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# Х А Б А Р Ш Ы С Ы

# ВЕСТНИК

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РЕСПУБЛИКИ КАЗАХСТАН

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**M.K. Alimardanova[1], Zh.B. Khamzina[1], D.T. Balpanova[2]**

[1]Almaty Technological University;
[2]Kazakh National Medical University named after S.D. Asfendiyarov, Almaty, Kazakhstan.
E-mail: alimardan.matu4@mail.ru, zhuldyz_hamzina@mail.ru, balpanova00@mail.ru

# KAZAKHSTAN ZEOLITES AS A PERSPECTIVE MATERIAL IN THE WATER TREATMENT OF THE DAIRY INDUSTRY

**Abstract.** In the modern world, there are practically no sources of clean water suitable for human consumption. As a result, at any enterprise in the food industry a number of sequential operations are carried out related to the purification of water necessary for the production of products. In all processes of processing dairy raw materials, water supply to enterprises is carried out from a public water supply network and / or from an artesian well. The article discusses the feasibility of using natural zeolites to ensure water safety. Research data of Kazakhstan zeolites of various deposits and their application in practice were studied.. As shown by the results of experimental studies, zeolites of the Taizhuzgen deposit significantly improve water quality. As a result of activation, the sorption ability of zeolite rises to 85-90%. Production tests of zeolites indicate the significant potential of this mineral raw material. Natural zeolites are cheap, resistant to mechanical wear and can be reused after regeneration. Studies have shown that Taizuzgen zeolite is a good sorption material and can be recommended in further studies with the aim of developing filters for water purification used in water treatment processes in the dairy industry.

**Key words:** safety, water, zeolites, sorption ability, zeolite benefits, modification.

**Introduction.** In the modern world, there are practically no sources of clean water suitable for human consumption. That is why at any enterprise in the food industry a number of sequential operations are carried out related to the purification of water necessary for the production of products.

For the production of dairy products of a wide range, they are increasingly resorting to the use of dry canned milk. An integral part of the production process is the restoration of these products with drinking water. There are many operations to purify water, but having received purified drinking water and directing it to restore dry canned food, we will no longer receive dairy products that are as full in their properties and benefits as they were before drying. There will be no longer that original natural structure that exists in fresh milk [1].

Today, research is being conducted on many biological objects, the study of their energy and structural properties, including water, as well as its effect on other biological fluids.

Special water treatment methods imply the purposeful imparting of certain properties to water. For example, milk powder in such water can dissolve better, acquire better organoleptic and physicochemical properties, and the system itself – a solution obtained by reconstituting dry components – can acquire a certain structure similar to that of natural dairy products [2].

The aim of the study is to ensure water quality using zeolites in dairy production technology.

**Research results and discussion.** Food industry enterprises mainly use water from the drinking water supply system in food production [3]. The water used in the production of milk and dairy products must comply with the requirements of the Technical regulation of the Custom Union's TR 033/2013 'On the Safety of Milk and Dairy Products" [4]. Water used in the process of the manufacturing of food products and in direct contact with food raw materials and packaging materials must meet the requirements for drinking water established by legislation of the Member States of the Customs Union TR

CU 021/2011 "On Food Safety" and the Technical Regulation of the Republic of Kazakhstan "Requirements for the Safety of Drinking Water for the Population" (table 1) [5].

Table 1 – Basic requirements for water quality in the production of reconstituted dairy products

| Indicators | Standards (maximum permissible concentrations), no more | Hazard indicator <1> | Hazard Class |
|---|---|---|---|
| Hydrogen indicator, pH | Within 6-9 | - | - |
| Total mineralization (dry residue), mg/l | 1000(1500)<2> | - | - |
| Total hardness, mEq/L | 7,0(10)<2> | - | - |
| Permanganate oxidation, mg/l | 5,0 | - | - |
| Petroleum products, total, mg/l | 0,1 | - | - |
| Surfactants (anionic surfactants) anionic, mg/l | 0,5 | - | - |
| Phenolic Index, mg/l | 0,25 | - | - |
| Aluminum ($Al^{3+}$), mg/l | 0,5 | S.-t. | 2 |
| Barium ($Ba^{2+}$), mg/l | 0,1 | S.-t. | 2 |
| Beryllium ($Be^{2+}$), mg/l | 0,0002 | S.-t. | 1 |
| Boron (B, total), mg/l | 0,5 | S.-t. | 2 |
| Iron (Fe, total), mg/l | 0,3 (1,0)<2> | Org. | 3 |
| Cadmium (Cd, total), mg/l | 0,001 | S.-t. | 2 |
| Manganese (Mn, total), mg/l | 0,1 (0,5)<2> | Org. | 3 |
| Copper (Cu, total), mg/l | 1,0 | Org. | 3 |
| Molybdenum (Mo, total), mg/l | 0,25 | S.-t. | 2 |
| Arsenic (As, total), mg/l | 0,05 | S.-t. | 2 |
| Nickel (Ni, total), mg/l | 0,1 | S.-t. | 3 |
| Nitrates ($NO^{3-}$), mg/l | 45 | S.-t. | 3 |
| Hydrargyrum (Hg, total), mg/l | 0,0005 | S.-t. | 1 |
| Lead (Pb, total), mg/l | 0,03 | S.-t. | 2 |
| Selenium (Se, total), mg/l | 0,01 | S.-t. | 2 |
| Sulphates ($SO_4^{2-}$), mg/l | 500 | Org. | 4 |
| I and II, mg/l | 1,5 | S.-t. | 2 |
| III, mg/l | 1,2 | | 2 |
| Chlorides ($Cl^-$), mg/l | 350 | Org. | 4 |
| Chromium ($Cr^{6+}$), mg/l | 0,05 | S.-t. | 3 |
| Cyanides, mg/l | 0,035 | S.-t. | 2 |
| Zinc ($Zn^{2+}$), mg/l | 5,0 | Org. | 3 |
| $\gamma$-HCH (lindane), mg/l | 0,002 <3> | S.-t. | 1 |
| DDT (sum of isomers), mg/l | 0,002 <3> | S.-t. | 2 |
| 2,4-D, mg/l | 0,03 <3> | S.-t. | 2 |

Notes: <1> - The limiting sign of the harmfulness of the substance for which the standard is established: S.-т. - sanitary toxicological, org. - organoleptic. <2> - The value indicated in brackets can be established by decision of the chief state sanitary doctor in the appropriate territory for a particular water supply system based on an assessment of the sanitary and epidemiological situation in the village and the water treatment technology used. <3> - Standards adopted in accordance with WHO recommendations.

Water from the drinking water supply system is characterized by a rather high content of impurities (table 2) [6]. It can be confirmed by the current provisions of the Technical Regulation of the Customs Union «On the Safety of Milk and Dairy Products», as well as the Technical Regulation of the Republic of Kazakhstan «Requirements for the Safety of Drinking Water for the Population» dated May 13, 2008, used by various enterprises producing dairy products.

Table 2 – The content of heavy metals in natural waters of Zhambyl and Almaty regions

| Indicator of quality | Normative value (maximum permissible concentration) no more | Water intake (samples) | | | |
|---|---|---|---|---|---|
| | | sample №1 (Issyk city, Almaty region) | sample №2 (Karasai district, Almaty region) | sample №3 (Auezov district, Almaty) | sample №4 (village Merke, Zhambyl region) |
| Hydrogen indicator, pH | Within 6-9 | 8,14 | 7,91 | 7,92 | 6,64 |
| Total hardness, mEq//l | 7,0(10)<2> | 2 | 2,9 | 4,4 | 3 |
| Chlorides ($Cl^-$) | 350 | 7 | 7,5 | 20,5 | 14,5 |
| Iron (Fe, total), mg/l | 0,3 (1,0)<2> | 3,1969 | 0,096 | 0,091 | 17,16 |
| Lead (Pb, total), mg/l | 0,03 | not detected | 0,04079 | not detected | not detected |
| Cadmium (Cd, total), mg/l | 0,001 | 0,00091 | 0,0039 | 0,00018 | not detected |
| Arsenic (As, total), mg/l | 0,05 | 0,00882 | 0,02716 | 0,05936 | not detected |
| Zinc ($Zn^{2+}$), mg/l | 5,0 | 0,00295 | 0,00382 | not detected | not detected |
| Copper (Cu, total), mg/l | 1.0 | not detected | 0,4892 | not detected | not detected |
| Chromium ($Cr^{6+}$), mg/l | 0,05 | not detected | not detected | not detected | not detected |
| $\gamma$-HCH (lindane), mg/l<br>- $\alpha - HCH$<br>- $\beta - HCH$<br>- $\gamma$- HCH | 0,002 <3> | <br>not detected<br>not detected<br>not detected | <br>not detected<br>not detected<br>not detected | <br>not detected<br>not detected<br>not detected | <br>not detected<br>***0,000013***<br>0,00002 |
| Heptachlor | 0,002 <3> | 0,04873 | not detected | not detected | 0,000007 |
| DDT (sum of isomers), mg/l | 0,002 <3> | not detected | not detected | not detected | not detected |

The dairy industry consumes significant amounts of water for technological needs, which must meet all the usual requirements for drinking water, and also have a higher degree of bacteriological purity, low stiffness and lack of iron, which even in minimal amounts can cause undesirable odor changes and taste of finished products [7].

Zeolite filters can become a solution to the problem of removing iron and other elements from water. Natural zeolites are widespread and cheap mineral raw materials, have a unique range of physicochemical, adsorption and ion-exchange properties, due to which they are widely used in wastewater treatment and drinking water treatment technologies [8].

The use of zeolites for processing raw materials and finished food products is interesting from the point of view of regulating the mineral composition of the product, as well as improving its hygienic, physico-chemical characteristics. This is primarily due to the unique combination of properties of natural zeolites. Due to its crystal-chemical structure, zeolites are biologically active and acid resistant. They have a prolonged adsorption, ion exchange, catalytic and detoxification ability. However, this raw material is not sufficiently used in the food industry, although it has been well studied from the biomedical, hygienic sides [9].

Currently, in the field of water treatment and water purification, natural materials (quartz sand, shungite, expanded clay, granite sand, burned rocks, zeolites, etc.) remain the main ones due to their better knowledge, accessibility, and relatively low cost [10]. Their reserves in Kazakhstan are significant: a large deposit of natural zeolites Taizhuzgen (Tarbagatai district of East Kazakhstan region, approved reserves - 7 million tons, forecasted - 215 million tons) and Shankanai (Kerbulak district, Almaty region, approved reserves – 5,5 million tons, forecasted-120 million tons), which are prepared for industrial development [11]. In Southern Kazakhstan, Altyn-Emel (41 million tons), Karzhantau and Daubabin zeolite deposits were pre-evaluated [12].

It is known that zeolites are characterized by favorable technological parameters for creating filtration materials on their basis. They do not swell in water, can be easily machined with subsequent fractionation, possess molecular sieve, absorption (ion exchange) and other useful properties, while being environmentally safe. Technical conditions have been developed and a sanitary-hygienic conclusion has been obtained on the zeolite of the Honguruu deposit as a filtering and sorbing material for the treatment of natural and waste water [13]. Countries with experience in using natural zeolites are Bulgaria, Hungary, USA, Turkey, Czech Republic, Japan, etc. [14].

The main indicators of the zeolites of Kazakhstan deposits are identical to the known deposits of Ukraine (Sokrinitskoe), Georgia (Tedzamskoe) and Russia (Kholinskoye, Kulikovskoye, Vanginskoye) [15]. Of the studied tuffs, Choline zeolite in any form is more effective in terms of the extraction of heavy metals, which, apparently, can be explained by the more rigid crystalline structure of the mineral and its chemical composition (table 3) [16].

Table 3 - Sorption of heavy metal ions from model solutions by zeolites

| Cations | The volume of filtrate before the breakthrough of the cation, in column volumes | | | | DEC, mmol/g Zeolite | | | |
|---|---|---|---|---|---|---|---|---|
| | Sakhaptin | | Choline zeolite | | Sakhaptin | | Choline zeolite | |
| | natural | $H^+$- form | natural | $H^+$- form | natural | $H^+$- form | natural | $H^+$- form |
| $Pb^{2+}$ | 1230 | 1353 | 1556 | 1690 | 0.98 | 1.10 | 1.15 | 1.32 |
| $Ni^{2+}$ | 280 | 340 | 420 | 480 | 0.81 | 0.90 | 0.94 | 1.04 |
| $Cu^{2+}$ | 215 | 310 | 380 | 420 | 0.35 | 0.40 | 0.43 | 0.52 |
| $Zn^{2+}$ | 156 | 200 | 205 | 293 | 0.27 | 0.31 | 0.36 | 0.40 |

Zeolites belong to the group of framework aluminosilicates, the infinite aluminosilicate framework of which is formed when articulating through common vertices of the $[AlO_4]^{5-}$ and $[SiO_4]^{4-}$ tetrahedra [17]. The frames have a regular system of cavities, interconnected by channels, in which there are metal cations and water molecules that are able to freely be removed and absorbed by the structure, due to which ion exchange occurs. The structural formula of zeolite can be represented as follows: $M_{m/n}[(AlO_2)_x(SiO_2)_y]\cdot zH_2O$ where x+y- is the sum of tetrahedra in the unit cell, m- is the number of cations M (potassium, sodium, calcium, magnesium) , n-is the valence of the cation [18].

Of the more than 40 mineral species and varieties of zeolites known in nature, only a few satisfy the requirements for practical use, namely: they form large almost monomineral concentrations and at the same time have the corresponding consumer properties (adsorption, ion exchange, acid and heat resistance, etc.) [17].

Zeolites of the Taizhuzgen deposit belong to clinoptilolite aluminosilicates with a content of clinoptilolite of 55-60%, mainly sodium and potassium type [19]. Table 4 presents the average chemical composition of zeolites in Kazakhstan deposits [20,21,22,29].

It was established that these zeolites comply with the RSS-99 standards for the content of toxic elements and radionuclides. The mineral composition of zeolites is represented by clinoptilolite, montmorillonite, quartz, feldspar and rock fragments [23].

For use in water purification processes, significant advantages of zeolites from East Kazakhstan deposits were revealed: high sorption properties; availability (deposits are located near consumers); low cost (in comparison with other technological reagents); resistance to temperatures and climatic conditions during transportation, storage and exploitation. The indicated advantages make it possible to successfully use natural zeolites for treating groundwater in the region, which are characterized by high hardness and a high content of heavy non-ferrous metal ions. As the results of the study showed, the zeolites of the Taizhuzgen deposit improve water quality by 30-50%, however, as a result of activation, the sorption ability of the zeolite rises to 85-90% [24].

Table 4 – The averaged chemical composition of zeolite tuffs of Kazakhstani deposits

| Components | Deposits, average content of components. % | |
|---|---|---|
| | Taizhuzgen | Shankanai |
| $SiO_2$ | 65,5 | 59,86 |
| $TiO_2$ | 0,20 | 0,46 |
| $Al_2O_3$ | 14,27 | 14,39 |
| $Fe_2O_3$ | 0,87 | 5,21 |
| FeO | 0,53 | - |
| $MnO_2$ | 0,04 | - |
| MgO | 0,8 | 1,71 |
| CaO | 3,2 | 4,93 |
| $Na_2O$ | 2,04 | 1,07 |
| $K_2O$ | 2,83 | 1,56 |
| $SO_2$ | 0,07 | - |
| $H_2O$ | 10,0 | 3,61 |
| $P_2O_5$ | - | 0.09 |
| MnO | - | 0.09 |
| Fe | - | 3,85 |
| Sr | - | 0,31 |
| l.o.i. | - | 3,38 |

Note: l.o.i.- loss on ignition

Plotnikov Ye.V. et al. [25, 26] studied the sorption and surface characteristics of filtering materials based on natural modified minerals. The obtained results confirm the possibility of using modified zeolites as a load for cleaning water from bacteria only when the size of the sorbent fraction is less than 0.1 mm (table 5). In general, zeolites have shown great efficiency in the removal of microbiological contaminants. The most promising is the use of a mixture of various fractions to obtain acceptable levels of water purification and hydrodynamic resistance (table 6).

Table 5 – Surface characteristics of modified mineral sorbents

| Material | Fraction, mm | Specific surface, $m^2/g$ | Specific pore volume, $cm^3/g$ | The average pore size, nm |
|---|---|---|---|---|
| Zeolite | <0,1 | 25,351 | 0,011 | 1,715 |
| Zeolite | 0,1-0,5 | 24,158 | 0,010 | 1,715 |
| Zeolite | 0,5-1,0 | 21,241 | 0,009 | 1,716 |
| Glauconite | <0,1 | 44,74 | 0,013 | 1,714 |
| Glauconite | 0,1-0,5 | 39,69 | 0,019 | 1,717 |
| Glauconite | 0,5-1,0 | 30,715 | 0,017 | 1,715 |

Table 6 - Assessment of the degree of extraction of microbiological contaminants from aqueous media with modified sorbents

| Parameter | Fraction (<0,1 mm) | | Fraction (0,1-0,5 mm) | | Fraction (0,5-1,0 mm) | |
|---|---|---|---|---|---|---|
| | Zeolite | Glauconite | Zeolite | Glauconite | Zeolite | Glauconite |
| The number of bacteria after filtration (CFU/ml) | 0 | 0 | $1,7 \cdot 10^5$ | $7 \cdot 10^6$ | $1,0 \cdot 10^7$ | $1,0 \cdot 10^7$ |

The mineralogical composition and thermal stability of the natural zeolite of the Taizhuzgen deposit are studied. The main component of the zeolite-containing rock is clinoptilolite. The possibility of using this zeolite to purify water from iron under dynamic conditions is shown. At the initial concentration of $Fe_{total}$ 1,5 mg/l, the removal efficiency was 100% after passing 300 ... 500 ml of water through the filter. The ion-exchange properties of zeolite by $Fe^{2+}$ ions were studied: the total ion-exchange capacity of $Fe^{2+}$ is 18,2 g-equiv / $m^3$ [27]. Studies have shown that Taizhuzgen zeolite is a good sorption material and can be recommended in further studies with the aim of developing filters for water purification used in water treatment processes of the dairy industry.

Thus, the use of zeolites for the preparation of water in the dairy industry:

- does not require high maintenance costs and reagents;
- makes it possible to additionally extract toxic substances in contrast to the reagent method;

- characterized by simplicity and accessibility in operation, in contrast to the electrodialysis method;
- does not require thorough water purification from mechanical impurities, the use of reagents to prevent precipitation;
- does not require maintaining a relatively high speed of mechanical cleaning;
- low cost of the installation of purification, unlike the reverse osmosis method [16].

Methods that prevent or exclude the ingress of harmful compounds into food products are quite limited, time-consuming and expensive, cumbersome, require frequent regeneration of plants and materials. Their use is effective at high productivity and high initial hardness of water. The cost of materials, for example, ion-exchange resins, is incommensurably higher than that for natural materials, for example, natural zeolites. However, their capabilities are much wider. Natural zeolites can soften water, release it from metal ions, organic impurities, colloids, and trap microorganisms. The use of zeolites of various modifications will make it possible to obtain water of the desired composition, microbiologically pure [28].

**Conclusion.** The chemical composition of zeolite tuffs of Kazakhstan deposits is identical to the chemical content of the known zeolite deposits of Ukraine, Georgia and Russia. Studies of drinking water from various regions of the country, conducted by the authors of the present paper, led to the conclusion about the presence of iron and cadmium in drinking water in quantities exceeding the allowable regulatory limits. As shown by the results of experimental studies, zeolites of the Taizhuzgen deposit significantly improve water quality. Thus, the studies performed have shown that the use of zeolites for preparing water in the dairy industry will make it possible to obtain dairy products with higher hygiene safety indicators.

**М.К. Алимарданова[1], Ж.Б. Хамзина[1], Д.Т. Балпанова[2]**

[1]Алматы технология университеті, Қазақстан;
[2]С.Д. Асфендияров атындағы Қазақ ұлттық медицина университеті, Алматы, Қазақстан

**ҚАЗАҚСТАНДЫҚ ЦЕОЛИТТЕР – СҮТ САЛАСЫНДАҒЫ СУ ӨҢДЕУ ҮДЕРІСІНДЕ ПЕРСПЕКТИВАЛЫҚ МАТЕРИАЛ РЕТІНДЕ**

**Аннотация.** Қазіргі уақытта тағамға қолдануға жарамды таза су көзі кездеспейді. Соның салдарынан тағам саласының кез келген кәсіпорнында өнім өндіру үшін қажетті суды тазартуға байланысты бірқатар жүйелі амалдар жүзеге асырылады. Сүт шикізатын өңдеу үдерістерінде кәсіпорындарды сумен жабдықтау қоғамдық су құбыры желісінен және/немесе артезиан ұңғымасынан жүзеге асырылады. Мақалада су қауіпсіздігін қамтамасыз ету үшін табиғи цеолиттерді қолданудың пайдалылығы қарастырылған. Түрлі кен орындарындағы қазақстандық цеолиттерге қатысты зерттеу материалдары мен оны тәжірибеде қолдану жолдары зерттелді. Тәжірибелік зерттеу нәтижелері көрсеткендей, Тайжөзген кен орнының цеолиттері су сапасын едәуір жақсартады. Белсендіру нәтижесінде цеолиттің сорбциялық қабілеті 85-90%-ға дейін артады. Цеолиттердің өндірістік сынақтары аталған минералды шикізаттың ерекше мүмкіндіктерін көрсетеді. Табиғи цеолиттер арзан, механикалық тозуға төзімді және регенерациядан кейін бірнеше рет пайдаланылуы мүмкін. Зерттеулер көрсеткендей, Тайжөзген цеолиті жақсы сорбциялық материал болып саналады және сүт саласының су дайындау үдерісінде қолданылатын суды тазарту үшін сүзгілерді әзірлеу мақсатында кейінгі зерттеулерде ұсынылуы мүмкін.

**Түйін сөздер**: қауіпсіздік, су, цеолиттер, сорбциялық қабілет, цеолиттер артықшылығы, модификация

**М.К. Алимарданова[1], Ж.Б. Хамзина[1], Д. Т. Балпанова[2]**

[1]Алматинский технологический университет;
[2]Казахский национальный медицинский университет имени С.Д.Асфендиярова, Алматы, Казахстан

**КАЗАХСТАНСКИЕ ЦЕОЛИТЫ КАК ПЕРСПЕКТИВНЫЙ МАТЕРИАЛ В ПРОЦЕССАХ ВОДОПОДГОТОВКИ МОЛОЧНОЙ ОТРАСЛИ**

**Аннотация.** В современном мире практически нет источников чистой воды, пригодной для потребления человеком. В результате на любом предприятии пищевой промышленности осуществляется ряд последовательных операций, связанных с очисткой воды, необходимой для производства продукции. Во всех

процессах переработки молочного сырья водоснабжение предприятий осуществляется из водопроводной сети общего пользования и / или из артезианской скважины. В статье рассматривается целесообразность использования природных цеолитов для обеспечения безопасности воды. Изучены данные исследований казахстанских цеолитов различных месторождений и их применение на практике. Как показали результаты экспериментальных исследований, цеолиты месторождения Тайжузген значительно улучшают качество воды. В результате активации сорбционная способность цеолита повышается до 85-90%. Производственные испытания цеолитов свидетельствуют о значительном потенциале этого минерального сырья. Природные цеолиты дешевы, устойчивы к механическому износу и могут быть повторно использованы после регенерации. Исследования показали, что цеолит Таизузгена является хорошим сорбционным материалом и может быть рекомендован в дальнейших исследованиях с целью разработки фильтров для очистки воды, используемых в процессах водоподготовки в молочной промышленности.

**Ключевые слова:** безопасность, вода, цеолиты, сорбционная способность, преимущества цеолитов, модификация.

**Information about authors:**

Alimardanova Marian Kalabaevna, Academician of the Academy of Agricultural Sciences of the Republic of Kazakhstan, Doctor of Technical Sciences, Professor of the "Technology of Food Products" Dept., Almaty Technological University, Almaty, Kazakhstan; alimardan.matu4@mail.ru; https://orcid.org/0000-0003-4861-7862;

Khamzina Zhulduz Bazaralievna., 2 year doctoral student of the specialty 6D073200-«Food safety», Almaty Technological University, Almaty, Kazakhstan; zhuldyz_hamzina@mail.ru; https://orcid.org/0000-0002-1530-5822;

Balpanova Didara Tleulessovna, Candidate of Chemical Sciences, Assistant Professor of the «Chemistry» Dept., Asfendiyarov Kazakh National Medical University, Almaty, Kazakhstan; balpanova00@mail.ru; https://orcid.org/0000-0002-6942-6435

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