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Oxidation kinetics of tetravalent uranium during sulfuric-acid leaching intensification under in-situ recovery conditions

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Abstract

The paper examines how redox phenomena govern the recovery of uranium by in-situ recovery (ISR) technology, with particular emphasis on the role of oxygen and iron in the controlling reactions. A review is presented of the standard electrode potentials of the ionic species participating in the process, followed by a detailed examination of the conversion of Fe(II) to Fe(III) and its effect on uranium dissolution in acidic media. The experimental section addresses the oxygenation of acidic lixiviants through a Venturi-type nozzle. The findings demonstrate that tuning the redox conditions markedly enhances the productivity of ISR. Atmospheric oxygen, owing to its availability and cost efficiency, drives the Fe(II) → Fe(III) transition and thereby raises the solubility of uranium-bearing species. Through experiment, the flow velocity providing the maximum oxygen dissolution was identified. Incorporation of the Venturi nozzle substantially increased the dissolved-oxygen content of the lixiviant, which in turn raised the Fe(III) concentration and improved uranium recovery. The proposed approach yielded a 38.13 % increase in uranium extraction relative to the conventional procedure. The work confirms the importance of redox processes in uranium hydrometallurgy and justifies the need to optimize them for industrial gains. A Venturi nozzle embedded in an acidic recirculation loop, operated without any externally supplied oxidant, sustains an Eh elevation sufficient for the U(IV) → U(VI) transition through enhanced oxidation of Fe^{2+} to Fe^{3+}. The study quantitatively documents a ~60-70 % rise in dissolved O_2, an Eh increment of ~30-80 mV, and a corresponding gain in dissolved uranium.

Keywords

uranium leaching; in-situ recovery; redox processes; ferric ions (III); oxygen saturation; Venturi nozzle

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Introduction

Redox reactions underlie a wide range of chemical transformations and are particularly central to the in-situ recovery (ISR) of uranium. A high value of the standard redox potential (E°) signals a strong tendency to oxidize, which is decisive for uranium dissolution. The present study considers the principal oxidation-reduction reactions and the factors that govern their efficiency, with a focus on the practice of uranium ISR in Kazakhstan [1-3].

Lixiviation of uranium from ore is a key stage of its industrial production and is closely linked to the energy needs of modern society. The performance of this step depends on many variables, among which the choice of oxidant is critical for dissolving uranium minerals. In this

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respect, atmospheric oxygen stands out as the most accessible and economically reasonable oxidant [4-6].

Unlike hydrogen peroxide, oxygen can be obtained directly from ambient air at negligible cost, which makes it the preferred reagent for ISR. It enters the oxidation cycle by converting ferrous iron (Fe^{2+}) into the ferric form (Fe^{3+}). This conversion not only raises the oxidative capacity of the solution but also markedly increases the solubility of uranium – a prerequisite for efficient recovery from the ore.

The kinetics and thermodynamics of redox reactions in solution likewise influence leaching efficiency. Parameters such as pH, temperature, and oxygen content shape the rate and direction of these reactions, and their optimization is what allows the uranium recovery to be substantially raised [7, 8].

In the present work particular attention is paid to the function of oxygen in uranium leaching and to the influence of various factors on the redox reactions involved. Separately, the use of a Venturi nozzle as an innovative means of saturating the lixiviant with oxygen and thus intensifying the process is considered.

In uranium hydrometallurgy, especially under Kazakhstani conditions where ISR is widely deployed, the conversion of ferrous to ferric iron plays a pivotal role. The presence of Fe^{2+} in the lixiviant is the norm, and its conversion to Fe^{3+} is necessary to obtain a high uranium recovery [9-11].

In the presence of Fe^{3+} ions, complexes form with uranium that markedly increase its solubility in acidic media. Provided the ferric-iron concentration is sufficient, uranium remains in solution and is recovered efficiently. In Kazakhstani deposits, where the ores carry substantial amounts of Fe^{2+}, the oxidation of this iron to Fe^{3+} becomes one of the most important stages of the leach. Fe^{3+} not only enhances uranium solubility but also takes part in reactions that stabilize the dissolved uranium species, so that uranium remains in the dissolved form even when pH fluctuates.

The rate of the $Fe^{2+} \rightarrow Fe^{3+}$ conversion depends on the oxidant concentration, the temperature, the pH, and the presence of competing anions. At low temperature or at high Fe^{2+} load, the oxidation can proceed slowly, which slows the overall uranium leach.

Under such conditions, active oxidants such as hydrogen peroxide (H_2O_2) or oxygen (O_2) become necessary. They deliver enough oxidative capacity to drive Fe^{2+} to Fe^{3+} at a useful rate and thereby accelerate uranium dissolution. Among them, atmospheric oxygen remains the most accessible and economically attractive reagent. In contrast to hydrogen peroxide, which is effective but considerably more expensive, oxygen is obtained directly from the surrounding atmosphere at no additional cost. This makes O_2 the natural choice for ISR operations [12-14].

The aim of this work is the experimental justification of using a Venturi-type nozzle to intensify the oxidative stages of sulfuric-acid ISR of uranium. Special consideration is given to the effect of aeration on the $Fe^{2+} \rightarrow Fe^{3+}$ and U(IV) → U(VI) transitions, on the Eh and dissolved-oxygen content, and on the resulting increase in uranium concentrations in the pregnant leach solution.

Methods

Within the framework of this study, a bench-scale rig reproducing the uranium ISR process was designed and assembled. The rig allowed the principal stages of the technology to be replicated under controlled conditions, which made it possible to examine in detail the features and performance of the procedure previously described in the doctoral dissertation [15].

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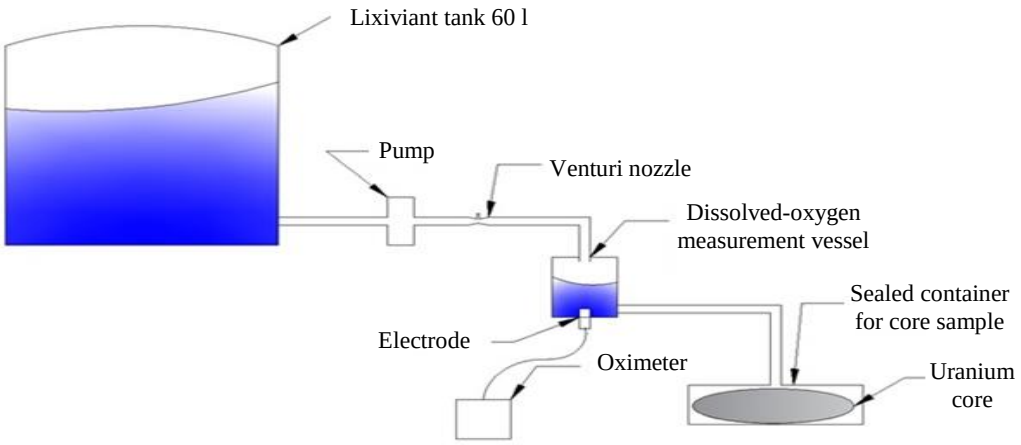


Fig.1. Schematic of the laboratory rig

Standard redox potentials E° serve to gauge the ability of a given species to act as an oxidant or a reductant in a chemical reaction. The higher the value of E°, the stronger the oxidizing ability of the substance. This information is important for understanding reactivity in uranium leaching, where efficient metal recovery from the ore requires the strict observance of specific conditions [16, 17].

A detailed treatment of the standard E° values and their meaning in the context of electrochemical reactions is given below:

$$UO_2^{2+} + 2e^- \rightleftarrows UO_2,\ E° = 0.221\ V.$$

The uranyl ion (UO_2^{2+}) accepts two electrons and is reduced to uranium dioxide (UO_2). This indicates that UO_2^{2+} is a relatively weak oxidant:

$$Fe^{2+} \rightleftarrows Fe^{3+} + e^-,\ E° = 0.771\ V.$$

The oxidation of Fe^{2+} to Fe^{3+} is a one-electron process and is reversible – the Fe^{2+} ion releases an electron and acts as a reductant. Fe^{3+} is a stronger oxidant than the uranyl ion:

$$NO_3^- + 4H^+ + 3e^- \rightleftarrows NO + 2H_2O,\ E° = 0.96\ V.$$

In acidic media the nitrate ion can accept three electrons and is reduced to nitric oxide. The nitrate ion is an even stronger oxidant than Fe^{3+}:

$$HNO_2 + H^+ + e^- \rightarrow NO + H_2O,\ E° = 0.99\ V.$$

Nitrous acid (HNO_2) in acidic media accepts one electron and is reduced to nitric oxide (NO) and water. With a standard potential E° = +0.99 V it behaves as a moderately strong oxidant, comparable in strength to the nitrate ion (NO_3^-):

$$MnO_2 + 4H^+ + 2e^- \rightleftarrows Mn^{2+} + 2H_2O,\ E° = 1.23\ V.$$

Manganese dioxide (MnO_2) in acidic media accepts two electrons and is reduced to Mn^{2+}. It is a fairly strong oxidant:

$$MnO_4^- + 8H^+ + 5e^- \rightleftarrows Mn^{2+} + 4H_2O,\ E° = 1.52\ V.$$

The permanganate ion (MnO_4^-) is one of the strongest oxidants in acidic media, accepting up to five electrons.

The bench-scale rig consists of three principal elements (Fig.1). The first is a tank for the leaching agent (LA): it is made of chemically resistant plastic and has a capacity of 60 l, which provides

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enough fluid for the experimental runs. The second element is a sealed vessel filled with drill-core material reproducing the natural ore environment of uranium deposits. The third is a dedicated chamber in which the uranium-bearing solution is collected for analysis. Compared with the configuration originally described in [15], the principal layout of the aeration and circulation node is retained; the present work differs in the volume of the LA tank, which has been raised to 60 l.

To verify the starting composition of the ore, an elemental analysis was performed on composite technological sample 1T, assembled from partial samples taken at well 3023. The uranium content was 0.13 %, radium 0.09 %, and the total iron 1.69 % (of which Fe^{2+} = 0.94 % and Fe^{3+} = 0.75 %); the carbonate content recalculated to CO_2 was below 0.1 %. Granulometrically, the sample is a fine-to-medium-grained sand, with the fraction below 0.1 mm accounting for 17.5 %, including ~10.5 % silt-clay material. Mineralogically, quartz dominates (up to 75 %), with feldspars (~5 %) and subordinate mica and chlorite. Significant quantities of colloidal iron hydroxides, iron sulfides (~1.5 %), and carbonized organic matter (~3.3 %) are also present. Among the clay minerals montmorillonite prevails, with smaller amounts of kaolinite. The uranium mineralization is represented by uranium oxide.

To circulate the liquid, a sixty-centimeter flexible hose made of a chemically resistant elastomer was connected to the tank. A horizontal pump, model 15WG-90A (rated power 90 W), was installed in line to deliver the required flow rate of the leaching agent. It develops the necessary head at a nominal throughput of 8 l/min, allowing the liquid to pass through the Venturi nozzle – a device that accelerates the flow and promotes efficient dissolution of atmospheric oxygen into the lixiviant. This reproduces the oxidative reactions that occur during uranium leaching. The arrangement is based on the configuration tested previously [15] and has been adapted in the present work to the chosen pump throughput and tank volume [18-20].

Downstream of the Venturi nozzle, the oxygen-enriched liquid enters a separate vessel designed specifically for measurement of the dissolved-oxygen content. The vessel is purpose-built: it is sealed by a lid at the top that prevents gas loss and maintains stable internal conditions, with a small inlet in the lid through which the air-water mixture enters. A highly sensitive portable oximeter with a remote electrode 8403 is installed in the lower part of the vessel; it measures dissolved oxygen over the range 0-19.99 mg/l with a precision of ±1.5. This arrangement provides the accurate readings required to evaluate the performance of the leaching process.

The experimental runs were carried out with a lixiviant containing 20 g/l of sulfuric acid. The oxidation-reduction potential (ORP) was measured with an IT-1101 meter. The uranium content of the pregnant leach solution was determined titrimetrically following the standard procedure "Uranium-bearing process solutions. Determination of uranium mass concentration by the ferrous-phosphate-bichromate method". The procedure relies on titration with ferrous-ammonium alum and potassium bichromate, has a detection limit of about 0.5 mg/l and a relative error of ±2-3 %, and provides reliable measurement of uranium in the range 10-30 mg/l. To confirm the reliability of the data, a fraction of the samples was additionally analyzed by inductively coupled plasma mass spectrometry (ICP-MS); the discrepancy between the two methods did not exceed 5 %. The duration of leaching was 9 h and the total volume of the lixiviant was 12 l. Pregnant leach solution aliquots were collected hourly throughout the run, giving a total of 10 baseline samples (untreated lixiviant from the core) and 10 samples after the lixiviant had been oxygenated.

Oxygen plays the central part in converting iron from the divalent to the trivalent state, which in turn drives uranium into solution. Molecular oxygen reacts with iron ions and raises their oxidation state.

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During the oxidation reactions energy is released, which additionally promotes uranium dissolution. These reactions require a certain activation energy that can be reached either by raising the temperature or by adjusting the pH of the medium.

The rate of uranium dissolution depends on the oxidant concentration. A higher oxygen content increases the dissolved uranium in solution. Above a certain threshold, however, oxygen begins to escape as free gas, which lowers leaching efficiency. Thus, at an oxygen concentration of 0.1 mol/l the solubility of uranium can rise by 50 %, whereas above 0.2 mol/l free-gas formation begins and the solubility decreases.

Redox transformations are the basis of uranium leaching. Oxygen and Fe^{3+} are the principal species that govern its efficiency. Understanding the kinetics and thermodynamics of these reactions and optimizing the conditions under which they occur are decisive for raising the productivity of uranium hydrometallurgy [21-23].

The oxidation of iron can be represented as:

$$Fe^{2+} \rightarrow Fe^{3+} + e^{-}.$$

The standard electrode potential of this reaction is +0.771 V, which means that only a small energy input is required for it to proceed. In the present case the principal oxidant is the oxygen dissolved in water. The oxygen half-reaction can be written as:

$$O_2 + 4H^{+} + 4e^{-} \rightarrow 2H_2O.$$

Combining the two half-reactions yields the overall redox equation:

$$4Fe^{2+} + O_2 + 4H^{+} \rightarrow 4Fe^{3+} + 2H_2O.$$

In the presence of dissolved oxygen, Fe^{2+} is readily oxidized to Fe^{3+}, so that the concentration of Fe^{3+} in solution rises significantly [8].

The study of this process is of considerable practical importance for raising the efficiency of leaching, because a high concentration of Fe^{3+} increases the solubility of the uranium compounds. As a result, the higher trivalent-iron level promotes the more efficient recovery of uranium from the ore.

Growth in the Fe^{3+} content of the solution directly affects the solubility of uranium species. Trivalent iron acts as a strong oxidant and promotes more vigorous leaching of uranium from the ore.

Uranium present in various minerals occurs most often as oxides and sulfides. The leach reaction can be written as:

$$UO_2 + 2Fe^{3+} \rightarrow UO_2^{2+} + 2Fe^{2+}.$$

Trivalent iron thus serves as the oxidant and converts U(IV), present as UO_2, into U(VI), represented by the uranyl ion UO_2^{2+}. This redox transformation greatly increases the solubility of uranium in acidic media and therefore improves the efficiency of its recovery from the ore.

The kinetics of the $Fe^{2+} \rightarrow Fe^{3+}$ oxidation depends on the pH, the temperature, and the oxygen content. For modelling this process, the rate-law expression can be used:

$$\text{Reaction Rate} = k[Fe^{2+}][O_2],$$

where k is the rate constant; $[Fe^{2+}]$ and $[O_2]$ are the concentrations of the reactants.

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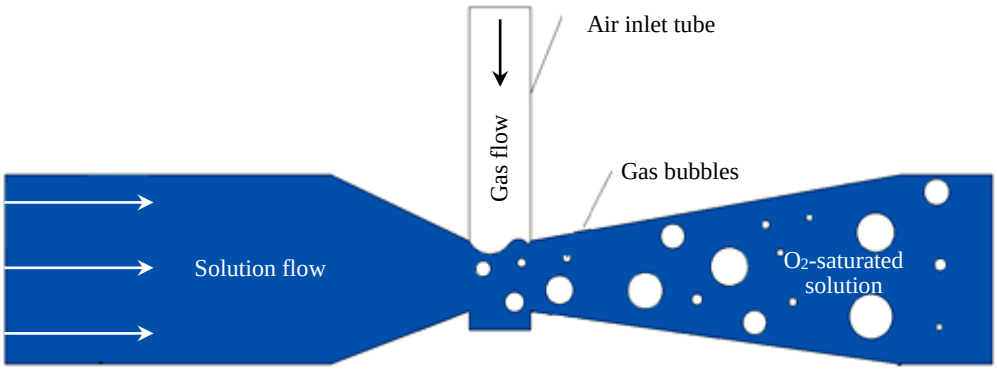


Fig.2. Air-entrainment scheme of the lixiviant using a Venturi nozzle

The pH of the medium likewise plays a decisive role. Optimal oxidation conditions lie in the range pH 2-4, which corresponds to a stable acidic environment in which the reaction proceeds most efficiently.

Efficient oxidation of divalent iron to the trivalent form in a sulfuric-acid solution requires an adequate dissolved-oxygen content, and one of the ways to achieve this is to oxygenate the solution by means of a Venturi nozzle.

The Venturi nozzle is a device that exploits hydrodynamics to manage the flow of a liquid and a gas. Its operation is based on Bernoulli's law and the associated changes in the energy parameters of the flow, which provide the conditions for air entrainment and gas dissolution as the liquid passes through the device (Fig.2) [24-26].

The experimental runs were carried out at ambient temperature (22-24 °C) for 9 h. The starting pregnant leach solution contained, on average, H_2SO_4 ≈ 15 g/l, Fe^{2+} ≈ 310 mg/l, U ≈ 19-20 mg/l, pH ≈ 2.0-2.2, and a total mineralization of ≈ 8-9 g/dm^3. After passage through the Venturi nozzle, the dissolved-oxygen concentration rose from 6.3 to 10.45 mg/l, Fe^{2+} fell to ≈ 260 mg/l, Fe^{3+} rose to 330-365 mg/l, the oxidation-reduction potential increased from 334-342 mV to 364-418 mV, and the uranium concentration in solution rose from 19.3 to 26.7 mg/l. The velocity at the throat of the nozzle was ~0.65 m/s, and samples were withdrawn every 60 min.

The principle on which the device operates is the pressure drop that occurs as a liquid passes through the constriction in a pipe. As the flow passes the narrow throat its velocity rises and the static pressure falls. A zone of reduced pressure is thus created, into which atmospheric air is drawn from outside. This mechanism ensures effective mixing and dissolution of oxygen in the lixiviant and thereby promotes the $Fe^{2+} \rightarrow Fe^{3+}$ conversion that is critical for raising uranium-leaching efficiency [27-29].

Under industrial conditions, depending on the parameters and capacity of the surface installation, different volumes of leaching agent are injected into the production wells. To establish how the flow rate and the volume of injected liquid affect the degree of oxygenation, dedicated laboratory tests were carried out. The results, which summarize the influence of flow velocity and air flow on dissolved oxygen, are given in Table.

Effect of flow velocity and LA volume on the degree of oxygenation

Flow velocity, m/s	Air flow, l/h	Oxygen concentration, mg/l
0	–	4.03
0.3	60.0	6.3
0.65	110.0	7.8
1.0	97.0	8.2
1.8	97.0	8.5

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As Table shows, the initial oxygen concentration in the lixiviant is 4.03 mg/l, whereas at a flow velocity of 0.3 m/s it reaches 6.3 mg/l. A further rise in the flow velocity from 0.3 to 1.8 m/s increases the oxygen concentration from 6.3 to 8.5 mg/l, more than double the value of the initial solution. A sharp jump in oxygen content is observed as the velocity reaches 0.65 m/s, whereas further increase up to 1.8 m/s adds only a marginal amount. The optimal flow velocity in the nozzle is therefore 0.65 m/s, since beyond this value further acceleration of the flow produces no meaningful increase in oxygen dissolution. The tabulated data were obtained during the earlier verification of the procedure [30] and are used here to justify the choice of 0.65 m/s as the operating velocity in the present experiment [31-33].

The experimental work confirmed that aeration by means of a Venturi nozzle is a highly effective and comparatively low-labour route for oxygenating the lixiviant. Traditional aeration uses costly equipment – compressors and oxygenators – together with pure oxygen, hydrogen peroxide, or other reagents. A compressor forces air into the liquid through submerged pipes; the maximum oxygenation attainable with air, however, is comparable to that achieved by the proposed scheme, which is much easier to deploy at sites where large volumes of liquid must be oxygenated [34-36].

Processing of the laboratory measurements made it possible to establish the dependence of the dissolved-oxygen concentration on flow velocity and air flow rate (Fig.3).

For the calculation of the hydrodynamics inside the Venturi nozzle, a flow velocity of 0.65 m/s was selected as the optimum. The solution density was taken as 1000 kg/m^3; the internal diameter of the wide section of the nozzle was 15 mm (0.015 m) and that of the throat was 3 mm (0.003 m).

The operation of the Venturi nozzle follows Bernoulli's principle, which is written as:

$$P_1 + \frac{1}{2}\rho v_1^2 + \rho g h_1 = P_2 + \frac{1}{2}\rho v_2^2 + \rho g h_2,$$

where P is the pressure, Pa; ρ is the fluid density, kg/m^3; v is the flow velocity, m/s; g is the gravitational acceleration, m/s^2; h is the elevation, m.

Because the process under consideration takes place in a horizontal plane, the gravitational term (ρgh) can be omitted from the calculation.

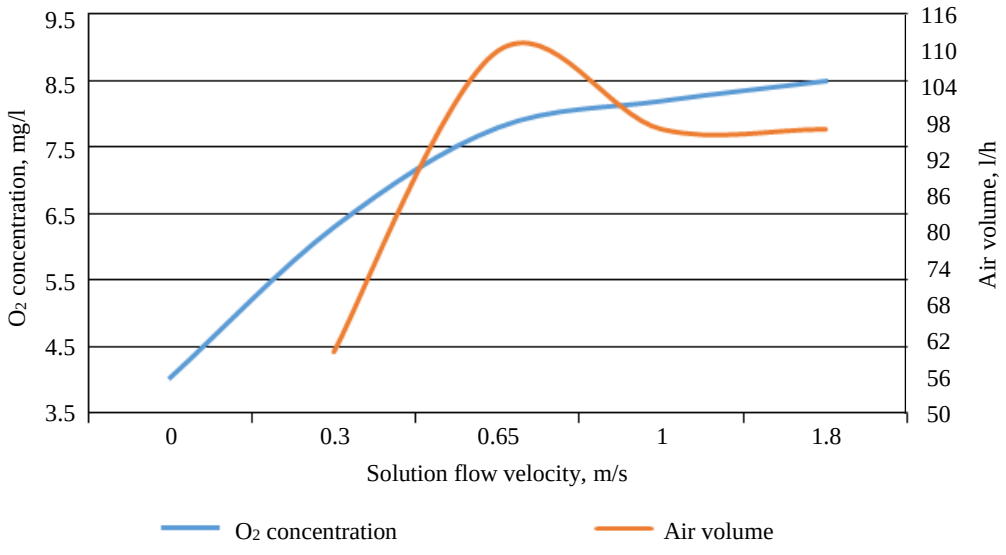


Fig.3. Dissolved oxygen concentration as a function of flow velocity and air flow rate

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Cross-sectional area of the wide section of the nozzle

$$A_1 = \frac{\pi D_1^2}{4} = \frac{\pi\left(0.015\right)^2}{4} = 1.767 \cdot 10^{-4}\ \text{m}^2.$$

Cross-sectional area of the throat of the nozzle

$$A_2 = \frac{\pi D_2^2}{4} = \frac{\pi\left(0.003\right)^2}{4} = 7.069 \cdot 10^{-6}\ \text{m}^2.$$

Let us calculate the flow velocity in the throat. By the continuity equation, the throat velocity v_2 is given by:

$$A_1 v_1 = A_2 v_2 \Rightarrow v_2 = \frac{A_1}{A_2} v_1,$$

let us insert the values:

$$v_2 = \frac{1.767 \cdot 10^{-4}}{7.069 \cdot 10^{-6}} \cdot 0.7 = 16.25\ \text{m/s}.$$

Pressure change from Bernoulli's law

$$P_1 + \frac{1}{2}\rho v_1^2 = P_2 + \frac{1}{2}\rho v_2^2.$$

The expression is rearranged to give the pressure difference between the wide section and the throat

$$\Delta P = P_1 - P_2 = \frac{1}{2}\rho\left(v_2^2 - v_1^2\right),$$

let us insert the values:

$$\Delta P = \frac{1}{2} \cdot 1000\left(16.25^2 - 0.65^2\right) = 131{,}820\ \text{Pa}.$$

The suction force is the product of the pressure difference and the cross-sectional area of the throat:

$$F = \Delta P A_2,$$

let us insert the values:

$$F = 131{,}820 \cdot 7.069 \cdot 10^{-6} = 0.932\ \text{N}.$$

The suction force of 0.932 N indicates that the Venturi nozzle is able to generate a marked rarefaction. During the oxygenation of acidic solutions this force is decisive: it ensures the effective introduction of gas into the liquid phase, improves dissolution, and produces a more uniform distribution of oxygen throughout the bulk of the solution.

The energy required for the oxidation reaction can be obtained from the standard Gibbs free-energy change ΔG. The ΔG values for the iron-oxidation reactions are tabulated in standard thermodynamic handbooks.

The oxidation of divalent iron in an acidic medium is written as $Fe^{2+} \rightarrow Fe^{3+}$. For this reaction $\Delta G \approx -74$ kJ/mol.

Taking the standard Gibbs free-energy change as –74 kJ/mol and a dissolved-oxygen mass concentration of 7.3 mg/l (i.e. $7.3 \cdot 10^{-3}$ g/l), and the molar mass of O_2 is 32 g/mol, the number of moles of oxygen per litre of solution is

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$$n = \frac{7.3 \cdot 10^{-3}}{32} = 2.28 \cdot 10^{-4} \text{mol}.$$

The energy released by the oxidation reaction is then evaluated from the molar amount:

$$\Delta G_{\text{reactions}} = n\Delta G = 2.28 \cdot 10^{-4} \cdot (-74) \approx -0.0169 \text{ kJ} = -16.9 \text{ J}.$$

Thermodynamically, $\Delta G < 0$ means that the reaction releases energy, whereas $\Delta G > 0$ means that the reaction requires energy input.

The fact that energy is released during the conversion of divalent to trivalent iron therefore indicates a spontaneous and thermodynamically favourable process. When oxygen dissolves in sulfuric acid and then oxidizes Fe^{2+} to Fe^{3+}, about 16.9 J of energy is released. This energy can promote uranium solubilization. The release of 16.9 J shows that the $Fe^{2+} \rightarrow Fe^{3+}$ oxidation is exothermic, that is, accompanied by heat evolution. This heat can raise the local temperature of the solution and activate the molecules participating in the reaction, which further accelerates the uranium leach.

The operation of the Venturi nozzle, governed by Bernoulli's principle, demonstrates the complex energy transformations that occur as a fluid moves. Variations in kinetic and static energy across the different sections of the nozzle produce air entrainment and gas dissolution. This phenomenon is of considerable practical relevance to the chemical and food industries, where control of gas solubility is critical for raising the productivity of process operations [30, 37].

The expected economic effect E (USD/year) was estimated taking into account the reduced consumption of sulfuric acid and electric power that result from a shorter time to mine out a production block when the proposed oxygenation technology is applied. The calculation was carried out for a single block with reserves of 300 t of uranium:

$$E = E_{ac} + E_{el} - Z_{pt},$$

where E_{ac} is the saving on sulfuric-acid consumption, USD; E_{el} is the saving on electric-power consumption, USD; Z_{pt} is the additional expenditure associated with the proposed technology, USD.

Saving on sulfuric-acid consumption:

$$E_{ac} = (T_b - T_p) Q_{LA} q C_{ac} / 1{,}000{,}000 =$$
$$= (19{,}187 - 17{,}268)\ 120{,}000 \cdot 6 \cdot 50 / 1{,}000{,}000 = 69{,}000 \text{ USD},$$

where T_b and T_p are the times to mine out the block under the conventional and the proposed technologies, h; Q_{LA} is the lixiviant flow rate per block, l/h; q is the consumption of sulfuric acid for fortification of 1 l of solution, g/l; and C_{ac} is the price of 1 t of sulfuric acid, USD/t.

Saving on electric-power consumption:

$$E_{el} = (T_b - T_p) q_e C_{el} N_p =$$
$$= (19{,}187 - 17{,}268) \cdot 7 \cdot 0.1 \cdot 15 = 20{,}000 \text{ USD},$$

where q_e is the hourly electric-power consumption, kW·h; C_{el} is the price of electricity, USD/kW; and N_p is the number of pumps.

Additional expenditure with the proposed technology:

$$Z_{pt} = C_V + 2C_{kr} + C_m + C_r =$$
$$= 200 + 2 \cdot 500 + 30 + 30 = 1260 \text{ USD},$$

where C_V is the price of the Venturi nozzle, USD; C_{kr} is the price of a shut-off valve, USD; C_m is the price of a pressure gauge for the parallel line, USD; C_r is the price of a flowmeter, USD;

$$E = 69{,}000 + 20{,}000 - 1260 = 87{,}740 \text{ USD}.$$

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Discussion of results

In the first stage of the work, runs with the conventional configuration of the rig were carried out. A series of experiments was then performed in which the lixiviant was oxygenated, in order to assess the influence of this factor on the concentrations of divalent and trivalent iron, on the ORP, and on the uranium content of the pregnant leach solution.

In the starting lixiviant the dissolved-oxygen concentration was 6.3 mg/l. At the bench scale, the flow velocity was set to 0.65 m/s, matching the field conditions. Once the Venturi nozzle had been used to entrain air, the oxygen concentration in solution rose to 10.45 mg/l – an increase of 66 %.

The oxidation of divalent iron in the lixiviant is a key process from both the chemical and the environmental standpoints. Introducing oxygen into the solution can radically alter the speciation of iron through its transition between the two valence states (Fig.4).

Analysis of the Fe^{2+} content over the 9 h that followed the oxygenation of the lixiviant showed a marked drop in the divalent-iron level. Before oxygenation the Fe^{2+} concentration was in the range 288-294 mg/l. After oxygenation, the level fell from a starting value of 310 mg/l to 260 mg/l by the end of the 9-hour period. This dynamic clearly indicates that the Fe^{2+} concentration decreased as a consequence of oxygenation.

A rise in the dissolved-oxygen content accelerates the Fe^{2+} oxidation and lowers its concentration in solution.

To quantify the rate of Fe^{2+} oxidation in the acidic medium, a pseudo-first-order kinetic treatment was applied:

$$\ln\left(\frac{\left[Fe^{2+}\right]_0}{\left[Fe^{2+}\right]_t}\right) = k_{obs}t,$$

where $[Fe^{2+}]_0$ is the initial Fe^{2+} concentration (310 mg/l); $[Fe^{2+}]_t$ is its concentration at time t (260 mg/l at 9 h); k_{obs} is the observed rate constant.

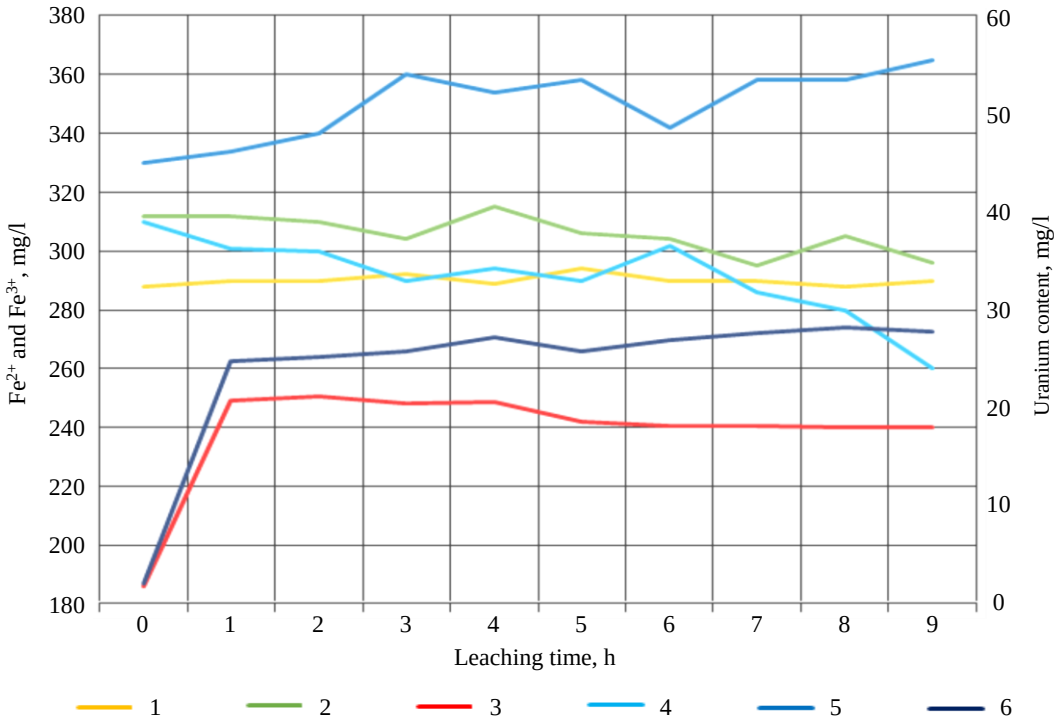


Fig.4. Uranium concentration as a function of the change in Fe^{2+} and Fe^{3+} concentrations before and after lixiviant oxygenation

1 – Fe^{2+} before oxygen saturation, mg/l; 2 – Fe^{3+} before oxygen saturation, mg/l; 3 – uranium content before oxygen saturation, mg/l; 4 – Fe^{2+} after oxygen saturation, mg/l; 5 – Fe^{3+} after oxygen saturation, mg/l; 6 – uranium content after oxygen saturation, mg/l

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Substitution of the experimental values Fe^{2+} gives $k_{obs} = (0.0195 \pm 0.003)\ h^{-1}$. This figure confirms that the Venturi nozzle accelerates the $Fe^{2+} \rightarrow Fe^{3+}$ conversion.

Before oxygenation, the Fe^{3+} concentration ranged from 296 to 315 mg/l. After oxygenation, it rose to 330-365 mg/l. This change reflects a substantial increase in trivalent-iron content driven by the higher dissolved-oxygen level.

Before oxygenation, most of the iron was present as Fe^{2+}. Only once the dissolved-oxygen content had increased did vigorous oxidation begin, leading to a rise in Fe^{3+} and a drop in Fe^{2+}.

In solutions containing Fe^{3+}, oxidative reactions affecting the valence of uranium can proceed. In particular, Fe^{3+} is able to oxidize uranium to more soluble forms, which facilitates leaching and can enhance the efficiency of uranium extraction.

The oxidation-reduction potential reflects the capacity of a solution to participate in redox reactions. It is widely used to characterize the chemical state of solutions, especially in the context of uranium leaching. To track the ORP dynamics, 10 samples were taken, with measurements performed before and after lixiviant oxygenation (Fig.5).

Before oxygenation, the ORP fluctuated only slightly, staying in the range 334-342 mV. After oxygenation, it rose substantially, reaching values between 364.4 and 418 mV.

The ORP level is therefore directly related to the dissolved-oxygen concentration. Before oxygenation, the O_2 content was low, which kept the ORP correspondingly low. Once the lixiviant had been oxygenated, the O_2 level increased, activating the oxidative reactions and raising the ORP.

Oxygen is a strong oxidant. As its concentration in the solution rises, the intensity of the oxidative reactions grows, and the ORP increases. These reactions include the oxidation of ions and molecules in solution and generate higher redox-potential values.

Processing of the experimental data made it possible to construct curves showing the uranium content of the pregnant leach solution as a function of leaching time, for both the conventional and the oxygenation-based configurations.

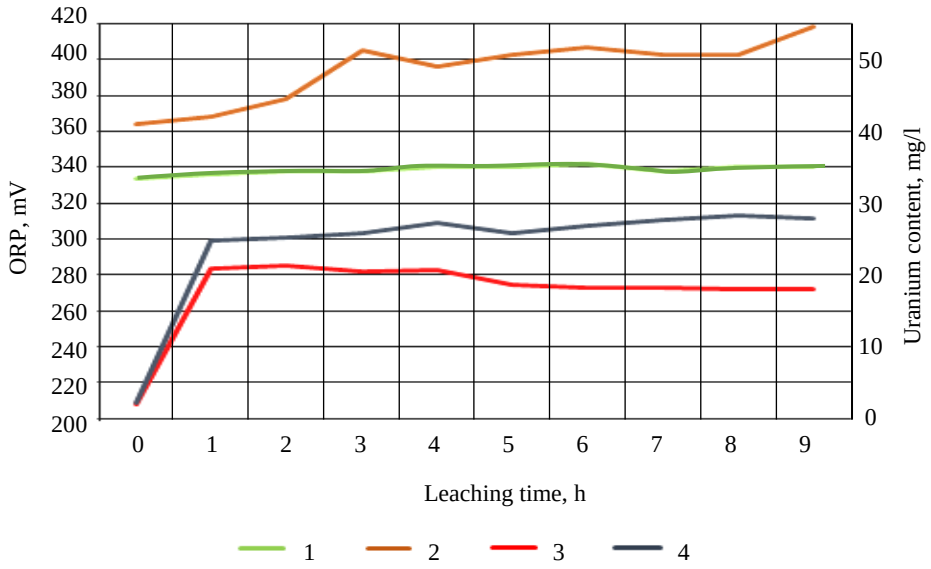


Fig.5. Uranium content as a function of the change in ORP
1 – ORP before oxygen saturation, mV; 2 – uranium content before-oxygen saturation, mg/l;
3 – ORP after oxygen saturation, mV; 4 – uranium content after oxygen saturation, mg/l

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A comparative analysis of the laboratory results shows that with the conventional procedure the mean uranium content of the pregnant leach solution is 19.3 mg/l. When the oxygenation scheme is used, this value rises to 26.66 mg/l. The proposed technology therefore raises the uranium content of the pregnant leach solution by 38.13 % relative to the conventional procedure.

The Venturi nozzle intensifies the redox processes through a combination of hydrodynamic and chemical factors: higher mass-transfer coefficient during oxygen dissolution; accelerated oxidation of Fe^{2+} to Fe^{3+}; and regeneration of Fe^{3+} within the Fe^{2+}/Fe^{3+} cycle, which sustains the efficient oxidation of U(IV) to U(VI). Turbulence and microcavitation further accelerate the reaction kinetics and promote the Eh rise documented by the experimental data.

It should be noted that the U(IV) → U(VI) transition can proceed via an intermediate $U(V) - UO_2^+$ stage. However, under the conditions of the present experiment (pH 2-4, O_2 saturation, presence of Fe^{3+}) this intermediate ion is thermodynamically and kinetically unstable and rapidly disproportionates to U(IV) and U(VI). The analytical methods used in the work (titrimetric procedure / ICP-MS) determine the total U(VI) content, so U(V) was not detected as such. The macroscopic description of the process can therefore be treated as U(IV) → U(VI).

Conclusion

The present work allowed a detailed analysis of the physico-chemical mechanisms that govern the efficiency of sulfuric-acid uranium leaching and showed that the intensification of this process is dictated not only by technological measures but, above all, by the energetics and the kinetics of the redox reactions in solution. For the first time, the $Fe^{2+} \rightarrow Fe^{3+}$ oxidation has been examined from the standpoint of thermodynamics and the system's energy balance. This reaction was shown to be thermodynamically favourable ($\Delta G < 0$) and accompanied by the release of energy that raises the local temperature of the solution and, in turn, accelerates the kinetics of subsequent redox transformations.

Dissolved oxygen was found to play the key role in shaping the redox potential of the system and in setting both the direction and the rate of the Fe^{2+}/Fe^{3+} and U(IV)/U(VI) transitions. In this work, trivalent iron is treated not simply as an oxidant but as an active redox mediator that participates in the cyclic transfer of electrons and sustains stable oxidative conditions in solution. This view makes it possible to regard the Fe^{2+}–Fe^{3+}–O_2 system as a dynamically self-supporting redox loop rather than as a set of disconnected reactions.

For the first time, a pseudo-first-order kinetic model has been applied to the Fe^{2+} oxidation in a sulfuric-acid medium. This made it possible to quantify the influence of dissolved oxygen on the reaction rate and to confirm the acceleration of the oxidative processes during aeration. The efficiency of uranium oxidation was shown to be determined by the coordinated action of energetic, kinetic, and electrochemical factors, including the O_2 concentration, the Fe^{2+}/Fe^{3+} ratio, and the value of Eh.

It has also been substantiated that the transition of uranium from the tetravalent to the hexavalent state can proceed via a short-lived intermediate U(V), which under acidic conditions, elevated Eh, and in the presence of Fe^{3+} is both thermodynamically and kinetically unstable. This justifies a correct macroscopic description of the process as a direct U(IV) → U(VI) transition without distorting the physico-chemical essence of the phenomenon.

The article develops the theoretical understanding of sulfuric-acid uranium leaching as a managed redox system with a definite energy and kinetic regime, and complements the previously obtained technological results with a fundamental justification of the underlying mechanisms.

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