A UNION NEUTRON-GAMMA LOGGING METHOD FOR DETERMINATION OF URANIUM-RADIUM DISEQUILIBRIUM COEFFICIENT

by

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Owing to the influence of continuous running of ground water, the uranium atoms can be separated physically from their daughters for the reason of different solubilities and the uranium deposit often shows the disequilibrium feature between uranium and its daughter products (radium principally). It is important, when spectral gamma ray logging, to quantify the uranium content which can cause inaccuracy of the result. This paper, based on spectral γ ray logging method, proposes a neutron-gamma logging method to determine the coefficient of uranium-radium disequilibrium. In this method, characteristic peak count rate of uranium is taken from prompt fission neutron logging, whereas characteristic peak count rate of radium, thorium and potassium are taken from spectral gamma ray logging. Based on this method, the union logging tool including epithermal neutron, thermal neutron, and gamma detector along with D-T generator, have been developed. The experimental results, in standard model wells, show that this method is in good agreement within 7 % in core assay results. It shows that the union neutron-γ logging method can be used for field uranium logging jobs.

Key words: prompt fission neutron, spectral gamma ray, uranium logging, disequilibrium coefficient

INTRODUCTION

Uranium resources, as the raw material which produces fuel for nuclear power facilities, have a significant impact on low-carbon electricity generation. Generally, uranium deposits are distributed with feature of flat lying and weak deforming, which need to be detected by borehole logging technique and mined by *in-situ* leach method. The traditional borehole logging technique of exploring uranium deposits and quantifying the grade of the uranium, by spectral γ ray (SGR) logging mainly, is used to measure the spectrum of γ rays emitted via natural radioactivity of uranium (^{238}U and ^{235}U), thorium (^{232}Th and ^{230}Th), and potassium (^{40}K) in the borehole formation. These radioactive isotopes emit γ rays with characteristic energy level and can be used to quantify the grade of uranium in the deposit [1].

Typically, the SGR logging measures the radiation emitted by the daughter products (^{214}Bi principally) of the decay of uranium isotopes (as shown in fig. 1) and can be used to quantify the uranium grade. However, due to the continuous (or intermittent) movement of ground water, the uranium atoms can be separated physically from their daughters for the reason of different solubilities. Consequently, the radioactive daughter products can be originated from other long-lived daughter isotope (radium principally). As a result, the uranium-radium disequilibrium phenomenon is formed and the SGR logging is indistinguishable to differentiate the γ radiation from uranium atoms or their daughters.

Actually, the uranium-radium disequilibrium phenomenon is very ubiquitous in nature especially in the sandstone-hosted deposit, which is difficult for SGR logging alone to quantify the actual uranium grade accurately. The prompt fission neutron (PFN) logging, using a pulsed neutron source and looking for prompt neutrons from ^{235}U fission, is an efficient way to detect ^{235}U directly [2, 3]. In this method, epithermal and thermal neutron detectors are usually both equipped to measure the counts of them as the response of the uranium grade [4] without requiring the disequilibrium coefficient. However, compared with

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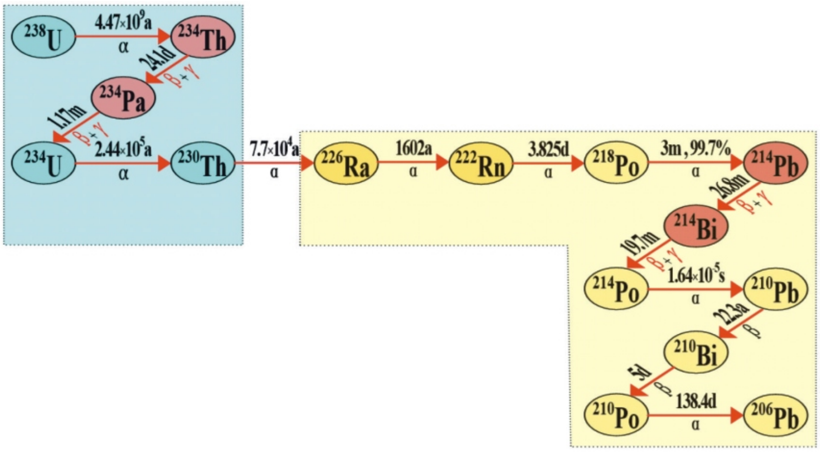

Figure 1. Part of ^{238}U radioactive decay chain with emission products and half-life periods

SGR logging, the PFN logging requires almost seven times longer time than that of SGR logging due to the inherent characteristic of fewer yield of secondary neutrons production [5, 6].

This paper, based on SGR logging method, proposes a new union uranium logging method to solve the disequilibrium problem and improve the efficiency of PFN logging. In this paper, uranium and radium content are obtained from PFN and SGR logging tool separately. Accordingly, the disequilibrium coefficient is determined from the counters of PFN and SGR detectors. Based on this method, we have developed the union neutron-γ uranium logging tool. The experimental results in standard model wells show the disequilibrium coefficient based on this method is in good agreement within 7 % in results of core assay analysis.

METHODOLOGYS

Theoretical method of PFN logging

The principle of PFN logging can be illustrated in fig. 2. First of all, a deuterium-tritium (D-T) accelerator sends out a burst of 14 MeV neutrons into the formation around within 4 space of the borehole in a very short time (10~20 s). After that these neutrons are moderated to thermal neutrons (0.025 eV) about 1 m around the accelerator, meanwhile, the thermal neutrons are absorbed by the nucleus in the formation rock by less than 10 cm. When thermal neutrons encounter ^{235}U, most of them will induce a fission reaction and emit 2 or 3 fission neutrons (1.95 MeV), and more than 99 % of them are PFN. Before the PFN slowed down to a thermal neutron by borehole formation, neutron detectors can record the signal of the epithermal neutron (0.7 eV~1.0 keV) and thermal neutrons in the borehole. By recording these neutrons, one can quantitatively evaluate the content of ^{235}U (or uranium) in the formation.

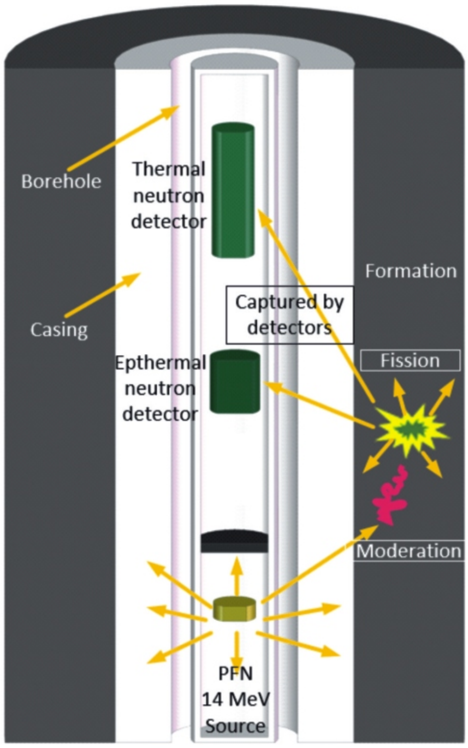

Figure 2. The principle of PFN logging

Assuming the time distribution of thermal neutron density in uranium formations is $n_{\mathrm{th}}(t)$, according to the principle of neutron and uranium interaction, the time distribution of epithermal neutron density $n_{\mathrm{ep}}(t)$, derived from thermal fission reactions of ^{235}U in the time range of detector measurement, can be represented as

$$n_{\mathrm{ep}}(t)\mathrm{d}t \quad n_{\mathrm{th}}(t)kq_{\upsilon}\mathrm{d}t \tag{1}$$

where the coefficient k is defined as

$$k \quad \frac{\alpha}{AN_A\sigma_f v} \tag{2}$$

where α, A are abundance and atomic weight of ^{235}U respectively, N_{A} – the Avogadro constant, σ_{f} – the fission cross-section between thermal neutron, and ^{235}U, v – average yield of secondary neutrons produced by fission, and q_v represented the uranium content of formation.

After $\Delta t = t_2 - t_1$ time interval of neutron source bombarding, the total counting rate of epithermal neutron detector can be expressed as

$$N_{\mathrm{ep}}(\Delta t) = \int_{t1}^{t2} n_{\mathrm{th}}(t)\, k q_v \mathrm{d}t = k q_v N_{\mathrm{th}}(\Delta t) \quad (3)$$

where $N_{\mathrm{th}}(\Delta t)$ is the total counting rate of thermal neutron detector after Δt time interval. From eq. (3) we can get the q_v by

$$q_v = \frac{N_{\mathrm{ep}}(\Delta t)}{kN_{\mathrm{th}}(\Delta t)} = k' \frac{N_{\mathrm{ep}}(\Delta t)}{N_{\mathrm{th}}(\Delta t)} = k' R_{e/t}(\Delta t) \quad (4)$$

where $k' = 1/k$.

Obviously, after a certain time interval of the neutron source emitting, the ratio of epithermal and thermal neutrons shows a certain proportional relationship with the uranium grade.

Union neutron-gamma logging method for determination coefficient

In SGR logging, characteristic peak count rates at specific energy represent the positive relationship with the quantitation of radioactive element. Consequently, characteristic peak count rates at depth z can be represented as $N_i(z)$, where $i = 1$ means the rate of Th (2.62 MeV), $i = 2$ denotes the rate of Ra (1.76 MeV), $i = 3$ indicates the rate of ^{40}K (1.46 MeV). As a result, $N_i(Z)$ can be expressed as

$$N_i(z) = B_i + \sum_{k=1}^{3} A_{ki}\, q_k(z) \quad (5)$$

where $q_k(z)$ is the quantitative content for element k, A_{ki} – the calibration factor matrix and need to be calibrated in different standard model well, k denotes the k^{th} radioactive element (k = 1-3 represents Th, Ra, and ^{40}K, separately) with unit content; i represents the i^{th} characteristic peak count rates (i = 1-3 represents count rates of Th, Ra, and ^{40}K, separately). $B_i = N_i(z_0)$ is the background count rate of i^{th} characteristic peak. The $N_2(Z)$ is equivalent to the rate of U, when uranium and radium are in balance. Otherwise, we have to use PFN result to represent the rate. In this case, as mentioned above, $R_{\mathrm{e/t}}$ in eq. (4) also represents the positive relationship with uranium quantitation in PFN logging. Hence, one can see that it is possible to combine PFN and SGR logging together for solving disequilibrium problem.

In order to determine the coefficient of uranium-radium disequilibrium, we add $N_0(Z)$ in eq. (5) which is represented by $R_{\mathrm{e/t}}$ in eq. (4) multiply the coefficient k' instead of uranium peak count rates. Thus, eq. (5) becomes

$$B_i + \sum_{k=1}^{4} A_{ki} q_k(z) = \begin{cases} k' R_{e/t}, & i = 0 \\ N_i(z), & i \neq 0 \end{cases} \quad (6)$$

and A_{ki} can be defined as

$$A_{ki} = \begin{pmatrix} A_{00} & A_{01} & A_{02} & A_{03} \\ A_{10} & A_{11} & A_{12} & A_{13} \\ A_{20} & A_{21} & A_{22} & A_{23} \\ A_{30} & A_{31} & A_{32} & A_{33} \end{pmatrix} \quad (7)$$

According to the different principles of PFN and SGR logging, the result of PFN logging does not depend on SGR logging, A_{i0} and A_{0i} are all equal to zero (i = 1-3). Besides, the count rate of γ ray at lower energy makes no contribution to that at higher energy. Consequently, eq. (7) can be simplified as the upper triangular matrix form as

$$A_{ki} = \begin{pmatrix} A_{00} & 0 & 0 & 0 \\ 0 & A_{11} & A_{12} & A_{13} \\ 0 & 0 & A_{22} & A_{23} \\ 0 & 0 & 0 & A_{33} \end{pmatrix} \quad (8)$$

Above all, eq. (6) is can be rewritten as

$$\begin{aligned} N_0(z) &= k' R_{\mathrm{e/t}} = B_0 + A_{00} q_0(z) \\ N_i(z) &= B_i + \sum_{k=1}^{3} A_{ki} q_k(z), \; i = 1,2,3 \end{aligned} \quad (9)$$

For solving uranium-radium disequilibrium problem, the ratio of $q_0(z)$ and $q_2(z)$ can be used to express the disequilibrium factor K_p as

$$K_p = \frac{q_2(z)}{q_0(z)} = \frac{(N_2(z) - B_2 - A_{12}q_1)/A_{22}}{(k' R_{\mathrm{e/t}} - B_0)/A_{00}} = \frac{A_{00}}{A_{11}A_{22}} \frac{N_2(z)A_{11} - B_2A_{11} - [N_1(z) - B_1]A_{12}}{(k' R_{\mathrm{e/t}} - B_0)} \quad (10)$$

Union neutron-γ logging method for high efficiency

Generally, the PFN uranium logging is a quite inefficient method due to its characteristic of fewer PFN. As a matter of fact, the PFN logging often requires almost seven times longer period than that of SGR logging in order to meet the same tolerance error.

In union neutron-γ logging method, for higher efficiency, SGR logging is used to delineate the boundary of ore beds, whereas the PFN logging can be started (or stopped) when the neutron detector entered (or left) the boundary. Besides, in PFN logging, we can gather Δt time (from 120 μs to 520 μs in fig. 3) interval of time spectra for two purposes: count rate of epithermal neutron substantially parallel with that of thermal neutron and higher count rate for higher efficiency.

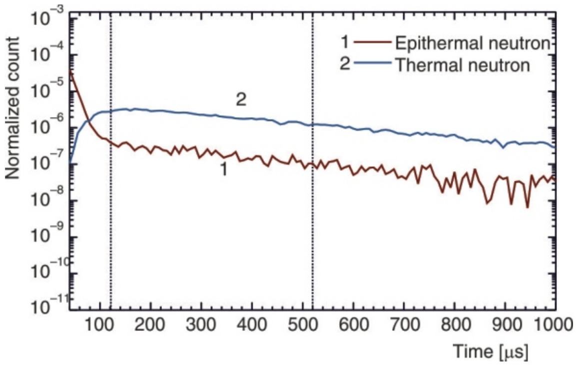

Figure 3. The segmentation of neutron time spectra

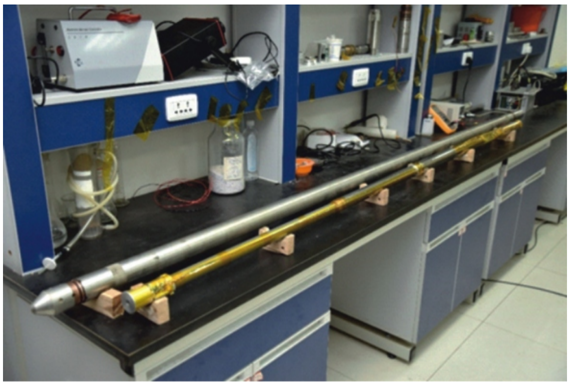

Figure 4. The union logging tool in laboratory

EXPERIMENTAL INSTRUMENTS AND RESULTS

Based on the principle and aforementioned, a union logging tool including a *D-T* pulsed neutron generator (shown in fig. 4), epithermal and thermal neutron detectors as well as a natural γ detector were placed in a stainless-steel cylinder, 380 mm length and 65 mm diameter. The *D-T* pulsed neutron generator provides 14 MeV neutrons with a yield of $4\ 10^8$ s^{-1}. The epithermal neutron detector is made up of a He-3 neutron detector covered by high-density polyethylene (5 mm) and cadmium (1 mm) for neutron moderation and absorption (130 mm length and 38 mm diameter). The thermal neutron detector is also made up of He-3 and the size is 130 mm length and 40 mm diameter and the γ detector is formed by $LaBr_3$:Ce and the diameter is 50 mm. The measurement structure of the logging tool is displayed in fig. 5.

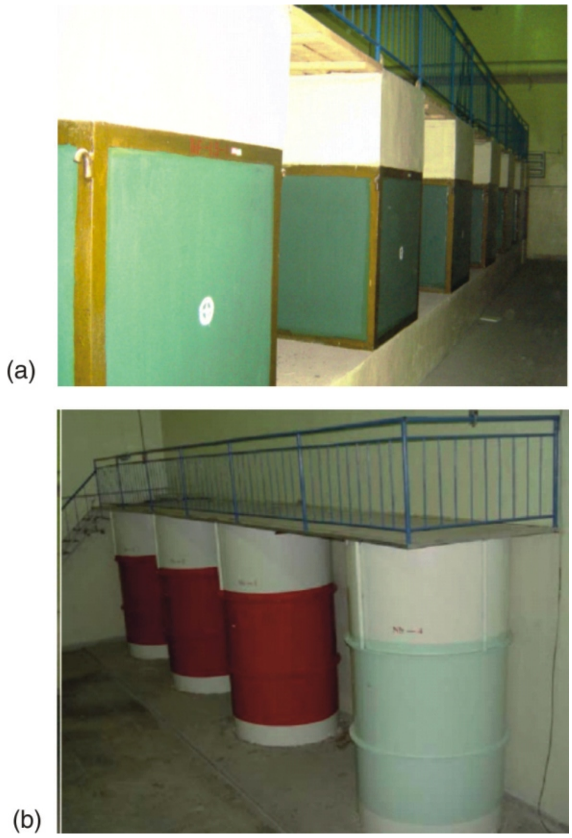

Figure 6. Standard model for measurement of coefficient of uranium-radium equilibrium; (a) standard well for logging, (b) standard well for neutron logging

The tool has been calibrated in standard models of Airborne Survey and Remote Sensing Center of Nuclear Industry of China. It provides 18 types of standard model wells and four of the cylindrical ones are designed for neutron logging, whereas others for γ logging purpose (shown in fig. 6). In this paper, we chose cylindrical model wells as standard wells named as Nu-1, Nu-2, Nu-3, and Nb-4 (background). The detail information of each well model is shown in tab. 1.

Table 1. The detail configuration of standard well model

No.	Radius [m]	Height [m]	Uranium content [ppm]*	Coefficient of uranium-radium equilibrium
Nu-1	0.7	1.8	281	0.867
Nu-2	0.7	1.8	685	0.919
Nu-3	0.7	1.8	983	0.919
Nb-4	0.7	1.8	1.56	0.73

*1 ppm = 10^{-6}

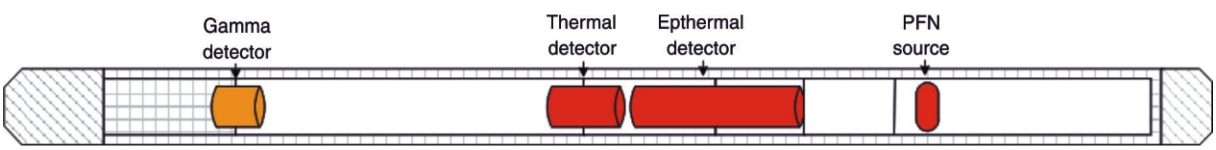

Figure 5. The structure of union logging tool

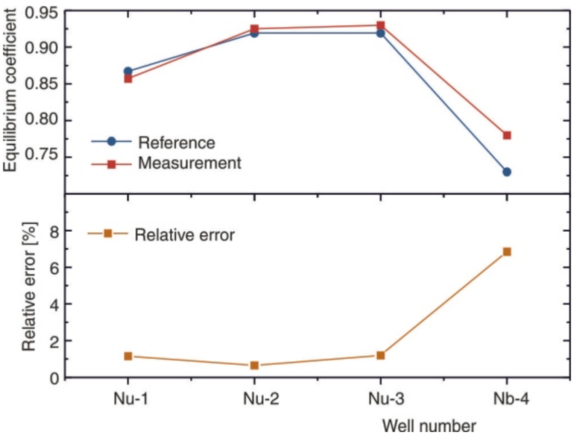

Figure 7. Result of coefficient of uranium-radium equilibrium in standard model

The result of coefficient of uranium-radium disequilibrium in standard model is shown in fig. 7. The results are in accordance with the reference values in of maximum relative error of 7 %, which is meeting the accuracy requirements of field logging. The large error occurs because of less data acquired in background well logging.

CONCLUSION

The method used for measurement of the coefficient of uranium-radium disequilibrium with PFN and SGR logging, have been proposed. Based on this method, the union logging tool, including epithermal neutron, thermal neutron and γ detector along with D-T generator, have been developed. The experimental results indicate that the instrument can be used to measure the coefficient of uranium-radium disequilibrium. The results agree with the reference values within maximum relative error of 7 %, well meeting the accuracy requirements of actual logging. In the future work, we will pay more attentions to the correlation of the influence factors such as oxygen activation during PFN logging, bore diameter and fluid type in borehole.

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AUTHORS' CONTRIBUTIONS

R. Chen, H-T. Wang and Z.-F. Liu designed the research, performed the research and analyzed the data. The paper was written by R. Chen. All authors discussed the results and revised the manuscript.

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НЕУТРОН-ГАМА МЕТОДА УЈЕДИЊЕНОГ ЕВИДЕНТИРАЊА ЗА ОДРЕЂИВАЊЕ КОЕФИЦИЈЕНТА НЕРАВНОТЕЖЕ УРАНИЈУМ РАДИЈУМА

Захваљујући утицају непрекидног струјања подземне воде, атоми уранијума могу се физички одвојити од својих потомака услед различитих растворљивости, те лежиште уранијума често показује неравнотежу између уранијума и његових потомака (углавном радијума). Када се квантификује садржај уранијума евидентирањем спектра гама зрачења то може проузроковати нетачан резултат. У овом раду, засновано на методи евидентирања спектра гама зрачења, предлаже се метода неутрон-гама евидентирања за одређивање коефицијента неравнотеже уранијум радијума. По овој методи, брзина бројања у карактеристичном пику уранијума добија се из евиденције промптних фисионих неутрона, док се брзина бројања у пику радијума, торијума и калијума добија евиденцијом из спектра гама зрачења. На основу ове методе развијен је алат за уједињено евидентирање укључујући епитермалне неутроне, термичке неутроне и гама детектор заједно са деутеријум-трицијум генератором. Експериментални резултати у стандардним моделима детектора облика јаме показују да се ова метода уједињеног неутрон-гама евидентирања може користити у пословима теренске евиденције уранијума.

Кључне речи: промптни фисиони неутрон, спектар гама зрачења, евидентирање уранијума, коефицијент неравнотеже