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SOURCES OF SILICA IN PRODUCTIVE SOLUTIONS DURING IN-SITU URANIUM LEACHING

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Annotatsiya. Maqolada uranni yer ostida eritib olish jarayonidagi mahsuldor eritmalarda kremniyning paydo bo‘lish manbalari, uning kimyoviy shakllari va eritmadagi xatti-harakatlari tahlil qilingan. Kremniyning ortokremniy kislotasi, polikremniy kislotasi va kolloid shakllarda mavjudligi ko‘rib chiqilgan. Silikat va alyumosilikat minerallarining kislota ta’sirida parchalanishi natijasida kremniyning eritmaga o‘tishi, uning eruvchanligi hamda pH ga bog‘liqligi aniqlangan. Shuningdek, kremniyning uranni yer ostida eritib olish jarayoniga hamda sorbsiya bosqichiga salbiy ta’siri ko‘rsatib o‘tilgan. Natijada mahsuldor eritmalardagi kremniyning miqdorini kamaytirish texnologik jarayon samaradorligini oshirish uchun muhim ekani asoslangan.

Kalit so‘zlar: Ortokremniy kislotasi, mahsuldor eritmalar, uranni yer ostida eritmaga o‘tkazish, silikat minerallar, kolloid kremney, polikremniy kislotasi, pH ta’siri.

Аннотация. В работе рассмотрены источники появления кремнезема в продуктивных растворах подземного выщелачивания урана, его химические формы и поведение в растворе. Проанализировано существование

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кремнезема в виде ортокремниевой кислоты, поликремниевой кислоты и коллоидных форм. Показано, что основным источником кремния является разложение силикатных и алюмосиликатных минералов под действием сернокислотных растворов. Рассмотрено влияние pH среды на растворимость и процессы поликонденсации кремнезема. Отмечено отрицательное влияние кремнезема на процесс выщелачивания урана и сорбционные переделы, включая снижение скорости выщелачивания коффинита и отравление ионообменных смол. Сделан вывод о необходимости снижения концентрации кремнезема в продуктивных растворах для повышения эффективности технологического процесса.

Ключевые слова: Ортокремниевая кислота, продуктивные растворы, подземное выщелачивание урана, силикатные минералы, коллоидный кремнезем, поликремниевая кислота, влияние pH.

Abstract. This paper considers the sources of silica formation in productive solutions of in-situ uranium leaching, its chemical forms, and behavior in solution. The presence of silica in the form of orthosilicic acid, polysilicic acid, and colloidal silica is analyzed. It is shown that the main source of silicon is the decomposition of silicate and aluminosilicate minerals under the action of sulfuric acid solutions. The effect of pH on silica solubility and polycondensation processes is discussed. The negative impact of silica on uranium leaching and subsequent sorption stages is highlighted, including the reduction of coffinite leaching rate and poisoning of ion-exchange resins. It is concluded that reducing silica concentration in productive solutions is essential for improving the efficiency of the technological process.

Keywords: Orthosilicic acid, productive solutions, in-situ uranium leaching, silicate minerals, colloidal silica, polysilicic acid, pH effect.

INTRODUCTION.

In uranium mining, the in-situ leaching (ISL) method is widely used due to its high efficiency and economic advantages. In this technology, sulfuric acid-based solutions are injected into the ore-bearing layer to dissolve uranium minerals, and the resulting productive solutions are pumped to the surface. However, along with uranium, various additional components, including silicon compounds, also enter the solution. The presence of silica in productive solutions is associated with the acid-induced decomposition of silicate and aluminosilicate minerals, which can significantly affect technological performance.

Silica in solution can exist in the forms of orthosilicic acid, polysilicic acid, and colloidal silica. These forms vary depending on the physicochemical properties of the solution, particularly the pH. High concentrations of silicon compounds can reduce the uranium leaching rate and negatively affect the performance of ion-

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exchange resins during sorption. Therefore, identifying the sources of silica formation in productive solutions and controlling its concentration is an important scientific and practical issue.

This study analyzes the sources of silica in productive solutions, its main chemical forms, and its impact on the in-situ leaching of uranium.

In this section, it is first necessary to define the terminology used. In productive solutions (productive leaching solutions) of underground uranium leaching, silicon can be present simultaneously in various forms and states. Therefore, in accordance with the recommendations of the most representative work on this topic – the monograph by R.K. Ayler – for convenience and simplicity, the term "silica" will be used to denote silicon dioxide SiO_2 in all its forms (crystalline and amorphous, anhydrous, hydrated, and hydroxylated) [1].

The main forms of silica in solution are generally considered to be:

– Orthosilicic acid, which, depending on its properties in a given case, can be represented as H_4SiO_4 or as silicon tetroxide $Si(OH)_4$;

– Polysilicic acid (oligomers) with a molecular weight of up to 100 kDa and particle sizes up to 5 nm;

– Colloidal silica with a molecular weight greater than 100 kDa and particle sizes greater than 5 nm.

The boundary between oligomers and colloids is arbitrary and based on general observations – below this threshold, polymers are usually unstable due to gelation or particle growth processes [2].

The dissolution and precipitation of silica in water involves hydration and dehydration chemical reactions catalyzed by hydroxyl ions:

$$(SiO_2)_x + 2H_2O \underset{\text{дегидратация}}{\overset{\text{гидратация}}{\rightleftarrows}} (SiO_2)_{x-1} + Si(OH)_4 \qquad (1)$$

Therefore, the solubility of silica in water depends on the composition of the substrate phase and is determined by the heat of hydration: at 25 °C, above partially hydrated amorphous silica it ranges from 100 to 130 mg/L (depending on the degree of hydration), above anhydrous amorphous silica (quartz glass) it is 70 mg/L, while above crystalline quartz it is only 6 mg/L [3]. According to data (2), the thermal effect of silica hydration reactions is:

$$Si(OH)_4 = SiO_2(\text{кварц}) + 2H_2O + 22{,}84\text{кДж} \qquad (2)$$

$$Si(OH)_4 = SiO_2(\text{аморф.}) + 2H_2O + 15{,}51\text{кДж} \qquad (3)$$

Calculations using the Nernst equation for equations (2) and (3) give silica solubility values of 0.0998 mM = 5.995 mg/L and 1.919 mM = 115.3 mg/L, respectively, which fully coincide with the data in (3). As a result of the ongoing dissolution and reprecipitation reactions in natural waters, the concentration of

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orthosilicic acid almost always falls within this range (on average, seawater contains 6.2 mg/L of silica, river water – 13.1 mg/L [4]; and the maximum permissible concentration for drinking water allows up to 21.4 mg/L).

However, these are average values: evidently, above the surface of partially leached uranium-rich deposits (URD) saturated with silicic acid, the concentration can increase up to 115 mg/L, while in clay-free quartz areas of sandstone it decreases to 6 mg/L (with silica reprecipitating on quartz in crystalline form). The equilibrium concentration of silicic acid above silicate minerals is closer to the lower limit: for example, above montmorillonite, according to [5], it is 15 mg/L. In particular, in most roll-type deposits in Uzbekistan, the concentration of orthosilicic acid in groundwater ranges from 13 to 30 mg/L.

It should be noted that the solubility of orthosilicic acid in its molecular form is practically independent of the pH of the medium. However, in alkaline conditions, silica solubility increases sharply due to the formation of $H_3SiO_4^-$ anions (and in strongly alkaline media – $H_2SiO_4^{2-}$; for orthosilicic acid, $pK_a1 = 9.7$, $pK_a2 = 11.7$). At concentrations of 100–130 mg/L and below, orthosilicic acid, over any pH range, is not prone to polymerization (more precisely, polycondensation) or gelation. At higher concentrations, in the presence of a solid phase, precipitation onto the substrate occurs, whereas in its absence, polycondensation takes place, with the rate being minimal at the isoelectric point (pH = 2.0–2.2), values quite close to the pH of productive solutions of underground uranium leaching.

At pH < 2, the polycondensation rate is proportional to the concentration of hydrogen cations, while at pH > 2.2 it is proportional to the concentration of hydroxyl anions. Near the isoelectric point, slightly supersaturated solutions of orthosilicic acid are very stable, and even highly supersaturated solutions do not polymerize instantaneously: for example, a solution with 60-fold supersaturation begins polymerizing after 2–3 hours, reaching only the dimer – dihydrosilicic acid $H_6Si_2O_7$ (DHA) – within a day. A solution with 17-fold supersaturation polymerizes 50% within a week, and a three-fold supersaturated solution begins polymerizing only after a month.

According to analysis data, at the Loylikent deposit, productive solutions contain 92 mg/L of silicon (corresponding to 197 mg/L of silica), i.e., the degree of supersaturation is only 1.64. This suggests that the silica is present exclusively as orthosilicic acid and DHA, especially since standard silica analysis (reaction with ammonium-molybdate reagent) identifies silica solely in these two forms [6].

During underground uranium leaching, sulfuric acid solutions actively react with silicates and aluminosilicates present in the ore mass. As a result of this interaction, ions of various metals, primarily sodium, potassium, magnesium,

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calcium, iron, and aluminum, are released into the solution in the form of their corresponding sulfates [7].

The behavior of silicon in these processes is largely determined by the structural features of silicate minerals. Based on their resistance to acid attack, three groups of minerals are distinguished [8]:

minerals that decompose forming non-gel silica;

minerals that are practically resistant to acids (acid-resistant);

minerals that dissolve forming silicic acid gel.

Multiple studies aimed at establishing a correlation between silicate solubility and their chemical composition, crystal lattice structure, or energetic parameters have not led to unequivocal conclusions. Nevertheless, they allowed the identification of several specific patterns [9]. In particular, it is known that feldspars belong to the first group (acid-resistant). Clay minerals, which are layered silicates, belong to the second group (after dissolution of vermiculite, biotite, and hydro-micas, a very fine white “skeleton” of silica remains, preserving the platy shape of the particles; similarly, after asbestos dissolution, fibrous silica forms in the residue, etc.) [10].

On the other hand, island silicates with isolated tetrahedra (coffinite, as well as olivine $(Mg,Fe)_2SiO_4$, larnite Ca_2SiO_4, monticellite $CaMgSiO_4$), as well as aluminosilicates with an atomic Si/Al ratio ≤ 2 (e.g., nepheline $(Na,K)AlSiO_4$ and anorthite $Ca[Al_2Si_2O_8]$)) belong to the third group, dissolving to form highly supersaturated solutions of orthosilicic acid (up to 100 g/L as silica) [11]. In these solutions, gelation in the region of the isoelectric point occurs only after several hours or even tens of hours (Fig. 1).

It is evident that the relatively low silica content in productive underground uranium leaching solutions is primarily explained by the fact that the abundance of these minerals in the ore mass is low [12].

Let us consider how silica affects the uranium leaching process during underground uranium leaching and subsequent processing steps. The main uranium minerals in roll-type deposits are nasturan and coffinite, which are leached by sulfuric acid in the presence of an oxidizer – ferric sulfate (Fe^{3+}), according to the equations:

$$UO_{2+X} + xH_2SO_4 + (1-x)Fe_2(SO_4)_3 = UO_2SO_4 + (2-2x)FeSO_4 + xH_2O \quad (4)$$

$$U(SiO_4)_{1-X}(OH)_{4X} + Fe_2(SO_4)_3 = UO_2SO_4 + 2FeSO_4 + (1-x)H_4SiO_4 + 2x\,H_2O \quad (5)$$

It is evident that the presence of orthosilicic acid in the solution does not affect the progress of reaction (4). However, it significantly influences reaction (5): according to Le Chatelier–Braun principle, an increase in the concentration of

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orthosilicic acid in productive solutions leads to a decrease in the uranium leaching rate from coffinite.

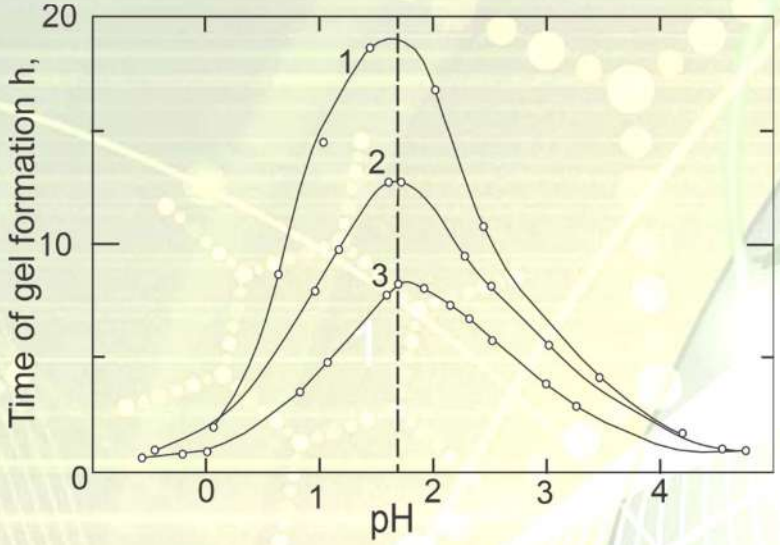


Fig. 1. Dependence of gelation time on pH for silica sols prepared from sulfuric acid and sodium silicate at different silica concentrations in the sol (according to [13]: 1 – 1.08 M (65.4 g/L); 2 – 1.33 M (79.8 g/L); 3 – 1.78 M (106.8 g/L)).

It is possible that the positive effect of the presence of fulvic acids in leaching solutions was partly due to their ability to complex with silica, thereby reducing its activity in the solution.

The most harmful effect of silica is observed on anion-exchange resins, as it accumulates in their pores and "poisons" them. For example, when the silica content in the resin reaches 10%, the uranium capacity of the resin decreases by 10% [14], while an increase in silica content to 19.6% reduces the uranium capacity by 32% [15]. Current methods for silica removal either cause a significant loss of mechanical strength of the resin (alkaline method) or are inefficient, achieving only a low percentage of silica removal (fluoride method) [16].

L.I. Vodolazov and B.N. Laskorin, along with colleagues, summarized the limited available literature on this issue and identified certain regularities in the silica removal processes [17]. However, this area of applied hydrometallurgy remains one of the least developed. The mechanism of this phenomenon is still unclear, as silica removal occurs both on cation- and anion-exchange materials (to a greater extent on the latter).

It can be hypothesized that silica can adsorb on ion-exchange resins when present in the form of sulfohumic acid, since it is known that their close analogs—lignosulfonic acids—can be adsorbed both on anion- and cation-exchange resins [18]. This hypothesis is supported by the fact that silica, like sulfohumic acids, can

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partially desorb from resins, contaminating the product desorbate and causing processing problems [19].

Thus, silica has a negative effect both on the in-situ uranium leaching process itself, by reducing the leaching rate of coffinite, and on subsequent processing stages, by poisoning ion-exchange resins and contaminating the product desorbate. Therefore, reducing the concentration of silica in productive solutions of in-situ uranium leaching is a highly relevant task. While it is advisable to remove organic matter from productive solutions before the sorption stage—due to its process-enhancing effect—and then return it to the circulation tanks to intensify uranium leaching, silicon must be removed completely. Otherwise, if silicon-containing organics are returned to the circulating leaching solutions, the problem of silica contamination arises.

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