (IV) Arsenazo (III) Complex and Adherence to Lambert- Beer Law.

Measured volumes like 0mL, 1.25mL, 2.5mL, 3.75mL, 5mL, and 6.25mL of uranium solution of concentration 10ppm were taken. The 10ppm uranium working solution was prepared by diluting 1 ml of uranium stock solution in 100ml of volumetric flask. The solutions taken in different beakers were acidified with 5ml concentrated hydrochloric acid and 2 g of granulated zinc was added. The reduction process was allowed to proceed for 5 minute. This solution was then filtered through glass wool; thorough washing with 1:1 hydrochloric acid ensured complete separation. The filtrate was collected in 25ml volumetric flask. Then 5ml of 0.02 percent Arsenazo (III) was added and the volume was made up to the mark using 1:1 hydrochloric acid. The U (IV) present in solution formed a pink color complex with Arsenazo (III). The absorption spectra of pink colored U (IV) Arsenazo (III) complex was recorded in Spectrophotometer against reagent blank and λ_{max} was determined. At λ_{max} the absorption spectra of other solution was measured against reagent blank. The absorption spectra and Lambert – Beer plot are given in figure 4.1 and 4.2(21).

3.3.3 Preparation of Calibration Curve for the Determination of Uranium

Measured volumes of uranium solution of 10ppm such as 0.5ml, 1ml, 1.5ml, 2ml, and 3ml were taken and acidified with 5ml of concentrated hydrochloric acid and 2g of granulated zinc was added. The reduction process was allowed to proceed for 5 minutes. This solution was filtered through glass wool and was washed with 1:1 hydrochloric acid. The filtrate was collected in 25ml of volumetric flask. Then 5ml of 0.02 percent Arsenazo (III) was added and made up to the mark with 1:1 hydrochloric acid. The resulting solutions were of concentration 0.2ppm, 0.4ppm, 0.6ppm, 0.8ppm and 1.0ppm. The absorbance of pink colored complex was measured against reagent blank at λ_{max}. Similarly, a calibration curve for various concentrations like 0.02ppm, 0.04ppm, 0.06ppm, 0.08ppm, 0.10ppm was prepared. The plots are given in Figure 4.2 A and figure 4.2 B (21) respectively.

3.3.4 Determination of Uranium in collected sample

1 kg of each sample was taken and crushed into fine powdered form by using grinder and was sieved (particle size 125 µm). Thus formed finely powdered sample was sholved into the conical pile as a standard procedure. The top of the cone was then flattened out and divided into the quarters. Then the opposite quarters of the pile were removed. The remaining quarters were mixed to form a smaller conical pile and again quartered. This process was repeated for each sample, further crushing being carried out when necessary to till a suitable weight (200 gm) was obtained. Then, 100 ml of distilled water was added into the sample. Few drops of concentrated HNO_3 were also added and stirred for half an hour. It was allowed for sedimentation. The solution was filtered. The sample was again washed with 100ml distilled water and filtered. Both the filtrates were mixed and dried to make about 5 ml. Then 10 gm of aluminium nitrate was added and stirred to dissolve. The pH of this solution was adjusted around 1.0 by using powdered ammonium carbonate and pH meter. The solution was quantitatively transferred to 100 ml separating funnel. Then 1 ml of saturated aluminium nitrate solution was used for each rinsing.

After extraction with 5 ml of IBMK for 3 minutes, the aqueous phase was separated. The organic phase was treated with 5 ml each of benzene and saturated aluminium nitrate solution. This solution was further shaken for 1 minute then the aqueous phase rejected while the organic phase was subjected for the recovery of uranium (21).

5 ml of 0.5M hydrochloric acid solution was added into organic phase and uranium was stripped from it. This action was repeated for two times and the extract was collected in a beaker. Two drops of concentrated sulphuric acid were added and evaporated till the fumes of sulphur trioxide ceased. The heating was done on the hot plate and the residue was treated with 1 ml of water, 5 ml of concentrated hydrochloric acid and 2 g of zinc granules. The reduction was allowed to continue for about 5 minutes. The solution was filtered through the glass wool and washed thoroughly with 1:1 hydrochloric acid. The filtrate was collected in a 25 ml volumetric flask. Then 5 ml of 0.02% of Arsenazo (III) solution was added and the volume was made up to the mark with 1:1 hydrochloric acid. The solution was homogenized and the absorbance was measured in 10 mm cell at 665 nm against a reagent blank after 20-25 minutes. The concentration of uranium was estimated with reference to calibration curve.

3.3.5 Gross Radioactivity Measurement by MCA

0.5 kg of the finely powdered (particle size 125 µm) marble and granite samples was taken and gross radioactivity was measured by using Caesium as source.

4. RESULTS AND DISCUSSION

4.1 Adherence to Beer's Law and stability

The main source of uranium minerals in the earth crust is during the formation of earth. These uranium minerals are both physically and chemically weathered in areas with humid climates. Thus weathered samples were carried to the different area of the earth by various factors like air, water, wind and so on. The uranium has greater tendency to form soluble uranium complexes. These complexes which are formed in the environmental samples contain uranium in its hexavalent state (6). The efficiency of extraction of uranium was enhanced by reducing the hexavalent state of uranium into tetravalent uranium by using zinc metal in hydrochloric acid. Since the uranium (IV) reacted with Arsenazo (III), to give pink colored U (IV) Arsenazo (III) complex whose showed its maximum absorbance at wave length 665 nm. This value is found to match with the absorption spectra of U (IV) Arsenazo (III) complex reported(5). The spectrum obtained here is shown in Figure 4.1 below. The complex was not stable for a long period of time. It was found that the maximum absorbance was between 20-25 minutes and decreased gradually.

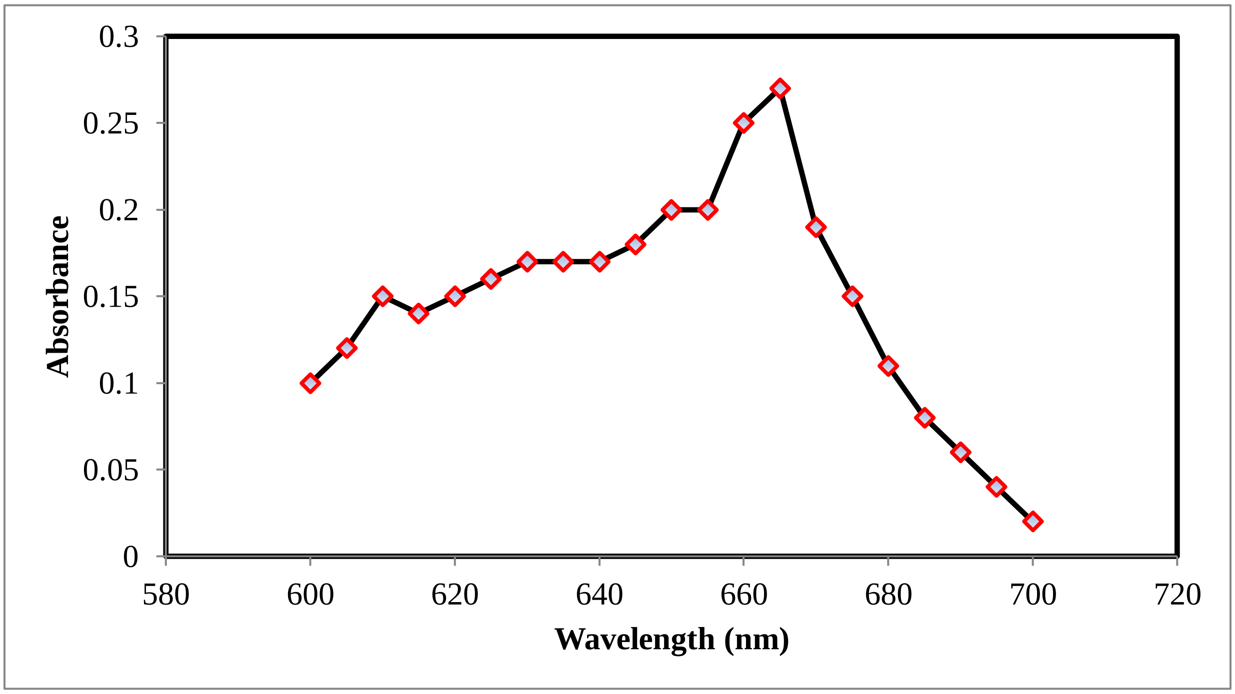

Fig4.1: Absorption spectra of Pink Colored Arsenazo (III) Complex

4.2 Calibration Curve for the determination of uranium

The Concentration of uranium in various building materials was determined using the calibration curve. Here, uranium (IV) Arsenazo (III) complex was prepared. The maximum absorbance of the colored solution was found to be 665 nm. The concentration is expressed in ppm.

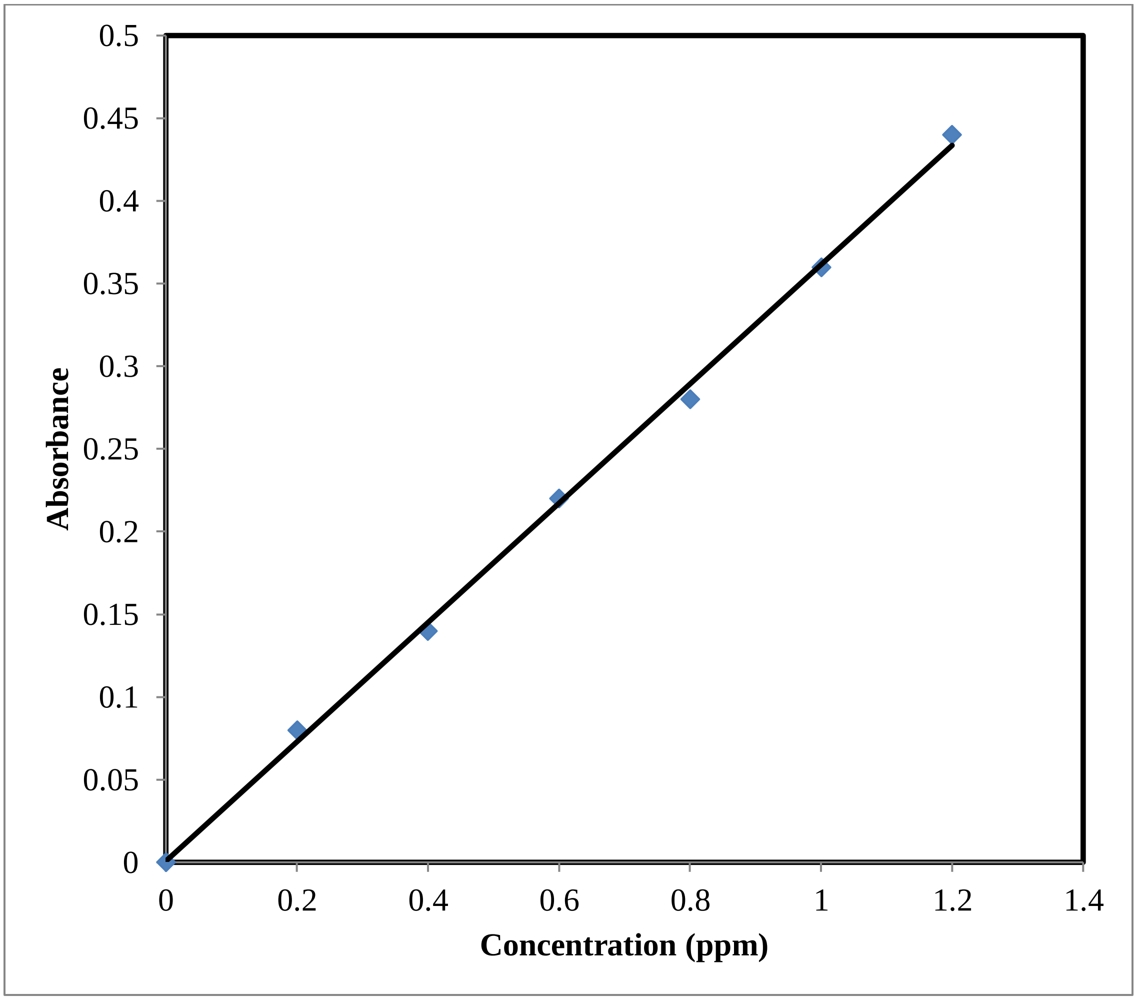

Fig. 4.2A: Calibration Curve for the Determination of Uranium.

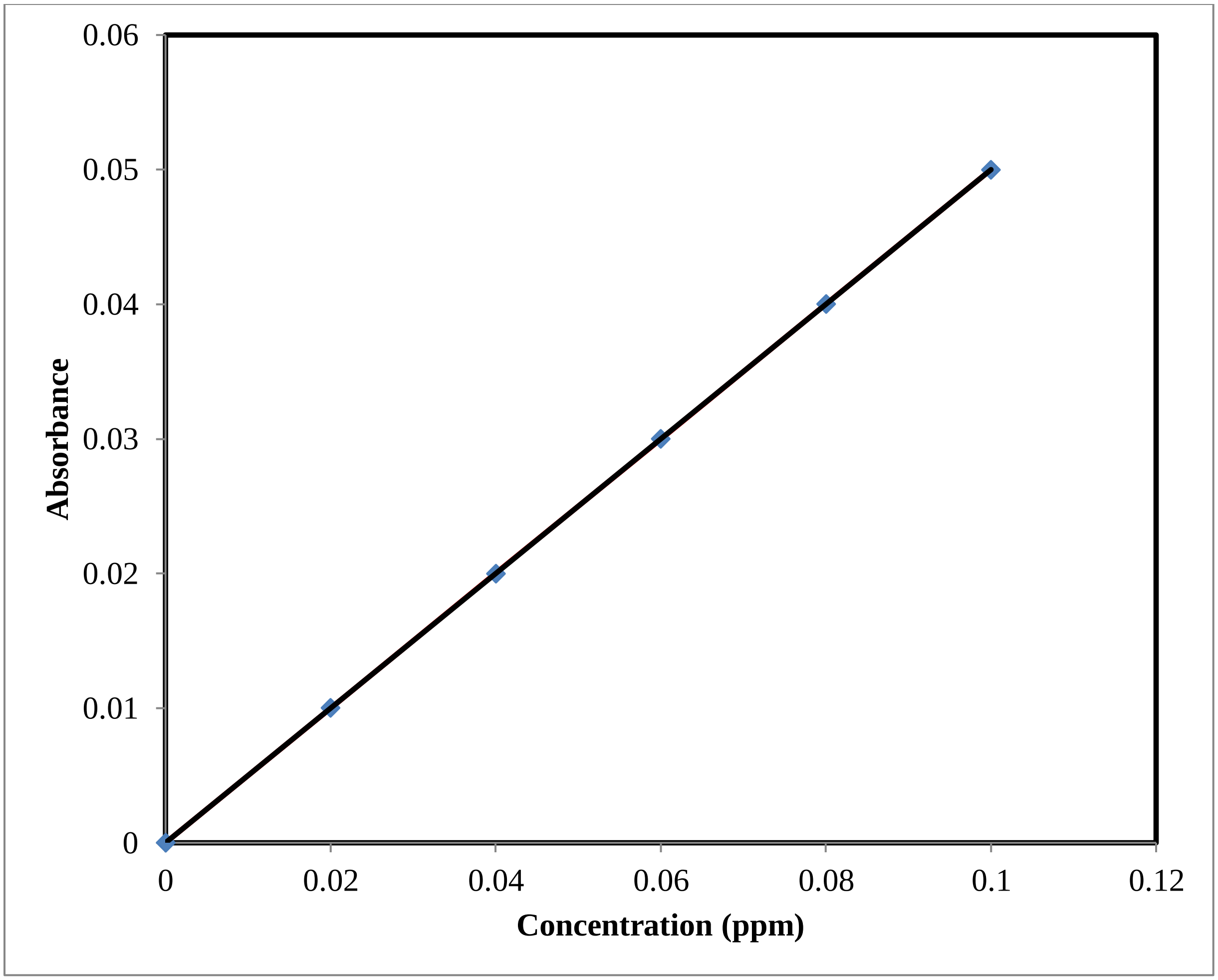

Fig. 4.2B: Calibration Curve for the Determination of Uranium.

4.3 Effect of amount of Aluminium nitrate on Extraction.

Uranium was extracted quantitatively in acidic medium using 6 M to 16 M nitric acid. Aluminium nitrate was used for extraction of uranium. The main role of aluminium nitrate was to make the solution more acidic. The efficiency of extraction was investigated by varying the amount of aluminium nitrate from 2 to 14 grams at pH 0.9 to 1.Table 4.2 shows the results. A plot of percentage extraction versus concentration of aluminium nitrate is shown in figure 4.4. This curve showed the uranium was quantitatively extracted into IBMK when amount of aluminium nitrate was 10 grams. If the concentration of aluminium nitrate was lowered, the extraction becomes less. At low concentration of aluminium nitrate, the solution was not acidic. The extraction of uranium remained unchanged on increasing aluminium nitrate and faced difficulty to dissolve more amount of aluminium nitrate in 5ml of sample solution (21).

Table 4.1: Effect of Al $(NO_3)_3$ concentration on extraction of uranium.

S.N.	Amount of Uranium (µgm)	Amount of $Al(NO_3)_3$ (gm)	% extraction
1	25	2.0	5
2	25	4.0	38
3	25	6.0	60
4	25	8.0	90
5	25	10.0	96
6	25	12.0	96
7	25	14.0	96

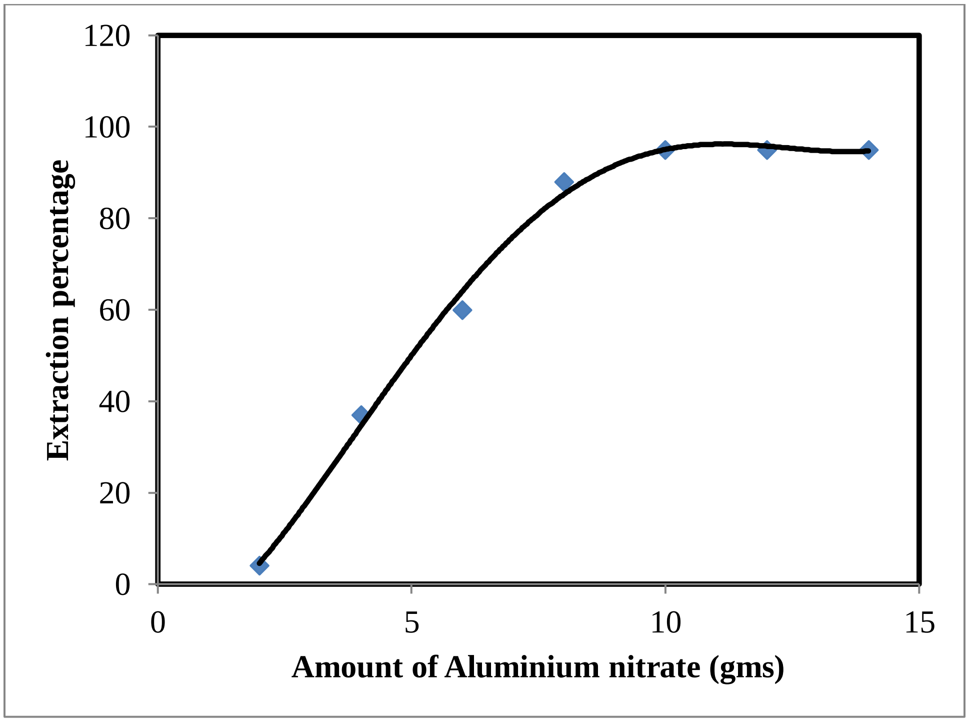

Fig4.3: Effect of Aluminium Nitrate on Extraction of Uranium

4.4 Effect of pH on extraction

On dissolving 10 gram of aluminium nitrate in 5 ml test solution, the pH decreased to 0.5. This pH value was adjusted to the order of 0.8 to 0.9 by using ammonium carbonate. The reduced pH was due to hydrolysis of aluminium nitrate. The variation of acidity was brought about by using ammonium carbonate. The table 4.2 shows the results. A plot of extraction percentage versus acidity (pH) values is shown in figure 4.4. This figure showed that extraction was quantitative at low pH values. On increasing the pH values, the percentage of extraction becomes less. At pH 2.5 and 3.0, only15 percent and 5 percent uranium was extracted respectively. At pH range of 0.5 to 1.5, the results showed quantitative extraction of uranium upto 98 percent.

Table 4.2 Effect of pH on Extraction of uranium

S.N.	Weight of Uranium (µgm)	pH adjusted	% extraction
1	25	0.5	98
2	25	0.8	98
3	25	0.9	98
4	25	1.0	95
5	25	1.5	90
6	25	2.0	60
7	25	2.5	15
8	25	3.0	5

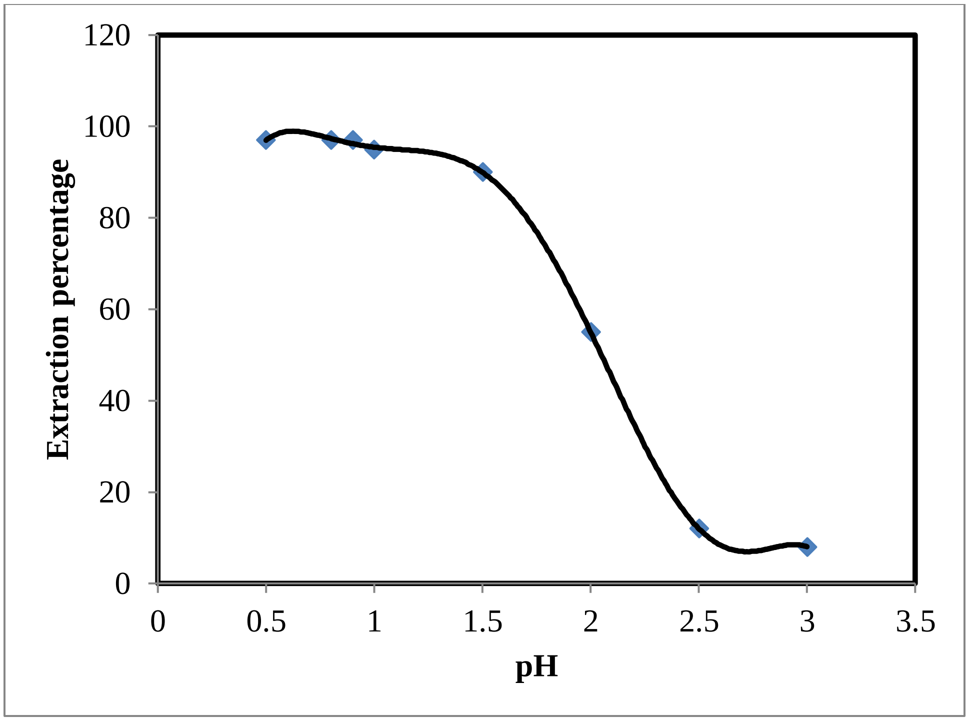

Fig4.4: Effect of pH on extraction of uranium.

4.5 Extraction of Uranium

The environmental uranium has greater tendency to form soluble complexes. These complexes which are found in the environmental samples contain uranium in its hexavalent state (6). The efficiency of extraction of uranium was enhanced by reducing the hexavalent state of uranium by using zinc metal in hydrochloric acid. One of the important factors affecting extraction of uranium is the pH. Here the pH was maintained around 1. This pH value was chosen because the extraction was significant at this condition. Extraction was conveniently brought about by dissolving 10g of aluminium nitrate $Al(NO_3)_3$ in 5 ml of the sample solution. The solubility of uranium salts in certain organic solvents was used to separate uranium from other metal ion by the solvent extraction process by the use of IBMK, diethyl ether and tributyl phosphate (TBP). These are natural extractants (2). The extraction is based on the formation of natural non-ionized complex according to the following reaction:

$$UO_2^{+2}\,(aq) + 2NO_3^{-}\,(aq) + 2TBP\,(org) \rightarrow UO_2\,(NO_3)_2.2TBP\,(org)$$

In fact the sample solution might contain thorium along with uranium. In this condition aluminium nitrate Al $(NO_3)_3$ at pH 1 was ineffective to extract thorium completely into aqueous phase. In order to bring the thorium selectively to the aqueous phase, benzene was used. Replacing the aqueous phase by 5 ml saturated aluminium nitrate Al $(NO_3)_3$ solutions and equilibrating for one minute with IBMK extract after the addition of 5 ml benzene increases the recovery of the uranium. A shaking time of 1 minute was found to be more efficient for quantitative extraction of uranium into IBMK. After equilibration in about 3 minutes with saturated aluminium nitrate and benzene did not affect the recovery of uranium and uranium was found in organic phase. The uranium present in the organic phase is stripped by using 0.1 $moll^{-1}$ hydrochloric acid. Thus obtained resulting solution was evaporated on the hot plate along with the few drops of concentrated sulphuric acid. The concentrated sulphuric acid helps in oxidizing the organic substances. The residue having uranium was then treated with 1 ml of water, 5ml of concentrated hydrochloric acid and 2 grams of Zinc acts as good reducing agent. By reducing U (VI) to U (IV), the determination of concentration of uranium in various samples greatly increased.

4.6 Efficiency of Ortec-Norland-5600 - MCA

The detector system coupled with 3”×3” NaI (Tl) crystal with 1024 channel is used for the process of measurement. Cs-137 (AMERSHAM, 74 KBq, manufactured on October 1988) was taken as standard source for calibration. The source was kept for set time 500 seconds within the lead shield above the crystal system.

Maximum peak observed at channel = 468

Region of Interest (ROI) = Lower limit: 234, Upper Limit: 702

Background =4500 counts in 500 seconds =9 cps

Actual Cs-137 counts in 500 seconds =1,410,000 = 2820 cps

Net counts =2820-9 = 2811 cps

We have,

N_o=74KBq (Given in source Tag)

Also, half life of Cs-137 = 30 years

Total time lapsed = 22 years (1988-2010)

Again, $N = N_o e^{-\lambda t}$

$= 74 \times e^{-0.693 \times 22/30}$

= 44.516 KBq

Efficiency (E_f) = Net Cs-137 count per second × 100/present activity in Bq

$= 2811 \times 100/44.516 \times 1000$

=6.31%

Activity of samples is calculated as follows:

Total activity of radionuclides in Bq/Kg (As) $=C_n/E_f \times V \times T$

Where, C_n = Net count or net activity of samples in cps

E_f = System efficiency

V = Volume or Weight of samples in litre or kg

T = Total duration

For example, for Red Kharda Granite

Background = 95 cps

Total activity = 122.5 cps

Net activity = 122.5-95 = 27.5cps

Total activity of radionuclide in Bq/kg (As) $=27.5 \times 100/6.31 \times 1$

= 871.63 Bq/Kg

The same procedure was repeated to calculate the activity of all other marble and granite samples. The total radioactivity measured in Bq/kg by the MCA for marble and granite samples were shown in table.

4.7 Sample Analysis

Natural radioactivity is wide spread in the earth's environment and it exists in various geological formations in soil, rocks, plants, water and air. The building materials like granite, marble are earth based, containing uranium in varying amounts which is one of the biggest inescapable sources of natural ionizing radiation for human exposure.

The spectrophotometric analysis for determination of uranium in the solution depends on the absorbance peak of uranium (IV) at 665 nm. First of all, 1 kg of each sample was taken and crushed into fine powdered form and sieved (particle size 125 µm). Thus formed finely powdered sample was taken and by standard procedure, as described earlier, a suitable weight (200 gm) was obtained. Then, 100 ml of distilled water was added into the sample. Few drops of concentrated HNO_3 were also added and stirred for half an hour. It was allowed for sedimentation. The solution was filtered. The sample was again washed with 100ml distilled water and filtered. Both the filtrates were mixed and dried to make about 5 ml. Then 10 gm of aluminium nitrate was added and stirred to dissolve. The pH of this solution was adjusted around 1.0 by using powdered ammonium carbonate and pH meter. The solution was quantitatively transferred to 100 ml separating funnel. Then 1 ml of saturated aluminium nitrate solution was used for each rinsing. The organic extractant named IBMK was used for extraction of uranium because of its low co-extraction percentage as well it did not bring down thorium and other metals to the aqueous phase completely. Benzene was chosen in order to return co-extracted metals to the aqueous phase. Equilibrium of phase after the addition of 5 ml of benzene, however, caused the decrease in the recovery of uranium. This was overcome by 5 ml of saturated aluminium nitrate solution. A shaking time of 1 minute was found to be sufficient for quantitative extraction of uranium in to IBMK. The uranium of organic phase was stripped using 0.1 M hydrochloric acid. The resulting solution was evaporated after adding two drops of concentrated sulphuric acid which helps in oxidation of the organic substances. The residue containing uranium was treated with 1 ml water and 5 ml concentrated hydrochloric acid acts as the best reducing agent. This reducing agent reduced the UO_2^{+2} to U (IV) as shown below:

$$UO_2^{+2} \xrightarrow{Zn/HCl} U(IV)$$

By using the Arsenazo (III) solution, U (IV) Arsenazo (III) complex was prepared and absorbance was measured at λ_{max} i.e. 665 nm. The concentration of uranium in different granite and marble was determined using calibration curve.

Table 4.3: Gross Radioactivity (Bq/kg) and Uranium concentration in different Marble Samples

S.N.	Samples	Origin	Gross radioactivity(Bq/kg)	Concentration of Uranium (ppm)
1.	Red Alikanta	Italy	613.31	0.04
2.	Siliviya	Egyptian	266.19	0.02
3.	Morwar White	India	557.15	0.04
4.	Dark Embryoder	Italy	644.268	0.06
5.	Dhermeta	India	596.23	0.04

Gross radioactivity of various marble samples was calculated (with 1 kg of sample) with the help of Multi Channel Analyzer (MCA) coupled with NaI (Tl) crystal detectors. The gross radioactivity of radionuclides among the samples varied from a minimum of 266.19 Bq/kg to the maximum of 644.268 Bq/kg. Also the chemical analysis showed that the concentration of uranium in the marble samples ranged from 0.02ppm to 0.06 ppm.

Table 4.4: Gross Radioactivity (Bq/kg) and Uranium concentration in different Granite Samples

S.N.	Samples	Origin	Gross radioactivity(Bq/kg)	Concentration of Uranium (ppm)
1.	Chima Red	China	994.24	0.08
2.	Red Kharda	India	871.63	0.08
3.	Rozy Pink	India	1127.32	0.12
4.	Cat Eyes	India	1052.87	0.10
5.	Green Pearl	India	1112.00	0.12

Gross radioactivity of various granite samples was calculated (with 1 kg of sample) with the help of Multi Channel Analyzer (MCA) coupled with NaI (Tl) crystal detectors. The gross radioactivity of radionuclides among the samples varied from a minimum of 871.63 Bq/kg to the maximum of 1127.32 Bq/kg. Also the chemical analysis showed that the concentration of uranium in the marble samples ranged from 0.08 ppm to 0.12 ppm.

It can be seen from the results that uranium concentration and gross radioactivity varied appreciably from sample to sample. It may be due to variation in the location of the raw materials from where they are derived and the percentage of constituents used by manufacturers as these parameters depends upon the distributions of naturally occurring radionuclides present in the earth crust since these radionuclides are not uniformly distributed. Here, the granite contained more uranium than marble. These results are in tune with Saleh et al. (40). All rocks have a small amount of radioactivity in them due to the presence of minerals that contain radioactive elements uranium (U), thorium (Th) and potassium-40 (K40). Because granite typically contains more of these elements than most other rocks, it will be more radioactive than a marble. All of the minerals in granite contain some radio elements; the white or pink feldspars contain K40, the black biotites and horn-blendes contain K40, U and Th, and the small inclusions of minerals such as zircon, apatite, sphene, etc. contain the most U and Th (11,12,43).

5. CONCLUSION

The physical method includes gross radioactivity in marble and granite samples by using the MCA. The gross radioactivity in the marble samples was found to be in the range 266.19 Bq/kg - 644.268 Bq/kg with maximum gross radioactivity for the “Dark embryoder”. However in case of granite, the gross radioactivity was found to be in the range 871.63 Bq/kg - 1127.32 Bq/kg with maximum gross radioactivity was for the “Rozy pink”.

The spectrophotometric determination of uranium in different marble and granite samples by was carried out by forming a complex of Arsenazo (III) with uranium in highly acidic medium. The soluble uranium complexes are found in the environmental samples with having uranium in its hexavalent state. The efficiency of the extraction was enhanced by reducing the hexavalent state of uranium into tetravalent uranium by using zinc metal in hydrochloric acid. For the extraction the use of aluminium carbonate to adjust the pH was found to be beneficial. IBMK used as organic solvent and benzene used to increase the efficiency of extraction were helpful.

The determination of uranium by this method was extremely sensitive, rapid and cheaper and can be used in the assay of ppm level of uranium. The concentration of uranium in marble samples was found in the range of 0.02ppm to 0.06 ppm. From the table, the “Siliviya” marble had the maximum uranium concentration which was found to be 0.06 ppm. Similarly, the concentrations of uranium in granite samples were found in between 0.08ppm to 0.12 ppm. The “Rozy Pink” and “Green Pearl” granite had the maximum concentration which was found to be 0.12 ppm. Furthermore, the granite has greater radioactivity than marble. Similarly, uranium concentration was also found greater in granite than that of marble.

6. SUGGESTON FOR FURTHER WORK

a) Radiochemical analysis of other construction materials like cement, sand, water, lime, bricks etc. needs to be carried out.

b) Exploration of uranium minerals needs to be done throughout the nation and areas with high content of uranium need to be specified.

c) Radon should be measured inside houses.

d) Correlation of environmental radioactivity with human health is essential and such studies need to be conducted.

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8. APPENDICES

APPENDIX I: Physical and chemical properties of uranium (1)

Property	Value
Symbol	U
Color	Silvery
Phase at Room Temp.	Solid
Density(g/cm^3)	19.05
Melting Point(K)	1405.2
Boiling Point (K)	4203
Heat of Fusion(KJ/mol)	12.6
Heat of Vaporization (KJ/mol)	417
Heat of Atomization(KJ/mol)	490
Thermal Conductivity (J/m sec K)	27.5
Electrical Conductivity (ohm^{-1} cm^{-1})	33.333
First Ionization Energy (KJ/mol)	584
Electronegativity	1.7
Atomic Weight	238.03
Atomic Volume (cm^3/mol)	12.5
Atomic Radius (pm)	156
Common Oxidation Number	+6
Other Oxidation Numbers	+3,+4,+5

APPENDIX II: Chemical Assay Values

Urany1 Nitrate $UO_2 (NO_3).6H_2O$	**LobaChemi**
Assay $UO_2(NO_3).6H_2O$	98.0-102.0%
Chlorine (Cl)	max.0.002%
Heavy metals	max.(Pb)0.002%
Insoluble matter	max.0.005%
Iron (Fe)	max.0.002%
Permanganet	max.0.6%
Sulphate	max.0.005%

IBMK $(CH_3)_2CHCH_2COCH_3$	**Qualigens**
Minimum assay	98%
Refractive index	1.3965
Free acid	1%
Water	0.5%
Wt. per ml at 20^0C	0.79-0.802g

Ammonium Carbonate NH_4CO_3	**Merck**
Assay (NH_3)	30%
Chloride	0.002%
Sulphate	0.005%
Heavy metals (as Pb)	0.002%
Iron (Fe)	0.001%

Calcium (Ca)	0.02%
Aluminium Nitrate Al $(NO_3)_3.9H_2O$	**Merck**
Assay	98.5%
Chlorine (Cl)	max. 0.001%
Sulphate (SO_4)	max.0.005%
Lead (Pb)	max.0.01%
Iron (Fe)	max. 0.002%
Calcium (Ca)	max. 0.005%
Sodium (Na)	max. 0.005%
Potassium (K)	max. 0.002%
Arsenazo(III)	**Loba Chemie**
Hydrochloric Acid	**Qualigens**
Assay (acidimetric)	35.37%
Wt. per ml at 20^0C	1.18g
Non-volatile matter	0.01%
Sulphuric acid	0.02%
Arsenic (As)	0.0001%
Iron (Fe)	0.0005%
Lead (Pb)	0.0005%
Free chloride	0.0005%

Sulphuric Acid	**Qualigens**
Assay (acidimetric)	97.99%
Wt. per ml at 20^0C	about 1.835g
Maximum limits of impurities	
Non- volatile matter	0.01%
Hydrochloric acid	0.0005%
Nitric acid	0.001%
Arsenic (As)	0.0002%
Iron (Fe)	0.002%
Reducing substances	0.02ml N/1%

Benzene Extra Pure	**S.D. Fine Chem. Ltd.**
Assay (GLC)	99.0%
Boiling range	min. 95%
Freezing point	not below 5.2^0C
Wt. per ml at 20^0C	0.875-0.879g
Maximum limit of impurities	
Non- volatile matter	0.002%
Water	0.1%

Appendix III: Units of Radioactivity

The activity of a radioactive material is measured in term of the disintegrations per unit time. Its SI unit is Becquerel (Bq), which is one disintegration per second.

i.e., 1Bq= 1 disintegration / second

The higher and the old unit of measurement radioactivity was Curie (Ci).

i.e., 1Ci= 3.7×10^{10} disintegration / second

Activity per unit mass is specific activity. The effect caused by radioactive isotopes to the biological system is measured in the units as expressed below.

For Dose equivalent:

Sievert (Sv)

i.e., 1Sv = 100 rem

1mSv = 100 mrem

For Absorbed Dose:

Gray (Gy)

i.e., 1 Gy = 100 rad

1 mGy = 1000 mrad

For activity:

1 Bq = 1 Disintegration / second

1 Bq = 2.7×10^{-11}Ci = 27 pCi

1 KBq = 100 rad

For Dose rate:

1 Gy = 100 rad

1 Sv = 100 rem 1 μSv = 0.1m rad = 10^{-4} rad

1 n Gy = $10^{-7} \times 1/10^{-4}$μSv = 10^{3} μSv

1 n Gy = $100 \times 10^{-9} = 10^{7}$ rad

1 mSv = 100 m rem