YU ISSN 0015—3206
FZKAAA 14 (1) 1 (1982)

# DETERMINATION OF THORIUM, URANIUM AND POTASSIUM IN SOME NATIONAL COALS

P. R. ADŽIĆ, A. H. KUKOČ and R. STEPIĆ

*Institute »Boris Kidrič«, P. O. Box 522, 11001 Belgrade, Yugoslavia*

Received 24 April 1981

Revised manuscript received 28 August 1981

UDC 513. 16

Original scientific paper

The natural gamma-ray spectra measurements were performed in order to determine Th, U and K concentrations in seven lignites and one bituminous coal. Th/U mass ratio and uranium isotopic ratio $^{238}U/^{235}U$ of the samples measured were determined and discussed.

## *1. Experimental procedure and aparatus*

The concentration of Th, U and K has been determined in seven lignites and bituminous coal by gamma-ray intensity measurements of relevant natural radioisotopes.

Measurements were performed with low-background Ge(Li) spectrometer ($N_B > 30$ keV) $= 0.88 \pm 0.01$ cps, efficiency 5.4% and FWHM $= 2.5$ keV, where $N_B$ denotes integral background count rate above the treshold of 30 keV.

The lowest concentrations of Th, U and K which can be determined with this spectrometer are $5 \cdot 10^{-7}$ g/g for Th and U and $2 \cdot 10^{-4}$ g/g for K for 1 kg samples and data acquisition time of 200 ks. However, data acquisition time could be shortened by an order of magnitude if now commercially available high efficiency Ge(Li) spectrometers are used.

Dried coal samples have been ground and enclosed in one litre air-tight polyethylene bottles, and were kept two to three weeks closed before the commencement of the measurements. The identical type of containers were used for the determination of experimental efficiency curve by measurement of the homogeneous admixture of 832 g »Banovići« lignite and 401 g of natural phosphate from Morocco. The uranium concentration in the natural phosphate had been determined previously ($125 \pm 12$ ppm). It has been also proved that $^{238}$U in phosphate is in equilibrium with its daughters ($^{226}$Ra)[1]. The efficiency callibration curve has been obtained in the energy range 144 - 2447 keV, using the strongest gamma-lines of $^{235,238}$U decay and their daughters.

The activities of Th and U have been determined by the intensity measurements of the most intensive single gamma-lines in the thorium and uranium families. The absolute gamma-ray intensity data for absolute activity calculations are chosen from several recent publications [2-5]. For each line chosen, the activity had been determined, and then weighted averaged for each spectrum. Finaly, activities were averaged out over different runs. Each sample was measured two to three times, and data acquisition time varied from 200 to 260 ks per spectrum.

The background spectrum has been measured several times between coal sample measurements (with one litre of triple distilled water in the sample position) so that possible variations of the background line intensities can be captured. Weighted average of background line intensities have been used for the background subtraction.

## 2. *Results and discussion*

All the coals measured, are located on the geological time scale between the Middle Miocene to the Upper Oligocene [6], and if left unperturbed in close geological system should contain $^{238}$U in the equilibrium with $^{226}$Ra ($^{234}$U).

Due to the fact that coal had been ground and heated, and that some of the samples had had different history after the removal from the deposite, it is important to check the $^{238}$U - $^{226}$Ra equilibrium, because the determination of U heavily rides on the gamma-spectrometry of radium daughters. (The equilibrium checking by the method used[1], is equivalent to the gamma-spectrometric determination of uranium isotopic ratio $R = {}^{238}\mathrm{U}/{}^{235}\mathrm{U} = 138$.)

All the results of the measurements are presented in Table 1, and the last column contains uranium isotopic ratio $R$. As it is evident, only for the lignite »Banovići« ratio $R$ has substantially smaller value than 138. In this case U has been determined from $^{235}$U gamma-lines. The average Th/U mass ratio calculated from the data in the Table amounts 1.2 and it is much smaller than the average value in the Earth's crust ($\approx 3$), probably due to the lower solubility of Th natural compounds in water[7]. It should be noticed a somewhat low concentration of potassium in the lignite »Bogovina« and small concentrations (the lowest for this gamma-spectrometer) of all three elements in the lignite »Kreka«.

Gamma spectra in Figs. 1 and 2 belong to the lignites »Bogovina« and »Morava«, respectively. The lignite »Bogovina« is unique sample in this set with five times higher concentration of U than Th (Th/U ratio 0.2). The lignite »Morava« has the highest concentration of thorium and potassium, and together with lignite »Rembas« the highest Th/U weight ratio, closest to that in the Earth's crust.

TABLE 1

| COAL SAMPLE | WEIGHT [g] | CONCENTRATION [g/g] $10^{-6}$ | | | Th/U | $R$ |
|---|---|---|---|---|---|---|
| | | Th | U | K | | |
| MORAVA | 920 | $10.9 \pm 0.6$ | $5.3 \pm 1.4$ | $(1.27 \pm 0.14)\ 10^4$ | 2.0 | $108 \pm 35$ |
| SOKO | 950 | $6.0 \pm 0.4$ | $5.1 \pm 0.3$ | $(5.05 \pm 0.62)\ 10^3$ | 1.2 | $141 \pm 50$ |
| REMBAS | 1000 | $4.7 \pm 0.4$ | $2.2 \pm 0.2$ | $(3.26 \pm 0.45)\ 10^3$ | 2.1 | $111 \pm 50$ |
| BOGOVINA | 1140 | $1.8 \pm 0.6$ | $8.9 \pm 1.1$ | $< 1.7 \cdot 10^2$ | 0.2 | $156 \pm 59$ |
| IBAR | 1215 | $7.6 \pm 1.1$ | $7.0 \pm 0.8$ | $(8.04 \pm 0.93)\ 10^3$ | 1.1 | $123 \pm 42$ |
| KREKA | 935 | $< 0.4$ | $< 0.5$ | $< 2 \cdot 10^2$ | — | — |
| ŽIVINICE | 952 | $1.4 \pm 0.4$ | $< 0.6$ | $(1.05 \pm 0.21)\ 10^3$ | — | — |
| BANOVIĆI | 1043 | $0.9 \pm 0.4$ | $1.6 \pm 0.6$ | $(1.15 \pm 0.19)\ 10^3$ | 0.6 | $70 \pm 32$ |

Concentrations of Th, U and K in coals in ppm. Th/U mass ratios and uranium isotope ratios $R$. All coals are lignites but »Ibar« which is bituminous. Coal samples are named after the relevant mines in S. R. Serbia and S. R. Bosnia and Herzegovina.

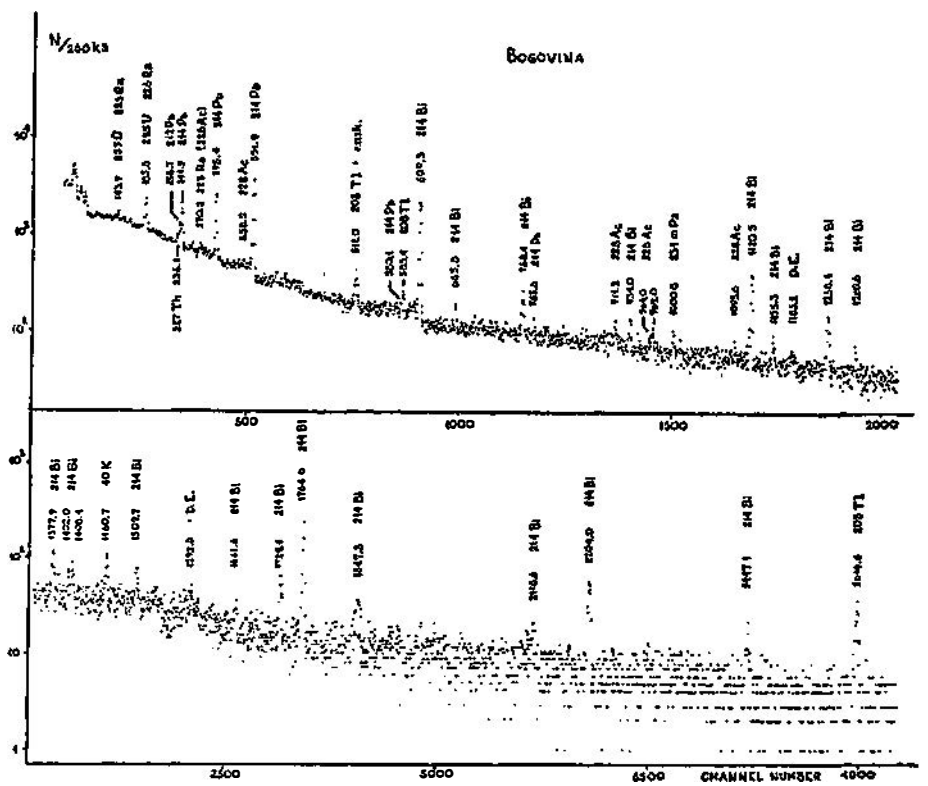


Fig. 1. Natural gamma-ray spectrum of »Bogovina« lignite. With D. E. double escape peaks are denoted. The background has not been subtracted.

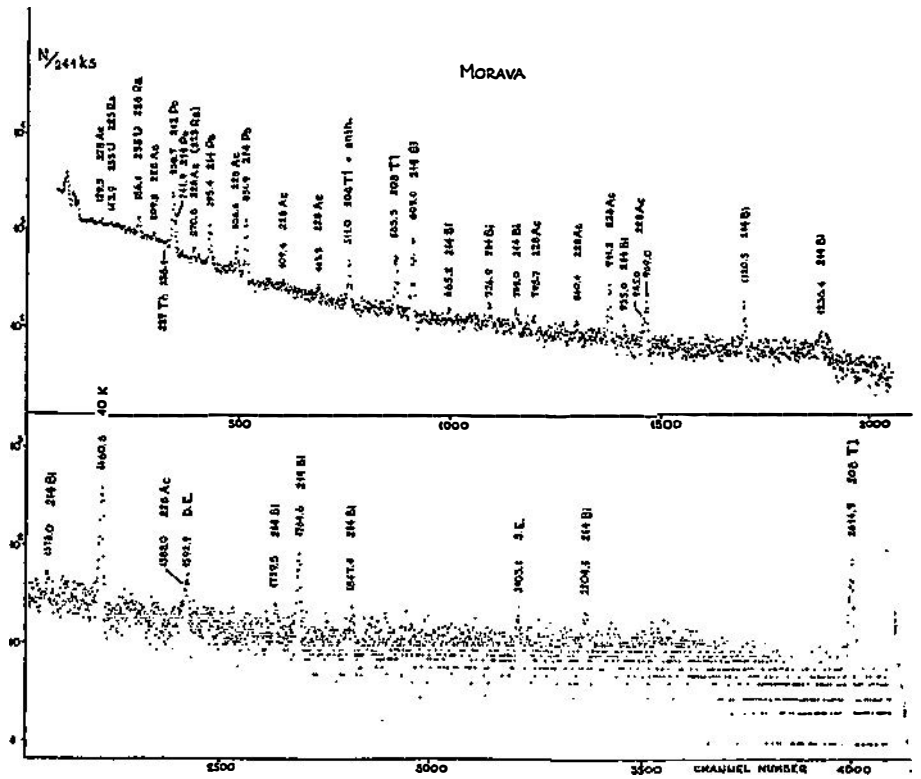


Fig. 2. Natural gamma-ray spectrum of »Morava« lignite. With S. E. and D. E. single and double escape peaks are, respectively, denoted. The background has not been subtracted.

## Acknowledgments

We would like to thank to Dr. M. Hadžiefendić and Mr. P. Bojović. B. Sci. for the coal samples.

## References

1) P. R. Adžić, Gamma-spectroscopy of Natural Samples, M. Sc. Thesis (in serbo-croatian), Belgrade 1980;
2) N. G. Gusev, P. P. Dimitriev, *Kvantovoe Izluchenie Radioaktivnih Nuklidov,* Atomizdat, Moscow 1977;
3) D. J. Horen, NDS **17** (1976) 367;
4) M. R. Schmorak, NDS **21** (1977) 91;
5) C. M. Lederer, V. S. Shirley editors, *Table of Isotopes,* seventh edition, John Wiley & Sons, New York 1978;
6) N. Pantić, P. Nikolić, *Ugalj,* Naučna Knjiga, Beograd 1973;
7) V. M. Gorbachev, Yu. S. Zamiatin, A. A. Lbov, *Osnovnie Charachteristiki Tjazholih Elementov,* Atomizdat, Moscow 1975.

# ODREĐIVANJE TORIJUMA, URANA I KALIJUMA U NEKIM DOMAĆIM UGLJENIMA

P. R. ADŽIĆ, A. H. KUKOČ i R. STEPIĆ

*Institut »Boris Kidrič«-Vinča, p. p. 522, 11001 Beograd*

UDC 513.16

Originalni naučni rad

Određene su koncentracije Th, U i K u sedam lignita i jednom mrkom ugljenu, na osnovu mjerenja prirodnih gama-spektara. Mjereni su i diskutirani maseni odnos Th/U i izotopski odnos $^{238}U/^{235}U$.