

URANIUM AND ITS MEASUREMENT IN ORE
BY RADIOMETRIC METHOD

By

A. K. SARANGI

Additional Superintendent (Geology)

URANIUM CORPORATION OF INDIA LTD.

The element uranium is very widely distributed through out the crust of the earth. Almost all type of rocks contains at least some amount of uranium. Natural waters including sea water also contain uranium in very small quantity. It is estimated that the average concentration of uranium in earth's crust is about 3gms per tonne of rock and in sea water, it is about 1gm per thousand tonnes. About 1.3×10^{14} tonnes of uranium exist in the earth's crust.

The element uranium with Atomic number 92 is positioned last in the periodic table amongst naturally occurring elements. It is radioactive and its atomic weight is 238. Uranium is known to occur in three isotopic forms in nature. Their approximate relative natural abundance is as follows.

U^{238} – 99.27%, U^{235} – 0.724%, and U^{234} – 0.006%

The proportion of these isotopes is constant in nature regardless of the type of mineral or deposit in which the uranium is found. (Another isotopic form of uranium is U^{233} , which is artificially produced in nuclear reactors.)

Uranium does not occur in native state. It has affinity for oxygen and occurs in nature mostly as oxides, phosphates, sulphates, vandates, arsenates, carbonates and silicates.

There are about more than 100 uranium bearing minerals known. But of these, only a few occur in economic concentration for commercial purpose. Uraninite, pitchblende and davidite are some of the primary uranium minerals. These minerals are refractory in nature. Naturally occurring some common secondary uranium minerals are autonite and meta-autonite, torbernite and meta-torbernite, carnotite, uranophane etc. These minerals are normally bright and occur in different colour shades.

In nature, uranium ions occur normally in 4 and 6 oxidation states. Sometimes the 5 oxidation state does occur in some environments but it is probably transitory. In hexavalent state, uranium is easily soluble in water.

Published in “Journal Mines, Metals and Fuels, Annual Review; 2000”.

Uranium metal is shiny, silvery, ductile and slightly paramagnetic. Chemically, metallic uranium is highly reactive. It dissolves in acids upon moderate heating and reacts with water at about 100⁰C.

ESTIMATION OF URANIUM:

Various methods / techniques, both physical and chemical are available for the determination of uranium content in rock / ore. During prospecting and / or exploration stage of uranium bearing minerals, it is necessary to make a quick but reliable estimate of uranium content in the rock. The properties of uranium described above are the basis for recognition, separation and determination of uranium. But for the purpose of field measurement, the most useful property of uranium is its radioactivity.

Radioactive property of uranium: Radioactivity is a spontaneous and self- destructive nuclear activity leading to the break-up of the element. It is spontaneous in the sense that it is intrinsic and is unaffected by external agents, either physical or chemical. All radioactive substances in nature undergo this disintegration / decaying process emitting three distinct types of radioactive rays and resulting in the formation of new elements. The new element thus formed, if radioactive, disintegrates again to form another element. This process continues covering a few elements until a stable end product is formed.

The emitted rays are designated as alpha(α), beta(β) and gamma(γ). Alpha rays are ionised helium particles having a positive charge of two units with a mass number of 4. Beta particles are electrons having a unit negative charge and are very light. Gamma rays are electromagnetic radiation of very high frequency, similar in nature to light or x-rays. α and β rays have very limited penetrating capacity. Even a couple of mm thick aluminum foil can completely arrest the free movement of these rays. γ rays, on the other hand are highly penetrative. They travel in straight lines in vacuum and the intensity follows the inverse square law ($I \propto I_0 / r^2$, where I is the intensity of gamma rays at a distance of r from the source and I_0 is the intensity at the point of origin). All the three rays are not transmitted simultaneously from any one substance.

Normally, the detecting devices using γ rays are most commonly used in the field because of their simplicity, ruggedness, sensitivity and relative low cost. Devices based on α and β detections are also available mostly used for laboratory purpose.

Uranium being a radioactive element emits radioactive rays, and in the process, decays into a series of successive daughter products finally reaching a stable end product – lead isotope (Pb^{206}). This activity is not instantaneous. It prolongs over a certain period of time, characteristic of the element.

The whole decay series of uranium is given below which is also known as **uranium-radium decay series**.

Table - 1

ELEMENT / ISOTOPE	EMISSION	HALF LIFE	GROUP
U^{238}	α	4.5×10^9 Years	URANIUM GROUP
Th^{234}	β, γ	24.1 days	
Pa^{234}	β, γ	1.18 min	
U^{234}	α	2.6×10^5 years	
Th^{230}	α, γ	8×10^4 years	
Ra^{226}	α, γ	1580 years	RADIIUM GROUP
Rn^{222}	α, γ	3.82 days	
Po^{218}	α	3.05 min	
Pb^{214}	β, γ	26.8 min	
Bi^{214}	β, γ	19.7 min	
Po^{214}	α	164 micro sec	
Pb^{210}	β, γ	22 years	
Bi^{210}	β	5 days	
Po^{210}	α	138.3 days	
Pb^{206}	Stable		

As can be seen from table-1, not all decay products of the entire series produce gamma rays. From the point of view of gamma emission, the whole series is divided into two groups.

- a) Uranium group and
- b) Radium group.

Considering the energy levels of gamma radiation, it has been determined that the uranium group members emit about 2% of total gamma while the remaining 98%

Published in “Journal Mines, Metals and Fuels, Annual Review; 2000”.

gamma come from radium group members. Therefore for all practical purposes, the measurement of intensity of gamma rays is the measurement of content of radium group of elements. However, the gamma measurements do indicate the uranium content, if the uranium series is in radioactive equilibrium.

Radioactive equilibrium / disequilibrium:- As explained above, the process of successive disintegration of radioactive substance continues till a stable end product is formed. But when the parent source and the daughter products in a sequence are radioactive, the growth of successive daughter products, their emission and their activity levels become very complex. At the initial stage, the successive product growth is a function of the decay of the parent. As the time becomes great, the daughter product activities approach that of the slow decaying parent and ultimately, a stage comes when the number of daughter nuclei produced per unit time by its parent becomes equal to the number of daughter nuclei disintegrating at the same time. It can be expressed as a steady-state condition where the rate of formation of daughter products is same as rate of disintegration of daughter products. This stage is known as secular equilibrium. In a closed system, all the members of a radioactive family eventually reach this stage and at this state, it is said that the radioactive series is in equilibrium. The large difference between the half-life periods of the parent and daughter element plays a prominent role in achieving equilibrium. In case of uranium series, the parent element U^{238} is the longest lived isotope (has a very long half-life period, equivalent to the age of earth) for which the process of disintegration has been continuing for a very long time and therefore, the stage of equilibrium is achievable. If this condition has been achieved for any ore assemblage or the deposit, then the counting / measure of gamma emission gives a very reliable estimate of uranium.

But there are some natural events which may occur during this long life period of parent element affecting the above mentioned steady-state condition. Let us consider the following two cases.

a) If the parent element uranium, during its long geologic history is subjected to oxidising environment, it changes to higher valency state (hexavalent) in which it is easily soluble in even a weakly acidic or alkaline water and is carried away in solution. It may happen to any of the long-lived daughter nuclei of the series also. As a result, the equilibrium in the whole series gets disturbed. Emission of gamma ray from the

Published in “Journal Mines, Metals and Fuels, Annual Review; 2000”.

daughter products will be in excess of the parent element uranium. In such cases, gamma ray measurement will indicate the presence of uranium, which will be more than that is actually present.

b) Sometimes the gaseous daughter products (mainly radon which is highly mobile and has a comparatively long half-life) escape out from the series disturbing the equilibrium. In this case, gamma measurement would indicate lesser amount of uranium than is really the case.

The above two cases show the conditions for disequilibrium. In case (I) the disequilibrium is said to be in favour of daughters whereas in case (II), it is in favour of parent i.e. uranium. Presence of thorium and potassium in uranium ore also affect the stage of equilibrium.

Generally, it has been seen that very old rocks containing primary uranium free from thorium and potassium, and not subjected to agents of natural denudation or chemical separation continue to remain in equilibrium.

For routine analysis, the physical method of gamma measurement is extremely useful. The only uncertainty is whether uranium is in the stage of secular equilibrium or not. Therefore, at the time of exploration, it is necessary to establish whether the ore is in equilibrium or disequilibrium. Since there are more than two factors to affect the equilibrium, any single measurement, whether based on alpha, beta or gamma count, would not suffice and this has to be supplemented by other measurements depending on the type of information needed. A simultaneous measurement of beta and gamma radiation from the sample is useful for this purpose. In order to establish the thorium and / or potassium factor, another independent measurement is also required. It is normally done by various methods of physical measurements and simultaneous chemical analysis. If the deposit is found to be in equilibrium, physical method of determination using gamma rays can be used without any doubt. In such measurements, the content of uranium computed is designated as % U_3O_8 equivalent or eU_3O_8 rather than U_3O_8 as determined by chemical analysis. Thus eU_3O_8 could be less than, equal to or more than U_3O_8 depending on the state of equilibrium of the series.

If the deposit is in disequilibrium, it is necessary to quantify the disequilibrium (direction and magnitude). The physical method of gamma measurements can then be applied with suitable corrections.

Instruments used for radiometric measurement (γ ray): - The basic philosophy of designing the radiation detective devices are

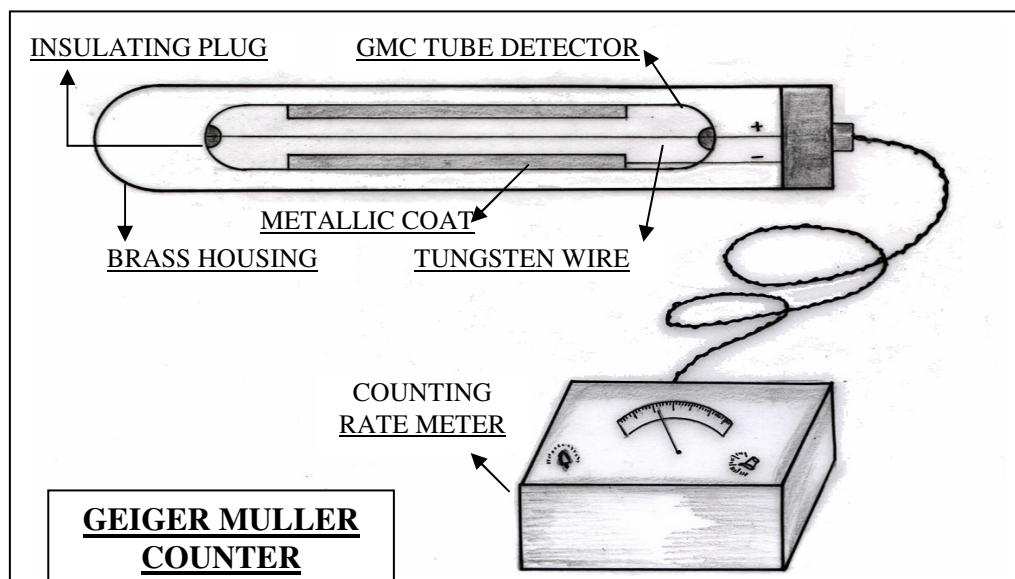
- i) to detect the incoming radiation by a radiation detector and
- ii) to amplify and record the electrical signals through some electronic devices.

Accordingly, the device consists of two essential parts - a) a radiation detector and b) an amplifier. For normal gamma measurement purpose in the field, two commonly used counters are

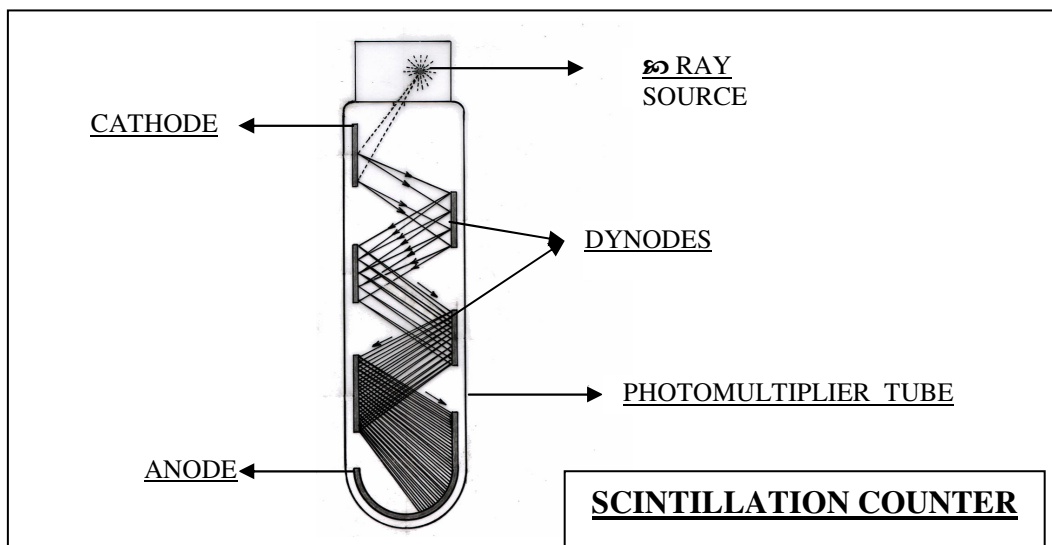
- i) Geiger Muller counter and
- ii) Scintillation counter

GEIGER MULLER Counter – The assembly consists of a Geiger Muller tube detector filled with a mixture of argon and ethyl alcohol at low pressure and enclosed in a moisture proof cylindrical housing. The interior is coated with iron or tungsten to act as the cathode. A thin metallic wire runs at the center fitted with insulating plugs at both ends of the tube that serves as the anode. The detector is connected to a composite count-rate meter with the provision for built-in high voltage power supply unit necessary for the detector and suitable electronic circuits to average out the detected signals.

The gamma radiation emitted by the source surrounding the detector are intercepted by the cathode and the resulting interaction produces ionization in the tube resulting electric signals. The signals are amplified by suitable electronic devices and are recorded. The converted electrical signals are read on the count rate meter which indicate the intensity of radiation in terms of current.



SCINTILLATION Counter – This consists of a thallium activated sodium iodide crystal called scintillator and an electronic device called photo-multiplier tube. Whenever gamma (γ) rays pass through this crystal, a part of the energy of the ray is absorbed in the crystal. The intensity of scintillation is proportional to the energy of the incoming radiation. The absorbed energy produces excitation in the crystal resulting in small specks of light called scintillations. These are picked up by photo-cathode of the photo multiplier tube. In the tube, there are several dynodes between cathode and anode and these are maintained at progressively high voltage. The scintillations on photocathode emit more scintillations towards the dynode and the process is accelerated with successive dynodes, thus sufficiently amplifying the signals. The amplified signals are shaped to equal amplitude pulses by means of electronic circuits. These signals are counted by scalar or are converted to current for measurement by a counting rate meter.



Compared to Geiger counter, the sensitivity of scintillation counter is much higher for detecting the rays.

Depending on the purpose and the working environment, the above two counters are modified with suitable attachments to meet the requirement. The modified instruments are calibrated against known standards.

Published in “Journal Mines, Metals and Fuels, Annual Review; 2000”.

INDIAN URANIUM DEPOSITS IN EQUILIBRIUM / DISEQUILIBRIUM: - Bulk of world's known uranium reserves are in sedimentary rocks and they are in disequilibrium. A sizeable quantity of India's uranium ore reserve lies in metasedimentary rocks of Singhbhum Thrust Belt, Bihar. All the uranium occurrences in this region including the three producing mines – Jaduguda, Bhatin and Narwapahar are in the state of equilibrium. Some other occurrences of uranium found elsewhere in the country namely, Bodal in Madhya Pradesh, Domiasiat in Meghalaya, Lambapur in Andhra Pradesh and Gogi in Karnataka etc are reported to be in disequilibrium.

ACKNOWLEDGEMENTS:

The author acknowledges the encouragement given by the Chairman & Managing Director, UCIL to prepare this paper and the kind permission given by him to publish it.

REFERENCES:

1. Evans Robley, D., The Atomic Nucleus, McGraw Hill Book Comp, USA, 1955
2. IAEA Technical Report Series, Measurement and calculation of radon releases from Uranium mill tailings, No. 333, 1992.
3. IAEA Technical Report Series, Analytical techniques in uranium exploration and ore processing, No.341, 1992.
4. Thompkins, R.W., Radiation in uranium mines, CIM Bulletin, Sept 1982, P. 149.
