

Uranium Occurrences in Coaly Meta-Pelites in the Jinsan Area

Se-Jung Chi*, Se-Hyun Kim* and Pyeong-Koo Lee*

Abstract: To understand the characteristics of uranium distribution, and the correlation of the uranium content and major constituents in uraniferous black slates from the Jinsan area of Ogcheon Fold Terrain, forty representative specimens were analyzed by mineralogical and radiometric techniques. According to statistical analysis, the uranium has a positive correlation with organic carbon and limonite, but a negative relation to muscovite and other opaques. The relationship with the highest and meaningful correlation is between log uranium and organic carbon. The log uranium-organic carbon correlation coefficient is 0.845 and these two constituents have about 71.4% association. It suggests that the abundance of organic carbon controlled the uranium precipitation. The relationship of organic carbon to log uranium can be expressed by following regression equation $\log (U_3O_8 \times 10^4 + 1) = -1.3447 + 2.5599 \log (\text{organic carbon})$. The multiple regression equation of different major components to log uranium is $\log (U_3O_8 \times 10^4 + 1) = 0.77396 + 0.04465 (\text{organic carbon}) + 0.00574 (\text{quartz}) - 0.00964 (\text{muscovite}) + 0.37827 (\text{biotite}) - 0.02286 (\text{clay substance}) + 0.01268 (\text{other silicates}) + 0.1032 (\text{barite}) - 0.00224 (\text{apatite}) + 0.01606 (\text{calcite}) + 0.08258 (\text{hematite}) - 0.02406 (\text{limonite}) - 0.01715 (\text{other opaques})$.

INTRODUCTION

It is well known that uranium was precipitated from uraniferous ground water by adsorption of organic matter under reducing condition. According to Kazumi, et al. (1975) and Hirano (unpub.) poorly coalified organic matter has the highest affinity for uranium fixation from uraniferous solutions over wide pH range.

The Jinsan area is located in the middle part of Ogcheon Fold Terrain and is comprised of the Ogcheon Group, Jurassic biotite granite and Cretaceous igneous rocks. The uranium-bearing black slates and phyllites occur mainly within the Changri Formation and Munjuri Formation of the Ogcheon Group. This Changri Formation has a coal bed with high uranium content in the studied area.

Some petrographic, geochemical and field studies of the uranium-bearing meta-pelites in the Ogcheon Fold Terrain have been previously carried out by So and Kang (1978) and So and Choi (1984).

The purpose of the present research was to make a detailed quantitative mineralogical and geochemical study of the uraniferous meta-pelites in the Jinsan area in an effort to increase understanding of the nature of these fine-grained rocks and to determine the location and specific associations of the uranium. The correlation of uranium to the major mineral constituents was studied.

SAMPLING AND TREATMENT

By using the UV lamp and portable radiological survey meter, specimens were selected to obtain materials representative of the uraniferous beds from the Jinsan area and to minimize the effect of weathering on mineralogical and

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chemical composition.

After uranium content of all specimens was examined by a Decade Scalar model 181B and a Scintillation Counter XIB Beta Crystal, 40 samples, weighing 1~2kg each, were taken for this study. Sampling localities are shown in Fig. 1. These samples were prepared for thin-section by slicing the rocks across the foliation, and counterparts were powdered (100 mesh) to analyze uranium and organic carbon content. Before producing thin-sections, consolidatory work was necessary in order not to obviate fine-grained mineral constituents with low coherence.

Modal analysis of at least 1,000 counts each were performed with a Swift automatic point counter on unstained thin sections. A counting intervals of $1/10 \times 2\text{mm}$ was used. Uranium composition was determined by comparison with a standard by using a spectrophotometer, and organic carbon was analyzed with a C-H-N Corder (YANAGIMOTOMT-2-model).

The data obtained are quantitative information comparing the mineral composition and texture of the samples to the distribution of organic carbon and to the concentration and distribution of uranium. The evaluation of the above quantitative data has been accomplished by the application of multivariate statistical analysis using an IBM 370 (model 115) computer.

The phases of the quantitative investigation are organic carbon, quartz, muscovite, biotite, clay substance, other silicates (tourmaline, sphene, zircon, chlorite, sericite, chloritoid, garnet), barite, apatite, calcite, hematite, limonite and other opaques. The statistical analysis of the data discussed in this paper deals with the relationship of these minerals with uranium content.

GENERAL GEOLOGY OF THE AREA

The area studied include the middle part of the Ogcheon Fold Terrain where the metasedi-

ments, Ogcheon Group, were deposited in an elongate basin north-eastwards across the Korea peninsula from Samcheog to Mogpo. The geotectonic lineation of the geology of the area is parallel to the Sinian direction.

The original shales, mudstones, siltstones, sandstones, dolomites, limestones, pebbly mudstones, basic tuffs and lavas which made up the Ogcheon Group, were intensely folded and variably metamorphosed during the Daebo Orogeny of the Jurassic period. The grade of metamorphism varies from greenschist to amphibolite facies. The Ogcheon Fold Terrain can be divided into two major zones; a metamorphic zone in southwest and a non-metamorphic zone in the northeast (Fig. 1). In addition to the contrasts in their stratigraphy and metamorphic grade, the two zones display differences in their structure. The metamorphic zone is characterized by similar major folds and ubiquitous penetrative minor structures, such as axial plane and crenulation foliation and a variety of types of tectonic lineation, all resulting from intense plastic deformation of the strata. The non-metamorphic zone is also highly folded but concentric folds are more common than in the metamorphic zone (Reedman, et al. 1976).

The Jinsan area belongs to the metamorphic zone, showing low metamorphic grade, and is comprised of the Ogcheon Group intruded by Jurassic biotite granite and Cretaceous intrusives. Their geologic distribution and sequence are shown in Figure 1. The Majeonri Formation is distributed chiefly in the southeast of the area. It appears to be the stratigraphically lowest unit in the Group. The formation consists of banded, crystalline limestone and limesilicates. The Changri Formation is widespread in most of the area. It conformably overlies the Majeonri Formation and is composed mainly of black slate, schist, phyllite, with minor limestones

and coal beds. Thin, low-grade, uranium-bearing coaly beds are locally interbedded in this formation. The Munjuri Formation is distributed in the area on a small scale. The formation overlies the Changri Formation

conformably and contains chiefly quartz biotite schist and sandy phyllite. The Odaesan Quartzite Formation is distributed in the southwest of the area. The formation overlies the Munjuri Formation conformably and consists of quartzite,

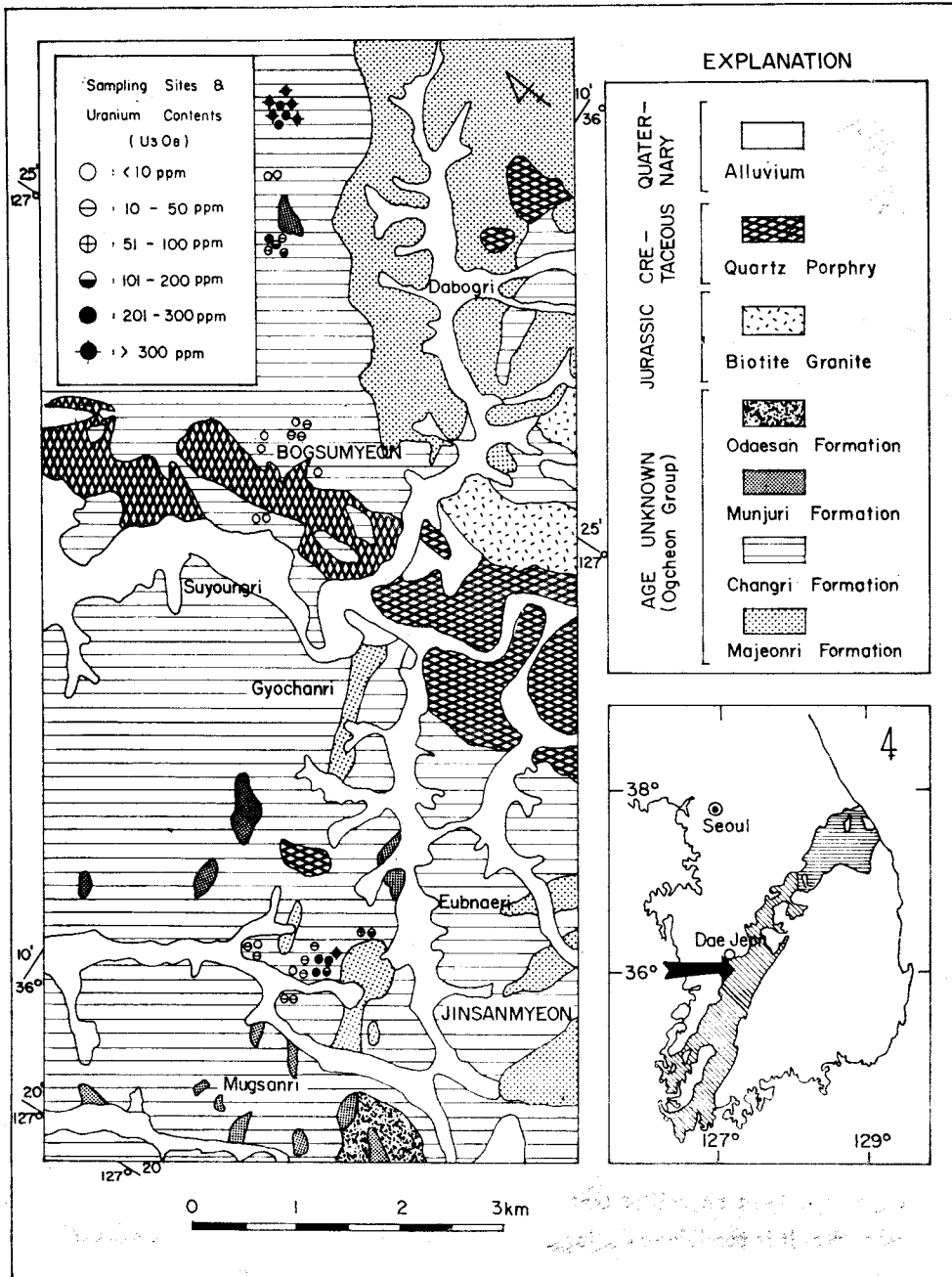


Fig. 1 Geologic map of the studied area, showing the Ogcheon Fold Belt (inset), sample localities, and uranium contents.

quartz schist, and conglomeratic quartzite. The intrusives consist of Jurassic biotite granite and Cretaceous quartz-porphry.

MINERALOGY AND URANIUM OCCURRENCE

The specimens studied at high magnification (10×40) are qualitatively different. The average of the main constituents are (in order of decreasing average vol. %); coal, 61.24; quartz, 21.60; muscovite, 7.62; followed by accessory minerals (in vol. %); other opaques (pyrite, ilmenite etc.), 1.72; limonite, 1.54; apatite, 1.09; barite, 0.82; calcite, 0.75; hematite, 0.61; chloritoid, 0.41; clay substance, 0.31; tourmaline, 0.15; biotite, 0.12; sphene, 0.08; zircon, 0.08; sericite, 0.03; garnet, 0.02; chlorite, 0.01. Average organic carbon and uranium contents analyzed are 17.21wt. % and 121.49ppm respectively. The organic materials are very important constituent of the studied specimens. They occur microscopically as matrix and mostly as masses of small black particles and coatings on the other mineral constituents. Most quartz grains held together by coal are poorly elongated and have slight wavy extinction. Many small metamorphic quartz veinlets let in irregular fractures of coal materials. Fine grains of micas show intensive minor folding. Apatite occurs as xenomorphic to hypidiomorphic, colorless grains. Calcite occurs as aggregates or often form veinlets. Xenomorphic barite is closely associated with quartz veins. Tourmaline and zircon occur as hypidiomorphic grains. Iron oxides (mainly hematite and limonite) are associated commonly with the coal material, either as a complete or partial rims around their outer edges and have an oolitic texture.

In the Jinsan area, it is possible to distinguish the uranium minerals only by fluorescent color under a UV lamp, because uranium minerals occur as a few very fine grains. The micaceous

uranium mineral showing cleavage in two directions in thin-section are pale, yellowish green colored, meta-uranocircite, which shows a strong yellowish green fluorescent color. Green minute crystals of torbernite also occur (So, 1976a, b).

The uranium minerals are mainly held in interstitial positions in the fine-grained organic matrix, whereas some of them occur often as fine grains in irregular fractures within the organic matrix and appears to be distributed randomly along structural plane of slates. In the studied area, all the uranium bearing black slates contain 5 to 400ppm uranium, though some portions contain as much as >300 ppm (Fig. 1). Highly uraniferous black slates contain large amount of organic matters.

The strong relation between uranium and organic matter in the black slates indicates that organisms in some way may have been instrumental in removing uranium from the sea water and concentrating it. Adsorption of uranium from a uranium-bearing solution in a host rock is controlled primarily by factors such as pH, Eh, temperature, activities of U^{+6} , U^{+4} and other cations and anions (Haung, 1978). Potential adsorbers commonly associated with the uranium deposits are clay minerals, organic carbonaceous matter, and zeolites. Organic matter is effective not only as adsorbent but also as reducing agent.

Uranium can be leached from source rocks when either alkaline solution or acidic solution moves through the permeable host rock. Mobility and accumulation of uranium are controlled by physical factors. The physical factors include the viscosity of the transporting fluid, the permeability of the medium, potential differentials, and temperature of the uranium-bearing solutions in the micro-environment (Haung, 1978).

At the surface, only that uranium reached by

leaching or eroding meteoric water is liberated. However, as the amount of uplift deformation, compression, and heating of rocks increases, the amounts of uranium mobilized also increase (Gabelman, 1977). As regional metamorphism intensifies, increasingly more corrosive fluids are released. As temperature and rock plasticity increases, these fluids are increasingly able to penetrate mineral lattices to oxidize and mobilize the more mobile elements such as uranium. Some uranium minerals developed within structural plane of coaly slates in the studied area is evidence that uranium was dissolved in ground water, and then was fixed in the minerals in various ways during regional metamorphism.

CONSIDERATION

The concentrations of uranium, organic carbon and various minerals are not the same in the various black slates, though mineral compositions are similar qualitatively. Since the geochemical distribution of uranium in the black slates is nearly log normal (So and Kang, 1978), the highest and most meaningful correlations are between each of the individual components and

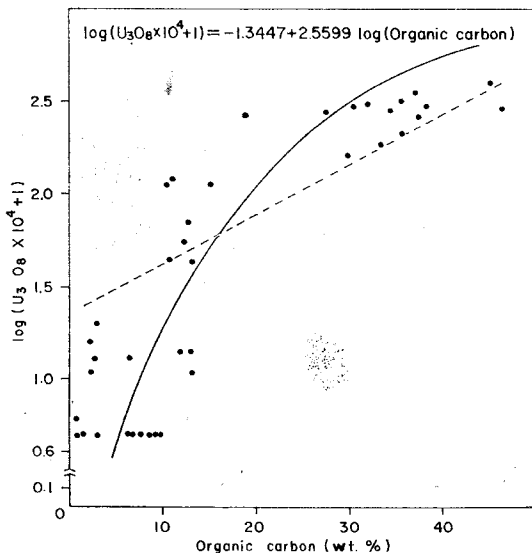


Fig. 2 The diagram of the relationship for organic carbon (wt. %) and log uranium (wt. %).

the logarithms of the uranium concentrations.

Table 1 shows the correlation coefficient between any two variables by statistical analyses. The degree of association between each two variables of constituent can be calculated by squaring the correlation coefficients. The most meaningful correlation in the black slates from Jinsan area is between log uranium and organic carbon ($r=0.845$) and these two constituents have a 71.4% association.

Figure 2 presents the calculated regression curve having the formula: $\log (U_3O_8 \times 10^4 + 1) = -1.3447 + 2.5599 \log (\text{organic carbon})$. The dotted line is a regression line having the formula of the Sintanjin-Beoun, Daejeon-Geumsan, Miwon, Geosan, Chungju area of Ogcheon Group (Park and So, 1980): $\log (U_3O_8 \times 10^4 + 1) = 1.1305 + 0.0083 (\text{organic carbon})$. It appears that, within a given specimens at least, the most uranium content is found in those specimens containing the most organic carbon.

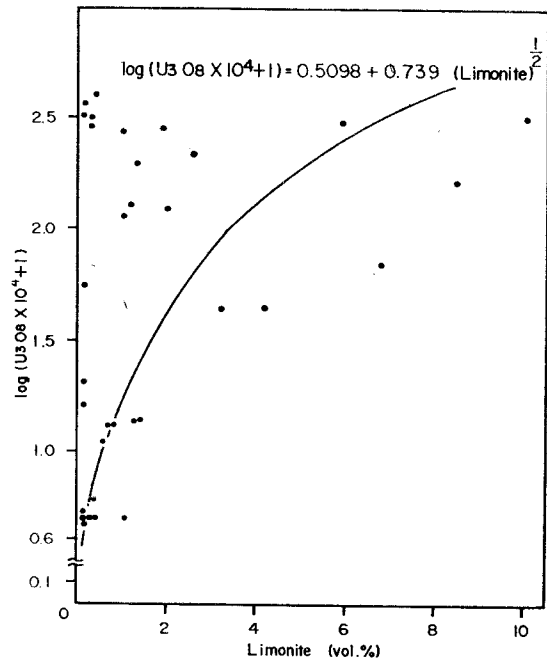


Fig. 3 The diagram of the relationship for limonite (vol. %) and log uranium (wt. %).

Table 1 Correlation coefficients between each two variables of constituents.

	Log (U ₃ O ₈)	Organic Carbon	Coal	Quartz	Muscovite	Biotite	Clay Substance	Other Silicates	Barite	Apatite	Calcite	Hematite	Limonite	Other Opaques
Log(U ₃ O ₈)	1.00	0.845	0.483	-0.364	-0.332	-0.106	-0.203	-0.068	-0.188	-0.174	0.284	0.302	0.300	-0.302
Organic Carbon		1.00	0.536	-0.531	-0.372	-0.225	-0.104	-0.098	-0.333	-0.186	0.318	0.202	0.236	-0.212
Coal			1.00	-0.820	-0.554	-0.030	-0.348	-0.125	-0.372	-0.010	0.081	-0.268	0.180	-0.525
Quartz				1.00	0.319	-0.102	0.265	0.153	0.492	-0.093	-0.092	0.164	0.066	0.369
Muscovite					1.00	0.276	0.052	-0.200	0.273	-0.098	-0.199	0.289	0.044	0.131
Biotite						1.00	-0.029	-0.045	0.179	-0.084	-0.152	0.070	0.232	-0.074
Substance							1.00	0.231	-0.070	-0.067	-0.063	-0.060	0.097	0.866
Other Silicates								1.00	0.160	-0.227	-0.055	-0.057	0.040	0.384
Apatite									1.00	-0.095	-0.142	-0.106	0.122	-0.066
Calcite										1.00	-0.005	-0.053	0.100	-0.099
Hematite											1.00	0.063	0.459	-0.094
Limonite												1.00	0.626	-0.031
//													1.00	-0.016
Other Opaques														1.00

* All significant at the 1% level of significance

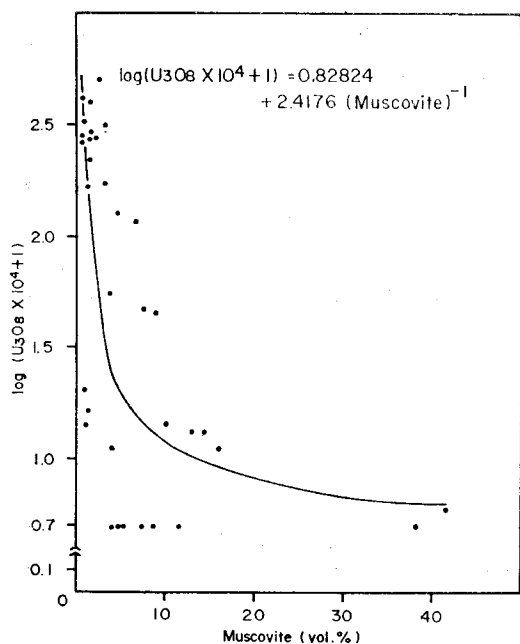


Fig. 4 The diagram of the relationship for muscovite (vol. %) and log uranium (wt. %).

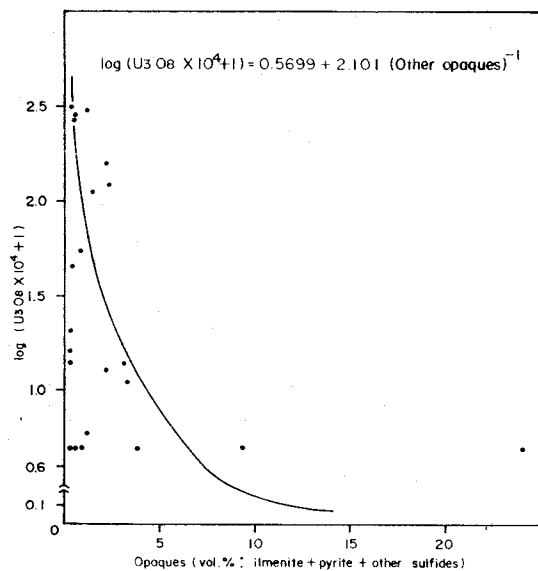


Fig. 5 The diagram of the relationship for opaques (vol. %) and log uranium (wt. %).

The strong relation between uranium and organic carbon in the black slates suggests that uranium was deposited together under similar physico-chemical environmental conditions, and

that the abundance of organic carbon controlled also the amounts of uranium. According to Haung (1978) the efficiency of the adsorption of uranium is related to the degree of maturity of organic matter or degree of carbonization attained by the humic materials.

Fig. 3 shows that limonite have a slight positive correlation, but muscovite and other opaques have a negative correlation in Figs. 4, 5.

The uranium is associated with and is genetically related to iron oxides and coal materials (Swanson, 1956). Kazumi (1978) suggested that adsorption and coprecipitation of uranium by and with limonite show a tendency similar to that of carbonaceous matter. However, in Fig. 4, correlation of uranium and limonite has no meaning ($r=0.3$). It indicates that some amount of iron oxides may be precipitated after uranium mineralization.

A multiple correlation and regression analysis of all black slates was employed to determine whether a significant improvement in the explained uranium variation could be obtained when all major constituents were considered rather than organic carbon alone in the black slates. This analysis resulted in a 5.89% increase in the percentage of uranium variation.

Since the 13 dimensional multiple regression line between uranium and the entire variation cannot be graphically represented, the deviation of each of the black slates from this line must be examined mathematically. In 95% of the specimens examined, the predicted log uranium values lie within the 2 Sy limits of 0.732 of the determined value. The relationship between the entire mineral composition and uranium in this 13 dimensional configuration can best be summarized by the prediction from the actual log uranium value, $\sum (Y - Y')/N$, for each black slates (0.118652) in Jinsan area, and the multiple correlation is 0.879.

From this $\sum(Y-Y')/N$ value it can be seen that all the black slates lie near the calculated regression line having the formula: $\log(U_3O_8 \times 10^4 + 1) = -1.3447 + 2.5599 \log(\text{organic carbon}) + 0.00574(\text{quartz}) - 0.00964(\text{muscovite}) + 0.37827(\text{biotite}) - 0.02286(\text{clay substance}) + 0.01268(\text{other silicates}) + 0.01032(\text{barite}) - 0.00224(\text{apatite}) + 0.01606(\text{calcite}) + 0.08258(\text{hematite}) - 0.02406(\text{limonite}) - 0.01715(\text{other opaques})$.

CONCLUSIONS

To better understand environmental controls of uranium distribution in black slates, a total of 40 specimens from the Jinsan area of Ogcheon Group were collected and analyzed by mineralogical and radiometric techniques.

Most of the uranium (meta-uranocircite and torbernite) in the black slates is of syngenetic origin, but some of them developed within structural plane of slates suggest that the uranium was dissolved in ground water, and then was fixed in minerals by various ways during regional metamorphism. 40 specimens from the Jinsan area show an organic carbon-log uranium correlation of +0.845 and these two constituents have about a 71.5% association. This suggests the chemical and geological factors that controlled the abundance of organic carbon also controlled the uranium content.

When all compositional factors are taken into account by multiple regression analysis, the association of mineral composition with log uranium for all the black slates improves 5.89% (from $r^2=71.4\%$ to $R^2=77.3\%$). The regression showing the relationship of the major components to uranium is as follows: $\log(U_3O_8 \times 10^4 + 1) = 0.77396 + 0.04465(\text{organic carbon}) + 0.00574(\text{quartz}) - 0.00964(\text{muscovite}) + 0.37827(\text{biotite}) - 0.02296(\text{clay substance}) + 0.01268(\text{other silicates}) + 0.01032(\text{barite}) - 0.00224(\text{apatite}) + 0.01606(\text{calcite}) + 0.08258(\text{hematite}) - 0.02406(\text{limonite}) - 0.01715(\text{other opaques})$.

Using this equation, the uranium content of any black slates which belong to the same population as those evaluated herein, can be calculated on the basis of modal analysis of the mineral constituents of the slates.

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珍山一帶 炭質變成泥岩層內的 우라늄의 賦存狀態

池世定* · 金世鉉* · 李 翊 九*

要約 : 本 究 研 是 忠 南 - 珍 山 一 帶 的 沃 川 系 炭 質 黑 色 粘 板 岩 中 賦 存 的 烏 拉 鈾 分 布 的 特 性 及 烏 拉 鈾 與 組 成 礦 物 的 關 係 를 밝 히 기 爲 하 여 시 도 되 었 다.

本 究 研 地 域 으 로 부 터 채 취 된 시 료 중 40 개 의 시 료 를 택 하 여 礦 物 學 的 地 化 學 的 究 研 을 實 施 하 고 콤퓨터 (IBM 370) 를 利 用 한 統 計 的 相 關 關 係 를 提 示 하 였 다.

우라늄과의 正의 相 關 關 係 를 갖는 광물은 有 機 炭 素, 赤 鐵 鑛, 褐 鐵 鑛 이 며, 負 的 相 關 關 係 를 갖는 礦 物 은 石 英, 白 雲 母, 沸 石 類 광 石 광 物 이 다. 이 중 가장 높은 正 的 相 關 關 係 를 갖는 것은 有 機 炭 素 이 며, 우라늄의 隨 伴 傾 向 은 71.4% 이 다. 우라늄 부존량과 有 機 炭 素 와 의 回 歸 方 程 式 은 $\log(U_3O_8 \times 10^4 + 1) = -1.3447 + 2.5599 \log(\text{Organic carbon})$ 으 로 表 示 되 고 추 定 표 준 오 차 Sy 값 은 0.1498 에 이 른 다. 이 는 유 기 탄 소 를 부 존 시 킨 地 質 環 境 은 역 시 우라늄 침 전 에 도 영 향 을 주 었 을 을 암 시 해 준다. 野 外 조 사 에 의 하 면 우라늄 광 물 은 주 로 黑 色 粘 板 岩 中 炭 質 物 內 에 產 出 되 고, 가 끔 黑 色 粘 板 岩 의 構 造 面 上 에 散 點 狀 으 로 分 布 하 는 데 후 자 의 경 우 는 우라늄 침 전 후 의 二 次 的 인 移 動 에 의 한 것 임 을 암 시 해 준다. 重 相 關 關 係 와 多 邊 數 回 歸 分 析 을 通 한 多 元 回 歸 方 程 式 은 $\log(U_3O_8 \times 10^4 + 1) = 0.77396 + 0.04465(\text{organic carbon}) + 0.00574(\text{quartz}) - 0.00964(\text{muscovite}) + 0.37827(\text{biotite}) - 0.02286(\text{clay substance}) + 0.01268(\text{other silicates}) + 0.1032(\text{barite}) - 0.00224(\text{apatite}) + 0.01606(\text{calcite}) + 0.08258(\text{hematite}) - 0.2406(\text{limonite}) - 0.01715(\text{other opaques})$ 로 表 示 되 고, 이 에 의 한 추 定 의 표 준 오 차 Sy 값 은 0.1186 이 며 重 相 關 關 係 의 값 은 0.879 이 다. 따 라 서 多 邊 數 回 歸 分 析 에 서 는 有 機 炭 素 만 을 包 含 한 回 歸 方 程 式 보 다 수 만 경 향 이 5.89% 증 가 된 다.

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