ISNN 3030-3907

2026
MAY
ISSUE - 5
VOLUME - 2

DEVELOPMENT OF SCIENCE

scientific journal



Development of science

Ilmiy jurnal

MAY 2026
Issue - 5, VOLUME - 2

ISSN 3030 -3907

Ilmiy jurnal OAK rayosatining 2024 yil 27 sentabrdagi 361-son qaroriga asosan 02.00.00 kimyo fanlari hamda 2025 yil 12 fevral 367- son qaroriga asosan 05.00.00 texnika fanlari bo‘yicha falsafa doktori (PhD) va fan doktori (DSc) ilmiy darajasiga talabgorlarning dissertatsiya ishlari yuzasidan dissertatsiyalari asosiy ilmiy natijalarini chop etish tavsiya etilgan milliy ilmiy nashrlar ro‘yxatiga kiritilgan.

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ISSN 3030-3907

Development of science ilmiy jurnali 2024 yil 15 aprel kuni №256445 sonli guvohnoma bilan Davlat ro‘yxatidan o‘tgan.

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ANALYSIS OF METHODS FOR REMOVING SILICA FROM URANIUM IN-SITU LEACHING PRODUCTIVE SOLUTIONS

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Annotatsiya. Mazkur maqolada uran ishlab chiqarish eritmalaridan kremniy (silika)ni ajratib olish usullari tahlil qilingan. Eritmalarda kremniy asosan ortokremniy kislotasi va uning dimer shakllarida, shuningdek suspenziya va sorbsiya holatida mavjudligi ko‘rsatib o‘tilgan. Tadqiqotda kremniyni ajratishning asosiy usullari — koagulyatsiya, flokulyatsiya, membrana texnologiyalari va sorbsiya jarayonlari solishtirma tahlil qilingan. Ayniqsa, ortokremniy kislotasini past konsentratsiyalarda ajratib olish muammolari yoritilgan. Geterokoagulyatsiya jarayonida alyuminiy va temir (III) gidroksidlari bilan birikmalar hosil bo‘lishi yuqori samaradorlikka ega ekani asoslab berilgan. Shuningdek, kremniy va organik moddalarni birgalikda cho‘ktirish, ularni keyinchalik ajratish hamda kristallanish jarayonlaridan foydalanish istiqbollari ko‘rib chiqilgan. Tadqiqot natijalari yer osti uranni eritib olish texnologiyalarini takomillashtirish uchun muhim ahamiyatga ega.

Kalit so‘zlar: kremniy, ortokremniy kislotasi, uran eritmalari, yer osti eritib olish, geterokoagulyatsiya, sorbsiya, flokulyatsiya, koagulyatsiya, alumosilikatlar, temir (III) gidroksidi, organik moddalar, kristallanish.

Аннотация. В данной работе проведён анализ методов удаления кремния (силики) из продуктивных растворов уранового производства. Показано, что кремний в растворах присутствует преимущественно в виде ортокремниевой кислоты, её димеров, а также в форме суспензий и сорбированных соединений. Рассмотрены основные методы удаления кремния, включая коагуляцию, флокуляцию, баромембранные процессы и сорбцию. Особое внимание уделено проблеме удаления ортокремниевой кислоты при низких концентрациях. Обоснована высокая эффективность гетерокоагуляции с гидроксидами алюминия и железа (III), приводящей к образованию труднорастворимых алюмосиликатных и ферросиликатных соединений. Также рассмотрены процессы совместного осаждения кремния и органических веществ, их последующее разделение и возможности кристаллизации осадков. Полученные результаты имеют важное значение для совершенствования технологий подземного выщелачивания урана.

Ключевые слова: кремний, ортокремниевая кислота, урановые растворы, подземное выщелачивание, гетерокоагуляция, сорбция, флокуляция, коагуляция, алюмосиликаты, гидроксид железа (III), органические вещества, кристаллизация.

Abstract. This paper presents an analysis of methods for removing silica from uranium production solutions. It is shown that silica in such solutions exists mainly in the form of orthosilicic acid, its dimers, as well as in suspended and sorbed forms. The main silica removal techniques, including coagulation, flocculation, baromembrane processes, and sorption, are comparatively analyzed. Particular attention is given to the challenges of removing orthosilicic acid at low concentrations. The high efficiency of heterocoagulation with aluminum and iron (III) hydroxides is substantiated, resulting in the formation of poorly soluble aluminosilicate and ferrosilicate compounds. The study also explores the co-precipitation of silica and organic matter, their subsequent separation, and the potential of crystallization processes. The results obtained are significant for improving in-situ uranium leaching technologies.

Keywords: silica, orthosilicic acid, uranium solutions, in-situ leaching, heterocoagulation, sorption, flocculation, coagulation, aluminosilicates, iron (III) hydroxide, organic matter, crystallization.

INTRODUCTION.

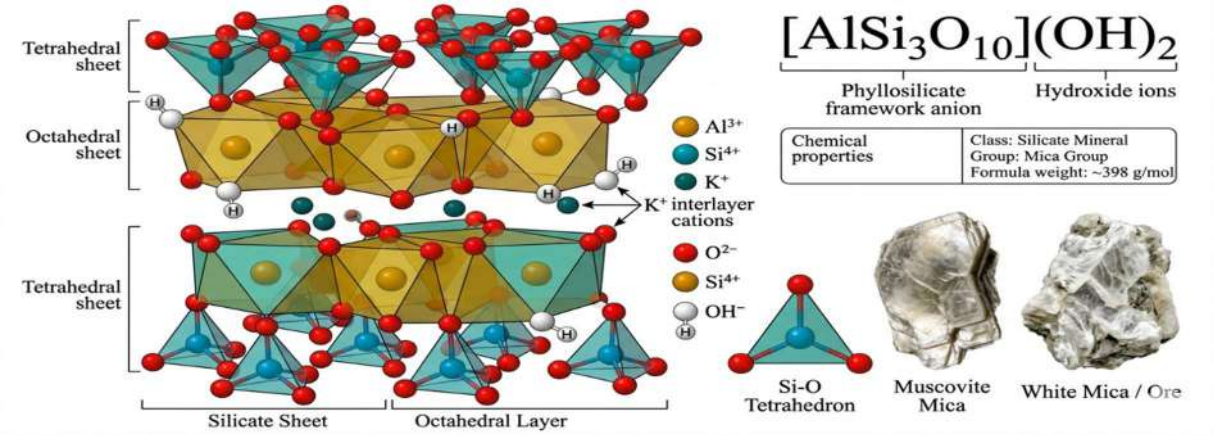
Analysis of Methods for Removing Silica from Uranium Production Solutions. Silica, as determined by analysis in the composition of productive solutions, is present in the form of a monomer (orthosilicic acid) and a dimer (DCC), since the standard analysis for silica (reaction with ammonium molybdate reagent) detects it exclusively in these two forms[1]. However, it is highly likely

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that a significant portion of it, not accounted for by this analysis, is present in the product solutions as suspensions and/or sorbates on organic matter.

Studies of deposits[2] indicate that uranium in the groundwater of the Tian Shan Basin occurs not only in the form of uranyl carbonate compounds, but also as an adsorbed layer on aluminosilicate hydrosols similar in composition to hydromicas. This explains the high silica content in carbonized plant detritus and, consequently, the high coffinite content in most domestic roll-type uranium deposits. At the same time, the data from[3] cited in Chapter 1 regarding the global average silica content in river water at 13.1 mg/L refer to orthosilicic acid. In the form of suspensions, it contains an additional 250 mg/L of silica and 38.2 mg/L of aluminum[4], which corresponds to an atomic ratio of Si/Al = 3.

This suggests that, in terms of composition, they are also similar to hydromicas, whose aluminosilicate framework is formed by the anion $[AlSi_3O_{10}](OH)_2$ in picture 1.



Picture 1. Layered structure and chemical formula of muscovite mineral.

Given that leaching solutions are typically prepared using formation water, it can be expected that productive solutions will contain significant amounts of silica in the form of suspensions. It should be noted that their presence in production solutions does not have a negative impact on the underground uranium leaching process, since the dissolution of ionites (as well as the vast majority of other organic compounds) is caused only by orthosilicic acid (and, possibly, DCC)[5]; however, when selecting a method for removing silica from productive solutions, the presence of these suspensions should be taken into account, and their behavior during the purification process should be analyzed.

A large number of studies have been devoted to the removal of silicic acid from solutions, spanning a wide range of scientific disciplines (as is the case with humic and fulvic acids). In particular, the problem of leaching silica from silicates arises in many other areas of metallurgy, not only in underground leaching of uranium and thorium, but also in agitation leaching of concentrates, where a certain amount of silicates remains as an unavoidable impurity (tailings). The

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main methods for silica removal are coagulation (including heterocoagulation), flocculation, baromembrane methods, and sorption[6]. Sometimes they are used in combination—for example, in baromembrane purification methods, the higher the MM value of the material being removed, the higher the efficiency—therefore, the most effective approach is to sequentially perform coagulation or flocculation of silica (to increase the MM of its aggregates) followed by ultrafiltration. Undoubtedly, these methods are highly effective for removing silica suspensions from production solutions—but by no means from orthosilicic acid. At the same time, as noted, at concentrations below 100–130 mg/L, orthosilicic acid is not prone to polycondensation or gel formation in any pH range, so the use of any flocculants will not reduce the orthosilicic acid content below the specified value. Obviously, for the conditions at the "Loyliken" deposit, this would mean a reduction in the initial orthosilicic acid concentration by only 40–50%, which is clearly insufficient. At the same time, in the presence of the solid phase, orthosilicic acid is deposited onto the substrate, and the residual concentration of orthosilicic acid is determined by the composition of the substrate—as noted in Chapter 1, this is 6 mg/L for quartz and 15 mg/L for montmorillonite[7]. The problem is that deposition onto the substrate is not a very fast process, and a large specific surface area of the substrate is required for it to proceed effectively. Thus, according to practical data, it is recommended to introduce a powdered acid-resistant substance (quartz sand or clay) with a particle diameter of no more than 0.06 mm into a silica-containing solution in an amount that ensures a total particle surface area of at least 0.3 m^2 per 1 g of SiO_2. An important advantage of this method is its suitability for use in highly acidic environments (up to 2.5 M nitric acid). It can be assumed that the precipitation rate would increase significantly in the presence of fluoride ions, which are among the most effective catalysts for silica polymerization and gelation, even at pH values close to the isoelectric points[8]. However, this method is not suitable for uranium in-situ leaching solutions due to the presence of aluminum and iron (III) ions in the solutions, which bind fluoride ions into a stable, inactive complex[9].

It seems that sorption-based purification methods—primarily using readily available, non-regenerable sorbents such as cellulose, clays of various compositions, zeolites, silica gel, iron and aluminum hydroxides, and others—share a similar mechanism of action[10]. Due to their versatility, these methods are widely used in various industries to remove silicon from solutions with different chemical compositions and pH levels. Therefore, it is more practical not to develop a new sorbent for the purification of productive solutions from underground uranium leaching, but to select the most effective one from those already known and adapt the technology to the specifics of their chemical

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composition, in particular taking into account the presence of organic compounds (this aspect will be discussed in more detail below)[11].

Another interesting and promising method for removing orthosilicic acid from solutions is heterocoagulation (i.e., co-precipitation) of orthosilicic acid with hydroxides of multivalent metals—for example, with aluminum and iron(III) hydroxides, orthosilicic acid forms, respectively, aluminosilicic and ferrosilicic acids[12]:

$$Al(OH)_3 + H_4SiO_4 = HAlSiO_4 \downarrow + 3H_2O \quad (1)$$

$$FeOOH + H_4SiO_4 = HAlSiO_4 \downarrow + 2H_2O \quad (2)$$

These acids have very low solubility, yet they are much stronger than orthosilicic acid (for example, fresh neutral sols of $Si(OH)_4$ and $Al(OH)_3$ produce a strongly acidic reaction when mixed), and tend to form stable complexes with alkaline earth metal ions, forming even less soluble compounds—for example, synthetic epidote $Ca_2Al_2Fe^{3+}(SiO_4)_3OH$ was detected in the precipitate formed upon neutralization of a commercial desorbate from uranium production to pH 3.0–3.5[13].

The exothermic effect of aluminosilicic acid formation is so significant that orthosilicic acid is capable of binding not only hydrolytically precipitated aluminum but also dissolved aluminum at pH 3.1—that is, at a pH one unit lower than the pH at which the hydrolysis of aluminum salts begins:

$$Al^{3+} + H_4SiO_4 = HAlSiO_4 + 3H^+ + 48{,}2kJ/mol \quad (3)$$

The heat effect of ferrosilicate formation is significantly lower, but the tendency of iron (III) to hydrolyze is higher—therefore, iron (III) from a 0.1 M solution is bound by orthosilicic acid at pH = 1.83, i.e., 0.14 units lower than the pH at which iron (III) salts begin to hydrolyze. This allows the same technology to be successfully used for the removal of orthosilicic acid as for the removal of organic matter from productive solutions of underground uranium leaching. If iron (III) is regenerated without prior acidification of the circulating leaching solutions, due to acid consumption according to the equation[14], the pH of the solution increases and orthosilicic acid begins to bind with iron (III), and ferrosilicic acid even before the precipitation of hydroxyetite begins according to the equation and its binding of fulvic acid and sulfohumic acid. This allows the process to be conducted in two stages, with the transition between stages controlled by a sudden change in pH: in the first stage, ferrosilicate is precipitated, forming a tailings cake, and in the second stage, hydroxyetite precipitates with organic matter, forming a return cake that is to be redissolved in sulfuric acid and combined with the return leaching solutions after sorption processing in order to restore both iron (III) and organic matter useful for underground uranium leaching to its composition.

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The fact that silica and organic matter are precipitated by the same reagent—iron (III)—in this case draws attention to the fact that, when using most of the proposed technologies for removing organic matter from production solutions, orthosilicic acid will be precipitated along with the organic matter, and in those technologies where orthosilicic acid is not precipitated, the organic matter will contain a large amount of silicon in the form of silicon and organic complexes. Since organic substances are planned to be returned to the underground uranium leaching process, and the presence of silica has a negative impact both at this stage and in subsequent processing stages, there is a clear need for the preliminary removal of silica from the organic matter before its reuse[15].

One promising approach in this regard appears to be the crystallization of precipitated silica. The fact is that silica's tendency to crystallize during heterocoagulation is much higher than during the coagulation of pure silica[16]—for example, in nature, the coagulation of silicic acid forms amorphous opal, whereas in the presence of small amounts of iron (III) and/or aluminum (up to 0.3% and 0.8%, respectively), the precipitate rapidly crystallizes to form microcrystalline quartz—the mineral chalcedony. If, however, the iron content in the precipitate ranges from 5 to 15%, the number of crystallization nuclei increases to such an extent that cryptocrystalline quartz forms—the mineral jasper, in which, due to the very high rate of nucleation, the crystal size is an order of magnitude smaller than in chalcedony. In particular, if organic matter and silica are sorbed onto metal hydroxides, it is possible to separate them subsequently—for example, if a spent bauxite filter is immediately washed with alkali, both the organic matter and the silica will be desorbed from it. However, if it is allowed to stand for a period sufficient for silicic acid to crystallize, and then washed with alkali, one can expect that the organic matter will be washed out in practically the same volume, while almost all of the silica will remain on the filter. If a limonite filter is used, it is advisable not to regenerate it after use, but to dissolve it in sulfuric acid, send the insoluble fraction (silica) to the waste pile, and return the iron (III) sulfate solution to the circulating leaching solutions as an oxidizing agent. Moreover, the organic matter present in the solution will serve as an intensifier of the underground uranium leaching process.

As noted above, in the pH range close to the isoelectric point, orthosilicic acid polymerizes very slowly. However, the reverse is also true: polymerized silicic acid depolymerizes at the slowest rate upon dilution within the same pH range — i.e., if a mixture of organic matter and a silicic acid gel is redissolved in a medium with pH ≈ 2, the organic matter will dissolve rapidly, while the silica will remain almost entirely in the precipitate.

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