Perspectives for the Development of Chemistry of Actinides in Kazakhstan

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Abstract: One of the urgent tasks of the uranium industry in Kazakhstan is the extraction of radionuclide residues from spent uranium wells. We proposed to extract the remains of radionuclides from the drilled wells and to improve the radiation safety of the contaminated area. We conducted a chemical analysis of the content of radionuclides in the ore material. The radionuclides U, Th, Pa, Ra, Ac, Rn were extracted, which can be used in industry and medicine in Kazakhstan.

Key words: Chemistry of actinides, radionuclides Th-234, Th-230, Th-227, Pa-231, Pa-234 m, Ac-228, Ra-223, Ra-224, Rn-219.

1. Introduction

Kazakhstan annually produces 20,000 tons of uranium. One of the urgent tasks of the uranium industry in Kazakhstan is the extraction of radionuclide residues from spent uranium wells. We proposed to extract the remnants of radionuclides from the wells and improve the radiation safety of the area. We carried out a chemical analysis for the content of radionuclides in the ore material. The results of x-ray diffractometry and atomic emission spectral analysis of ore are given below.

2. Experimental

X-ray diffractometric analysis was carried out on a DRON-3 automated diffractometer with CuKα radiation, β-filter. Conditions for recording diffractograms: U = 35 kV; I = 20 mA; θ-2θ survey; detector 2 deg/min.

The determination of the content of gamma-emitting radionuclides in the core were carried out by instrumental gamma spectrometry in accordance with the method “Activity of radionuclides in counted samples.” Measurement procedure using gamma spectrometers using SpectraLine. Spectra were recorded on a gamma spectrometer with a semiconductor germanium detector “Canberra” head. No. 5401, verification certificate No. VA 17-04 32399 dated November 12, 2018.

3. Results and Discussion

Earlier, we developed the Muhamedzhan-1 metal complex catalyst for the oxidation of uranium U⁴⁺ to U⁶⁺ [1]. In this paper, we tried to find a catalyst for the extraction of radionuclides from uranium ores. Semi-quantitative X-ray phase analysis was carried out according to the diffraction patterns of powder samples using the method of equal weights and artificial mixtures. Quantitative ratios of crystalline phases were determined. The diffraction patterns were interpreted using ICDD card index data: PDF2 (Powder Diffraction File) powder diffraction data base and diffraction patterns of minerals free of impurities. For the main phases, the content was calculated.

Research work was carried out to determine the content of gamma-emitting radionuclides in the liquid and solid phases of the incoming sample. The separation of fractions in the incoming sample was performed by filtering using a red ribbon filter. The filter cake was dried at a temperature of 100°C.
for 6 hours. Subsequently, the separated sample by fractions was transferred to a special container for measurement.

The determination of the content of gamma-emitting radionuclides in the core were carried out by instrumental gamma spectrometry. Samples were placed in a plastic cylindrical container with a bottom thickness of 1 mm and placed directly on the gamma-detector window. Gamma-ray spectra were recorded for 64 hours for a liquid sample and 16 hours for a dry one. The results of determining the specific activities of radionuclides are given in the Table 3.

Table 1  Interplanar distances and phase composition of the sample.

| d, Å | I % | mineral       |
|------|-----|---------------|
| 13.88818 | 17.1 | chlorite      |
| 9.81234   | 20.8 | mica          |
| 7.00014   | 24.4 | chlorite      |
| 4.95122   | 14.8 | mica          |
| 4.69151   | 11.0 | chlorite      |
| 4.22964   | 26.4 | quartz        |
| 3.51825   | 19.6 | chlorite      |
| 3.32627   | 100.0 | quartz     |
| 3.22388   | 18.5 | KPS           |
| 3.18648   | 12.9 | PS            |
| 2.98132   | 11.1 | mica          |
| 2.57916   | 17.2 | mica          |
| 2.55881   | 11.0 | KPS           |
| 2.48643   | 9.1  | mica          |
| 2.44694   | 14.0 | quartz        |
| 2.37573   | 8.9  | mica          |
| 2.27322   | 11.3 | quartz        |
| 2.22821   | 10.0 | mica          |
| 2.11990   | 13.1 | quartz        |
| 1.98700   | 11.3 | mica          |
| 1.97434   | 10.5 | quartz        |
| 1.81329   | 15.8 | quartz        |
| 1.70454   | 5.5  | chlorite      |
| 1.66786   | 10.0 | quartz        |

Table 2  Results of semi-quantitative x-ray phase analysis of crystalline phases.

| Mineral   | Formula                  | Concentration, % |
|-----------|--------------------------|------------------|
| quartz    | SiO₂                     | 59.4             |
| mica      | KAl₂(Si₃O₁₀)(OH)₂        | 14.6             |
| chlorite  | (Mg, Fe)₃Al(Si₃Al)O₁₀(OH)₈ | 12.1             |
| KPS       | KAlSi₃O₈                 | 8.0              |
| PS (albite)| Na(AlSi₂O₆)             | 5.9              |
Table 3  Results of gamma spectrometric analysis.

| Spectrum         | Th-234, Bq/kg | Pa-234m, Bq/kg | Th-230, Bq/kg | Pb-214, Bq/kg | Bi-214, Bq/kg | Pb-210, Bq/kg | Ac-228, Bq/kg | Ra-224, Bq/kg | Pb-212, Bq/kg | Bi-212, Bq/kg |
|------------------|---------------|----------------|---------------|---------------|---------------|---------------|---------------|---------------|---------------|---------------|
| U-SOLUTION       | 1110±120      | 1430±140       | 1430±160      | 162±20        | 168±20        | 1160±120      | 5.5±0.8       | 12.0±2.1      | 7.3±0.8       | < 6.5         |
| U-SEDIMENT       | 3420±340      | 4270±430       | 5930±620      | 2390±230      | 2480±240      | 5300±500      | 28.8±4.1     | 32±11         | 26.3±3.2      | < 32          |

| Spectrum         | Ti-208, Bq/kg | K-40, Bq/kg | U-235, Bq/kg | Pa-231, Bq/kg | Th-227, Bq/kg | Ra-223, Bq/kg | Rn-219, Bq/kg | Pb-211, Bq/kg | Bi-211, Bq/kg |
|------------------|---------------|-------------|--------------|---------------|---------------|---------------|---------------|---------------|---------------|
| U-SOLUTION       | 2.4±0.2       | < 12.0      | 44.5±4.6     | 35.0 ± 5.8    | 52.3±5.1      | 48.5±6.4      | 61.2±7.5      | 58.8±6.7      | 67±10         |
| U-SEDIMENT       | 5.9±0.8       | 831±81      | 276 ± 25     | 295±55        | 177±24        | 169±48        | 230±50        | 200±37        | 231±25        |

Note: SOLUTION - liquid phase, SEDIMENT - solid phase.
4. Conclusion

The results showed the possibility of extracting radionuclides and prospects for the development of chemistry of actinides in Kazakhstan.

We proposed to extract the remains of radionuclides from the drilled wells and to improve the radiation safety of the contaminated area. We conducted a chemical analysis of the content of radionuclides in the ore material. The radionuclides U, Th, Pa, Ra, Rn were extracted, which can be used in industry and medicine.

(1) A technology has been developed for the extraction of radionuclides from the core of the uranium deposit.

(2) Isolated radionuclides Th-234, Th-230, Th-227, Pa-231, Pa-234 m, Ac-228, Ra-223, Ra-224, Rn-219.

References

[1] Patent US # 201301046698 Method of catalytic oxidation of $\text{U}^{4+}$ to $\text{U}^{6+}$ using a catalyst Muhamedzhan-1.

[2] Aibassov, Y. 2015. The Complexes of Uranium with DNA. Scientific & Academic Publishing, USA, 224.