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«Жанармай, катализ және электрохимия институты» АҚ

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ИЗВЕСТИЯ

НАЦИОНАЛЬНОЙ АКАДЕМИИ НАУК
РЕСПУБЛИКИ КАЗАХСТАН
АО «Институт топлива, катализа и
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NAS RK is pleased to announce that News of NAS RK. Series of chemistry and technologies scientific journal has been accepted for indexing in the Emerging Sources Citation Index, a new edition of Web of Science. Content in this index is under consideration by Clarivate Analytics to be accepted in the Science Citation Index Expanded, the Social Sciences Citation Index, and the Arts & Humanities Citation Index. The quality and depth of content Web of Science offers to researchers, authors, publishers, and institutions sets it apart from other research databases. The inclusion of News of NAS RK. Series of chemistry and technologies in the Emerging Sources Citation Index demonstrates our dedication to providing the most relevant and influential content of chemical sciences to our community.

Қазақстан Республикасы Ұлттық ғылым академиясы «ҚР ҰҒА Хабарлары. Химия және технология сериясы» ғылыми журналының Web of Science-тің жаңаланған нұсқасы Emerging Sources Citation Index-те индекстелуге қабылданғанын хабарлайды. Бұл индекстелу барысында Clarivate Analytics компаниясы журналды одан әрі the Science Citation Index Expanded, the Social Sciences Citation Index және the Arts & Humanities Citation Index-ке қабылдау мәселесін қарастыруда. Web of Science зерттеушілер, авторлар, баспашылар мен мекемелерге контент тереңдігі мен сапасын ұсынады. ҚР ҰҒА Хабарлары. Химия және технология сериясы Emerging Sources Citation Index-ке енуі біздің қоғамдастық үшін ең өзекті және беделді химиялық ғылымдар бойынша контентке адалдығымызды білдіреді.

НАН РК сообщает, что научный журнал «Известия НАН РК. Серия химии и технологий» был принят для индексирования в Emerging Sources Citation Index, обновленной версии Web of Science. Содержание в этом индексировании находится в стадии рассмотрения компанией Clarivate Analytics для дальнейшего принятия журнала в the Science Citation Index Expanded, the Social Sciences Citation Index и the Arts & Humanities Citation Index. Web of Science предлагает качество в глубину контента для исследователей, авторов, издателей и учреждений. Включение Известия НАН РК в Emerging Sources Citation Index демонстрирует нашу приверженность к наиболее актуальному и влиятельному контенту по химическим наукам для нашего сообщества.

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SORPTION OF MANGANESE (II) AND VANADIUM (IV) CATIONS BY TAGAN BENTONITE IN AN AQUEOUS MEDIUM

Abstract: the process of sorption of Mn^{2+} and V^{4+} cations by bentonite of the Tagan deposit studied depending on various factors (T, °C; τ , min; S: L ratio; C_{Mn} and C_V , mg/l). It was found that bentonite exhibits sorption properties with respect to the studied cations in a wide range of pH values (from 1 to 10). The highest degree of their sorption is achieved at the consumption of bentonite 1.5 g per 100 mass. p.of metal-containing solution. The influence of the nature of the cation on the sorption capacity of Tagan bentonite was established. The most intense increase in the degree of sorption of manganese (II) occurs in 15 min with an increase in temperature to 35°C. The highest degree of sorption of V^{4+} cations is achieved in 60 min at 25°C in weakly concentrated vanadium-containing solutions (50 mg/l V^{4+}). Bentonite from the Tagandeposit exhibits a lower sorption capacity with respect to V^{4+} cations as compared to Mn^{2+} cations, which is due to the nature of the cation.

Key words: bentonite, montmorillonite, sorption, desorption, degree of sorption, Mn^{2+} and V^{4+} cations.

Introduction. In recent years, heavy metals have been entering the biosphere intensively from the wastewater of the light, heavy, oil-producing and oil-refining industries. Polluted industrial effluents contain Co, Mn, Ni, V, Cr, Pb, etc. metals that are able to concentrate in organisms and have a negative effect on human health. Heavy metals have a pronounced summation effect, so the combined presence of several elements increases their toxic effect several times [1]. The problem of purification of various media from heavy metals is solved by the sorption method using natural sorbents that have ion exchange and sorption properties, good physical and chemical characteristics, and low cost [2]. Such requirements are met by clays with a layered structure (bentonites) with the main rock-forming mineral montmorillonite, a highly developed internal surface, the ability to swell and chemical resistance [3, 4]. Bentonites exhibit sorption properties, first, due to a highly branched surface on which active adsorption centers are located [4-7, 8]. Secondly, due to the formation of intra-atmospheric complexes through the Si/O - and Al/O-groups at the edges of clay particles [9].

Manganese (II) and vanadium (IV) are toxic heavy metals. High concentrations of these metals have a toxic effect on living organisms, leading to irreversible violations of biochemical processes.

Currently, to remove manganese and vanadium cations from water, it is necessary to maintain the pH to a certain level in order to oxidize them, followed by precipitation of hydroxides of these metals and filtration of the resulting compounds.

The problem of wastewater treatment and water treatment largely rests on the lack of cheap multifunctional sorbents that are stable during operation. The search for sorbents with high sorption capacity, selectivity, and low cost is of great importance, since ion exchange has taken an important place among the main methods of water purification, which are diverse in origin.

Natural and synthetic materials – sand, wood, activated carbon, silica gels, ion-exchange resins, etc. - are used as sorbents for cleaning various media. However, these sorbents have a number of disadvantages, such as fast clogging, they have insufficient high adsorption properties, since their pore sizes are small and the internal surface area is small. Synthetic sorbents have a good absorption capacity, but are characterized by higher cost, complexity of production and the use of high temperatures, pressures and catalysts.

In recent years, natural layered aluminosilicates, such as zeolites, glauconites, bentonites, montmorillonites, kaolinites, illites, chlorites, etc., have been used in the practice of

water treatment. Special attention should be paid to clay minerals, which have a high specific surface area, chemical and mechanical stability, the ability to vary surface and structural characteristics, and low cost [10].

To remove toxic heavy metals, it is advisable to use a widespread, inexpensive clay mineral as an adsorbent - bentonite clays. The Republic of Kazakhstan has significant natural reserves of bentonite, the main mineral of which is montmorillonite, which determines their sorption properties [12, 13]. Bentonite clays are natural mineral sorbents, and rarely occur in the layers in their pure form, because they contain accompanying minerals (beidellite, hydrosludes, chlorites, kaolite, etc.) [11].

The aluminosilicate framework of clays mainly consists of alternating parallel two-dimensional layers formed by silicon-oxygen tetrahedra and aluminum-oxygen hydroxyl octahedra. In montmorillonite (MM), the main component of clays, the external and internal surfaces of the basal faces are active. In addition, its edge areas are active, creating a large active surface that reaches 700-800 m² / g [14].

Montmorillonite clays are characterized by a high cation exchange capacity of 60 to 150 meq/100g. The exchange cations of montmorillonite are located in the inter-pack space. The exchangeable cations of montmorillonite are more often represented by sodium, calcium, and magnesium ions, which are easily exchanged for other cations. This is due to the fact that the charge of the montmorillonite lattice is mainly due to the charge of the octahedral layer. The charge of the octahedral layer acts on the exchange cations at a greater distance than the charge of the tetrahedral layer. Consequently, the binding strength of cations to the octahedral layer is less strong than that of the tetrahedral layer [15].

The MM crystal lattice is electrically unbalanced (an excess of negative charges is created). The lack of positive charges is covered by exchange cations that are adsorbed on the outer and inner surfaces of the package and on its edge parts (in areas of chips and violations).

The authors of the article [16] determined the material composition and sorption properties of clays in relation to heavy metal ions: Cu²⁺, Pb²⁺, Fe³⁺. With the help of the studied sorbents, it was possible to reduce the concentration of Cu (II) ions by (83.5-93)%, Pb (II) ions by (94-97)%, and Fe(III) ions by (97-99)%, with an initial concentration of ions of each metal of 10 mg/l.

Methodology of the study. In this work, montmorillonite-containing bentonite from the Tagan deposit (East Kazakhstan) was used, in which

the content of montmorillonite reaches (90-92)%. Natural bentonite has the following chemical composition, wt.%: 69, 2SiO₂; 23.40 Al₂O₃; 3.05 Fe₂O₃; 1.78 Cat; 1.55 MgO; 0.47 Mn; 0.08 2; 0, 04Na₂O; 0.37 TiO₂. Judging by the chemical composition, it belongs to calcium bentonite.

To study the sorption properties of bentonite from the Tagan deposit, cations of heavy metals – manganese (II) and vanadium (IV), which are present in wastewater and natural waters, were selected. In order to exclude the influence of impurities, the sorption capacity of natural bentonite with respect to Mn²⁺ and V⁴⁺ cations was studied on the model systems "bentonite-Mn²⁺(V⁴⁺) - H₂O" under mixing conditions.

The set temperature was maintained in a thermostatically controlled reactor. A certain concentration of Mn²⁺ and V⁴⁺ cations was created by introducing an estimated amount of MnSO₄·5H₂O and VOSO₄·3H₂O sulfates into an aqueous solution.

The influence of time, temperature, norm of bentonite (S:L) and pH of the medium on its sorption capacity was studied at a constant C_{Mn} and C_V equal to 200 mg/l.

The effect of the concentration of V⁴⁺ and V²⁺ cations on their sorption by natural bentonite was studied at experimentally found optimal values of temperature, time, and sorbent consumption.

The degree of sorption (R, %) was found by the difference between the initial and equilibrium concentrations of metal solutions and calculated by the formula:

$$R = \frac{C_o - C_p}{C_o} \cdot 100\% ,$$

where C_o is the initial concentration of metal ions in solution, mg / l; C_p - concentration of metal ions in the solution after its purification, mg / l.

The initial and final concentrations of manganese and vanadium ions in the solutions were determined using an atomic absorption spectrophotometer of the AA-7000 type, manufactured by Shimadzu Corporation, Japan, no. A30664901456.

Results and discussion. The sorption capacity of natural bentonite with respect to V⁴⁺ and Mn²⁺ cations was studied on the model systems "bentonite-V⁴⁺ (Mn²⁺) - H₂O" under mixing conditions.

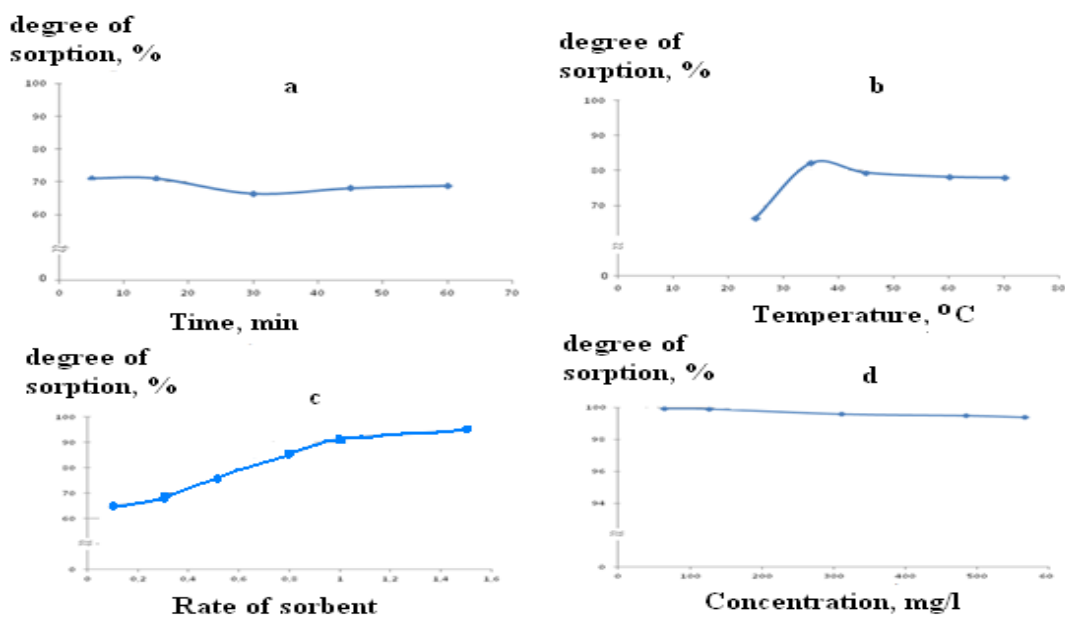
The study of the effect of time in the range of 5-60 min on the sorption capacity of bentonite with respect to Mn²⁺ and V⁴⁺ cations was carried out at 25°C and the norm of the sorbent (S: L) equal to 0.5: 100 at a constant C_{Mn} and C_V equal to 200 mg/l (Figure 1,2).

On the sorption curves of both cations, there is a minimum, which is due to the process of their desorption from the sorbent into the solution. A weakly expressed min on the sorption curve of manganese (II) cations is prescribed at 30 min (Figure 1 a), and a clearly expressed min on the sorption curve of V^{4+} is shifted to 45 min (Figure 2 a). The highest degree of solution purification from Mn^{2+} (88.9%) is achieved in the first 5-10 minutes of the process, and the highest degree of solution purification from V^{4+} (65-66%) is achieved in 60 minutes (Figure a).

The nature of the sorption curves of vanadium (IV) differs markedly from the sorption curves of divalent manganese (Figure 1 a, b, d). The exception is the dependence of the sorption of V^{4+} and Mn^{2+} cations on the sorbent rate (Figure 1, 2 c). The degree of purification of the solution from vanadium (IV), as well as in the case of Mn^{2+} cations (Figure 1c), increases with an increase in the load of the sorbent. It was found that the sorption of V^{4+} cations, as well as Mn^{2+} ions has lower limits from 45.8 to 46.5 %.

The sorption of Mn^{2+} cations is significantly affected by the process temperature (Figure 1b). The effect of temperature on the sorption of manganese (II) was studied in the range from 25 to 80°C at S:L equal to 1:0.5 for 15 minutes. With an increase in temperature to 35 °C, the degree of purification of the solution from Mn^{2+} ions increases sharply from 62 to 82 %, and above this temperature it decreases to 77 %, remaining almost unchanged in the range (45-70)°C. An increase in temperature above 35 °C initiates the desorption of manganese (II) from bentonite into the solution.

An increase in the process temperature contributes to a decrease in the degree of purification of the solution from V^{4+} cations (Figure 2b) in the entire temperature range under study, in contrast to the sorption curve of Mn^{2+} (Figure 1b). With an increase in temperature from 25 to 70°C, the degree of sorption of V^{4+} ions decreases by 21.1% due to their desorption from the sorbent into the solution. To obtain a high degree of purification of the solution from V^{4+} ions (52.2%), a temperature of no more than 25°C is required.



a - time; b-temperature; c-bentonite rate; d-concentration C_{Mn}

Figure 1 - Influence of the studied factors on the sorption of Mn (II) cations

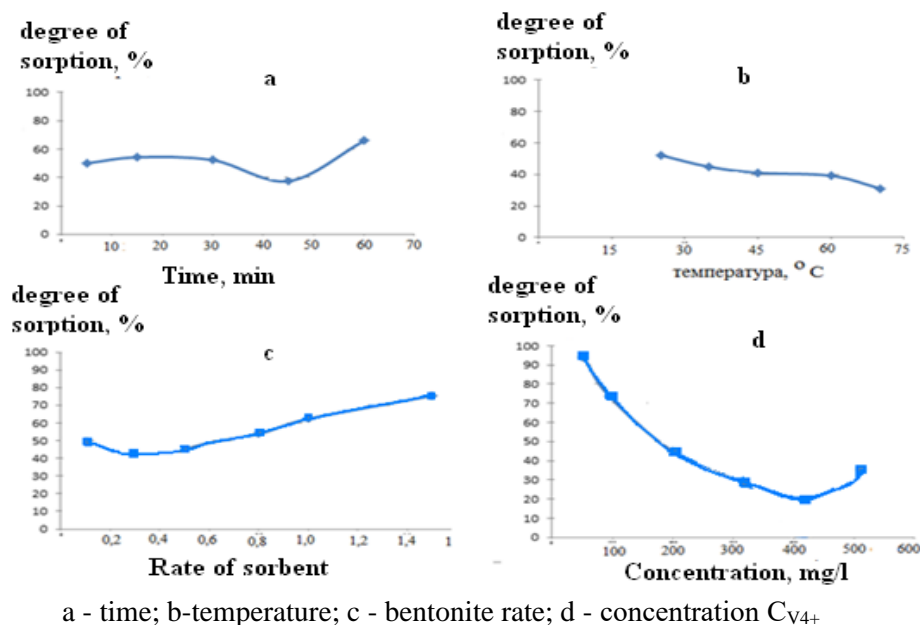


Figure 2 - Influence of the studied factors on the sorption of V (IV) cations

The extreme dependence of the sorption curves on time and temperature, that is, the appearance of min and max on them, is probably caused by the processes leading to the creation of an equilibrium concentration of manganese cations in the manganese-containing solution-sorbent system, since after a certain time of temperature, as a result of saturation of bentonite with these cations, the reverse process of transition of manganese to solution is carried out.

The effect of the sorbent consumption on the degree of purification of the solution from Mn^{2+} cations was studied at 15 min and 35°C (Figure 1c). The ratio of bentonite to the purified solution was changed in the range from 0.1: 100 to 1.5: 100. It is established that with an increase in the load of the sorbent, the residual content of Mn^{2+} cations in the liquid phase of the system decreases, respectively, the degree of their sorption increases. A high degree of sorption of Mn (II) ions is achieved at a S: L ratio of 1:1.5 (94.9 %), which is due to the large mass of the sorbent.

The curve of the sorption of Mn (II) ions from their concentration, obtained at the optimal temperature of 35 °C, the norm of bentonite of 1.5 g per 100 mass.p. of a manganese-containing solution for 15 minutes of the process, is rectilinear (Figure 1d). In the range of concentrations from 50 to 100 mg/l of Mn^{2+} , the total absorption of sorbed cations by bentonite occurs. Over 100 mg/l of Mn^{2+} in solution, there is a slight tendency to decrease the degree of manganese sorption. However, up to the content of Mn (II) cations of 600 mg/l, a high degree of purification of the solution remains (99.4 %).

With an increase in the concentration of V^{4+} cations in the solution, the degree of their sorption decreases, reaching a minimum (<1 %) in a vanadium-containing solution with a CV equal to 400 mg/l (Figure 2 d). At a concentration of vanadium (IV) 500 mg/l, the degree of purification of the solution increases to 47.2 %. But the solution that is weakly concentrated on vanadium (50 mg/l) is most deeply purified at 93.2%.

The extreme nature of the curves of the dependence of the sorption of V^{4+} cations on time and concentration is due to the same processes as in the case of the sorption of Mn^{2+} cations.

Conclusion. It follows from the conducted studies that bentonite of the Tagan deposit under optimal conditions (τ – 15 min; T – 35 °C, S: L =1:1.5) for all the studied concentrations of manganese (II) cations (50-600 mg/l) exhibits high sorption properties in acidic, alkaline and neutral solutions and can be used to purify various media from Mn^{2+} ions. It was found that under optimal conditions (25°C, 60 min, S: L =1.5:100) in low-concentrated vanadium-containing solutions (50 – 100mg/l V^{4+}), the degree of sorption reaches (93.2-73.7)%, and in high-concentrated (500 mg/l V^{4+}) - 47.2%.

Studies have shown that bentonite from the Tagan deposit exhibits a noticeably lower sorption capacity with respect to V^{4+} cations as compared to Mn^{2+} cations. Comparative analysis of the data obtained showed that in the aquatic environment, natural bentonite exhibits a preferable sorption capacity in relation to Mn (II) cations, regardless of the process conditions, which is due to both the nature of bentonite and the nature of the sorbed

cation. When bentonites contain more divalent exchangeable cations Ca^{2+} , Mg^{2+} , what we have in our case, they have a greater number of active centers on the surface of particles, through which the dispersed phase is hydrogen bonded with the dispersed medium [17]. If, however, monovalent metals, mainly Na-ions, prevail in the composition of exchangeable cations, then the sorption activity of the surface manifests itself to a lesser extent. In this regard, calcium bentonites are the best sorbents in comparison with sodium. Moreover, in our case, the sorption mechanism due to the emergence of uncompensated electronic states prevails over the cation exchange [17].

The studies have shown that the bentonite of the Tagan deposit exhibits a significantly lower sorption capacity with respect to V^{4+} cations compared to Mn^{2+} cations. A comparative analysis of the obtained data showed that in an aqueous

medium, natural bentonite exhibits a preferred sorption capacity with respect to Mn (II) cations, regardless of the process conditions, which is due to both the nature of bentonite and the nature of the sorbed cation. When bentonites contain more divalent exchange cations Ca^{2+} , Mg^{2+} , what we have in our case, they have a greater number of active centers on the surface of the particles, through which the hydrogen bond of the dispersed phase with the dispersed medium is carried out [17]. If the composition of the exchange cations is dominated by monovalent metals, mainly Na-ions, then they show less sorption activity of the surface. In this regard, calcium bentonites are the best sorbents in comparison with sodium. Moreover, in our case, the sorption mechanism due to the occurrence of uncompensated electronic states prevails over cation exchange [17].

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СУЛЫ ОРТАДАН ТАҒАН БЕНТОНИТІМЕН МАНГАНЕЦ (II) ЖӘНЕ ВАНАДИЙ (IV) КАТИОНДАРЫН СОРБЦИЯЛАУ

Аннотация: таган кен орнының бентонитімен Mn^{2+} және V^{4+} катиондарының сорбциялану процесі әртүрлі факторларға байланысты зерттелді ($T^{\circ}\text{C}$; τ , мин.; Қ:С қатынасы; C_{Mn} және C_{V} , мг / л). Бентонит зерттелген катиондарға қатысты рН мәндерінің (1-ден 10-ға дейін) кең диапазонында сорбциялық қасиеттер көрсететіні анықталды. Олардың сорбциялануының ең жоғары дәрежесі құрамында металл бар ерітіндіде 100 масс. бөл. бентонит шығыны 1,5 г болғанда қол жеткізіледі. Таган бентонитінің сорбциялық қабілетіне катиондар табиғатының әсері анықталды. Марганецтің (II) сорбциялану дәрежесінің ең қарқынды өсуі температураның 35°C дейін жоғарылауымен 15 минут ішінде болады. V^{4+} катиондарының сорбциялануының ең жоғары дәрежесіне әлсіз концентрленген ванадий құрамды ерітінділерде (50 мг / л V^{4+}) 25°C температурада 60 минут ішінде қол жеткізіледі. Таган кен орнындағы бентониті V^{4+} катиондарына Mn^{2+} катиондарына салыстырғанда төмен сорбциялық қабілеттілік көрсетеді.

Түйін сөздер: бентонит, монтмориллонит, сорбция, десорбция, Mn^{2+} и V^{4+} катиондарын сорбциялау дәрежесі.

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СОРБЦИЯ КАТИОНОВ МАРГАНЦА (II) И ВАНАДИЯ (IV) ТАГАНСКИМ БЕНТОНИТОМ В ВОДНОЙ СРЕДЕ

Аннотация: исследован процесс сорбции катионов Mn^{2+} и V^{4+} бентонитом Таганского месторождения в зависимости от различных факторов ($T^{\circ}\text{C}$; τ , мин.; соотношение Т:Ж; C_{Mn} и C_{V} , мг/л). Найдено, что бентонит проявляет сорбционные свойства по отношению к исследуемым катионам в широком диапазоне значений рН (от 1 до 10). Наибольшая степень их сорбции достигается при расходе бентонита 1,5 г на 100 масс.ч. металл содержащего раствора. Установлено влияние природы катиона на сорбционную способность Таганским бентонитом. Наиболее интенсивно увеличение степени сорбции марганца (II) происходит за 15 мин с повышением температуры до 35°C . Наибольшая степень

сорбции катионов V^{4+} достигается за 60 мин при $25^{\circ}C$ в слабоконцентрированных ваннах содержащих растворах ($50 \text{ мг/л } V^{4+}$). Бентонит Таганского месторождения проявляет меньшую сорбционную способность по отношению к катионам V^{4+} по сравнению с катионами Mn^{2+} .

Ключевые слова: бентонит, монтмориллонит, сорбция, десорбция, степень сорбции, катионы Mn^{2+} и V^{4+} .

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