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POROUS STRUCTURE AND ADSORPTION PROPERTIES OF MAGNESIUM SILICATES SYNTHESIZED BY THREE ROUTES

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The aim of presented work was to synthesize the ecologically friendly sorbents using low cost reagents based on magnesium silicates by precipitation, hydrothermal and sol-gel methods. Morphology of materials obtained was investigated by means of thermogravimetric analysis (TG-DTA), low temperature adsorption/desorption method, scanning and transmission electron microscopic studies (SEM and TEM). It has been found that all sorbents are obtained in a form of amorphous layer-structure magnesium silicates with the micro- and mesoporous structure. Based on low temperature nitrogen isotherms, the specific surface area and volume of micropores calculated by the Brunauer-Emmet-Teller (BET) and Barrett-Joyner-Halenda (BJH) methods were the greatest for the sample obtained by sol gel method ($S_{BET} = 640 \text{ m}^2/\text{g}$, $V_{\text{micro}} = 0.26 \text{ cm}^3/\text{g}$) while the sample synthesized by precipitation had the biggest volume of mesopores among the materials investigated ($V_{\text{meso}} = 0.39 \text{ cm}^3/\text{g}$). As follows from TEM images, all samples consist of particles with the size from 10 up to 50 nm, the sample synthesized by sol gel method had the most homogeneous structure (MgSi-3). The ion exchange capacities of materials in the process of Cs^+ , Sr^{2+} , Cu^{2+} , and Co^{2+} removing from aqueous solution were determined and it was found that these properties depend on the method of materials obtaining. Data showed that magnesium silicate synthesized by precipitation method has the higher capacity toward the heavy metal cations compared to the radionuclides (1.56 and 0.96 mmol/g for cobalt and copper, respectively). For two samples synthesized by hydrothermal and sol-gel methods the increasing was fixed of capacity towards cesium and strontium ions that could be explained by the significant amounts of pores approx 2.6 nm radii in their structure. Experimental data were fitted to the Langmuir models. Analysing the data of adsorption studies, it was noted that all materials obtained can be used in adsorption technology for purification of water from heavy metal ions and radionuclides.

Keywords: magnesium silicates, morphology, adsorption, heavy metals, impact of synthetic route

INTRODUCTION

Magnesium silicate is a compound of magnesium oxide and silicon dioxide with empirical formula $\text{MgO}\cdot\text{SiO}_2\cdot n\text{H}_2\text{O}$, the most common method for its synthesis is via a precipitation reaction between a soluble metal silicate (e.g., sodium orthosilicate, sodium metasilicate, potassium silicate, tetraethyl ortho silicate) and a soluble magnesium salt (e.g., magnesium sulphate, nitrate, or chloride) [1]. Silicate anions combine with Mg^{2+} ions in solution forming the low solubility magnesium silicates [2–4]. It is known that porous structure and surface of materials obtained depend on the main conditions of synthesis – nature of salts, ratio of components, temperature of drying, etc. Hydrothermal, microwave and ultrasound treatment of precursors positively improve on the properties of materials [5–7].

Silicates attract the increasing attention of scientists due to their capability the selectively

adsorb a lot of ions: cesium, strontium, heavy metals and lanthanides [6, 8, 9]. Practice poses well-known requirements to ion exchange materials, first of all, to low cost of their fabrication, to certain parameters of porous structure and to good cation exchange properties. The main advantage of synthetic materials is the possibility to control the textural and surface properties by correlation of synthetic conditions on the stage of mixing reagents and drying precursors [10, 11]. Much works in this field deal with a design the new methods and improvement existed for obtaining silicates of various metals such as titanium, zirconium, aluminum and calcium [12–15]. Magnesium silicates as sorbents have been intensively investigated only last decades.

The aim of presented work was to synthesize the ecologically friendly sorbents using low cost reagents (magnesium sulphate and sodium metasilicate) with the high surface area and to examine the adsorption capacity of materials

obtained towards Cs^+ , Sr^{2+} , Cu^{2+} and Co^{2+} ions. For synthesis were used the simple methods such as precipitation, hydrothermal and sol-gel technology. The solid phases are investigated by thermogravimetric analysis, scanning and transmission electron microscopic studies, low temperature adsorption/desorption method for better comprehending the morphology of sorbents. The ion-exchange capacity for desired metal ions have been evaluated and impact of method used on the properties of materials are established. Adsorption experiments have been supported by fitting of isotherms by Langmuir model.

EXPERIMENTAL

All chemicals used ($\text{MgSO}_4 \cdot 9\text{H}_2\text{O}$, NaOH , CsCl , $\text{SrCl}_2 \cdot 6\text{H}_2\text{O}$, $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ and $\text{CuCl}_2 \cdot 5\text{H}_2\text{O}$) were of analytical grade (OSTHIM, Ukraine) and used without further purification. The magnesium content in solutions was controlled by a complexometric titration [16]. The aqueous sodium metasilicate solution $\text{SiO}_2/\text{Na}_2\text{O}$ with molar ratio = 3.14 (Na_2O – 9.3 %, SiO_2 – 28 %, density – 1.42 g/L) was produced by Ukrainian enterprise (PJSC “Zaporizhskloflus”) and analyzed by acid-base titration method.

Magnesium silicates ion exchangers were prepared by three methods. The first one (precipitation method, MgSi-1) consists in drop wise addition of solution magnesium sulphate (0.5 M) to sodium metasilicate with equimolar ratio Mg/Si. The precipitate forming was activated by addition of diluted NaOH solution to the mixture up to $\text{pH} = 11$ [1]. The process was carrying out with continuous stirring in a water bath adjusted at room temperature. The precipitate formed was kept in the mother solution overnight. The powder were rewashed for several times by distilled water to remove unreacted reagents and the final products were dried at 120 °C, ground, sieved and store at room temperature.

Obtaining of second material (MgSi-2) included the hydrothermal treatment of reagents. At first, the magnesium sulphate solution (0.5 M) was added dropwise to the sodium silicate solution (0.5 M) heated up to 60 °C to generate suspension followed by continuous stirring for 1 h. Then, the suspension obtained was added to a 100 mL Teflon-lined stainless steel autoclave and maintained at 180 °C for 12 h to form hydrogel. The last one was repeatedly filtered and washed

by deionized water to remove Na^+ and SO_4^{2-} ions. Finally, the gel was dried at room temperature on air for 24 h to form powder.

Starting reagent for synthesis of sample using sol gel technology (MgSi-3) was also aqueous solution of magnesium sulphate (0.5 M). With vigorous stirring in this solution was added dropwise the accounted volume (needs for final ratio $\text{Mg}/\text{Si} = 1$) of concentrated sodium silicate (6.6 M) heated up to 60 °C. Spherical granules had formed in solution in the place of drop falling was filtered after day, washed by deionized water and diluted acid (HNO_3) to remove the excess of Na^+ and dried at room temperature as previous samples.

For researching the chemical stability of sorbents an about 0.05 g of magnesium silicates were mixed with 50 ml of deionized water solution at room temperature (25 ± 1 °C) and kept for 24 h with intermittent shaking. The total amount of Mg^{2+} (mmol/L) dissolved in the solutions was determined using complexometric titration [16].

X-ray diffraction (XRD) patterns were obtained on a DRON – 4-07 diffractometer (LOMO, Russia) using CuK_α radiation ($\lambda = 0.15418$ nm) in the 2θ angle range of 10–60° with a 0.02 step. Thermogravimetric analysis of samples (TG-DTA) was carried out in the interval of 20–900 °C in the air with a rate of temperature increase of 10⁰/min on a Derivatograph – Q equipment (Hungary). Specific surface areas and pore size distributions for the synthesized samples were calculated from nitrogen adsorption/desorption curves (NOVA 2200e, Quantachrome, USA) using the Nova Win 2.0 software. The total surface area of the materials S_{total} was calculated by the Brunauer-Emmet-Teller method (BET). The total pore volume (V_{total}) was calculated from the volume of nitrogen adsorbed converted to liquid at a pressure close to $P/P_0 = 1$. To acquire the volume and radii of mesopores (V_{meso} , R_{meso}), Barrett-Joyner-Halenda (BJH) was used. The average pore radii (R_{pore}) was determined from the total pore volume (V_{total}) of the materials and its specific surface area (S_{total}) by equation $R_{pore} = 2V_{total}/S_{total}$. The micropore volume (V_{micro}) was calculated by subtracting the value of V_{meso} from V_{total} . Pore radii distributions were obtained from isotherms in terms of the density functional theory (DFT).

Scanning (SEM) and transmission electron microscope (TEM) investigation was done with a

SEM JSM 6490 LV and a JEM 2100 F microscope (JEOL, Japan), respectively.

Sorption properties of the magnesium silicate samples were studied under static conditions. Concentrations of Cs^+ , Sr^{2+} , Cu^{2+} and Co^{2+} in initial and equilibrium solutions of chlorides were measured using a Shimadzu AA 6300 (Shimadzu, Japan) atomic absorption analyzer.

Adsorption capacity (q_e , mmol/g) was calculated by the next equation:

$$q_e = (C_0 - C_e)V/m$$

where C_0 and C_e are the initial and equilibrium metal concentration in solution, respectively, mmol/L; V is the aliquot volume (L); and m is the mass of the adsorbent (g).

Experimental data were fitted into the Langmuir models, which are common used to describe the liquid–solid systems [17] by the following equations:

$$q_e = Q_0 K_L C_e / (1 + K_L C_e)$$

where q_e is the adsorption capacity (mmol/g); C_e is the equilibrium concentration of the adsorbate (mmol/L); Q_0 is the maximum adsorption capacity of the adsorbent (mmol/g); and K_L is the Langmuir sorption equilibrium constant (L/mmol). To evaluate the correlation between the experimental data and theoretical models, the coefficient of determination (R^2) was calculated.

RESULTS AND DISCUSSION

Ion exchange materials have been obtained at 120 °C in the amorphous phases. Thermal decomposition of samples has been investigated by differential thermal analyses (data for sample MgSi-3 are given in Fig. 1), other materials decompose by similar way. The TG curve of material shows the first loss in mass accompanied by endothermic peaks in DTA curve at 80 °C, associated with the removing water from hydrated magnesium silicate gel [3, 18, 19]. After that a continuous loss in mass is gradually observed up to 650 °C (9.4 %) with the main peaks in region ~ 220–350 °C. The second peak is possibly due to the loss of constitution water with transformation amorphous hydroxides to poorly crystalline layer-structure magnesium silicate [18]. According the X-ray data poorly crystalline phases formation is started at ~650 °C (weak broad diffraction peaks

around $2\theta = 28\text{--}35^\circ$ and $\sim 43^\circ$, Fig. 2) regardless the method used. As seems from data [18, 19], these peaks indicate the beginning to form the crystalline enstatite MgSiO_3 (JCPDS 011-0273) that is finished after 900 °C.

Next step of investigation is researching the porous structure of materials by means of two methods. Data received by low-temperature nitrogen adsorption/desorption method are presented in Table 1 and Fig. 3 *a, b*. Nitrogen adsorption/desorption isotherms of the all samples of magnesium silicates according the IUPAC classification [20] belong to the II type with the hysteresis loop of the H3 type (Fig. 3 *a*). Generally, the magnesium silicates obtained by precipitation and sol gel method (MgSi-1 and MgSi-3) manifest the highest BET specific surface area, total pore volume and volume of micropores calculated by the BET and BJH methods (Table 1). No micropores it was fixed in the porous structure of material synthesized by using hydrothermal treatment of reagents (MgSi-2). It should be noted that parameters of porous structure of MgSi-1 and MgSi-3 are equal to magnesium silicate samples synthesized by more difficult methods with using of expensive reagents [9, 21].

Analyzing the pore size distribution calculated from desorption branches of the isotherms in term of DFT method (presented in Fig. 3 *b*), it is found that in all samples there are mesoporous with approx. 1–3 nm radii. Samples synthesized by precipitation method (MgSi-1) contain the great amount of micropores supported by data received using BET and BJH methods (V_{micro} , Table 1). In the case of samples synthesized by hydrothermal and sol-gel method (MgSi-2, MgSi-3) it has been indexed the great amount of mesopores approx. 2.6 nm that are absent in the structure of the first one.

In Fig. 4 are presented TEM and SEM images for samples synthesized by precipitation and sol gel technology (MgSi-1 and MgSi-3, respectively). According TEM studies all samples consist of particles with the size from 10 up to 50 nm. The main differences in texture of sample synthesized by sol gel method (MgSi-3) are the smallest particles size (~20 nm), the most homogenous structure among samples investigated and the honeycomb-like surface (SEM, *b*).

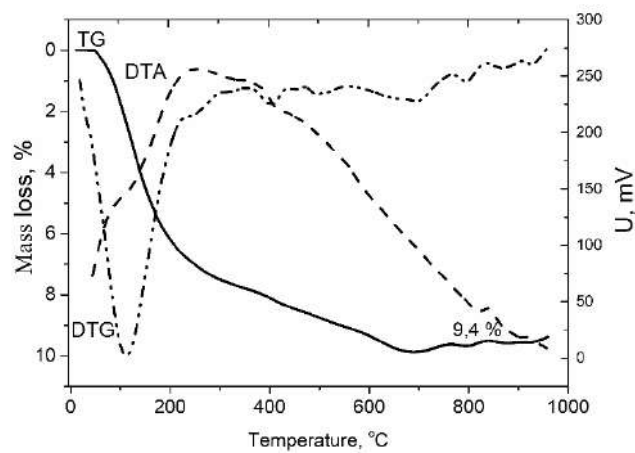


Fig. 1. Thermal analysis of magnesium silicates obtained by sol-gel method (MgSi-3)

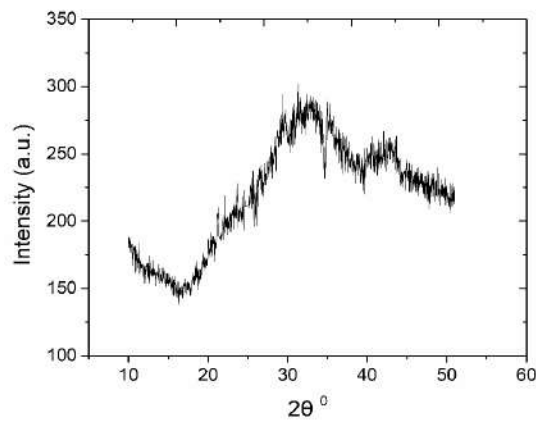


Fig. 2. X-ray diffraction data for magnesium silicate obtained by sol-gel method (MgSi-3) treated at 600 °C

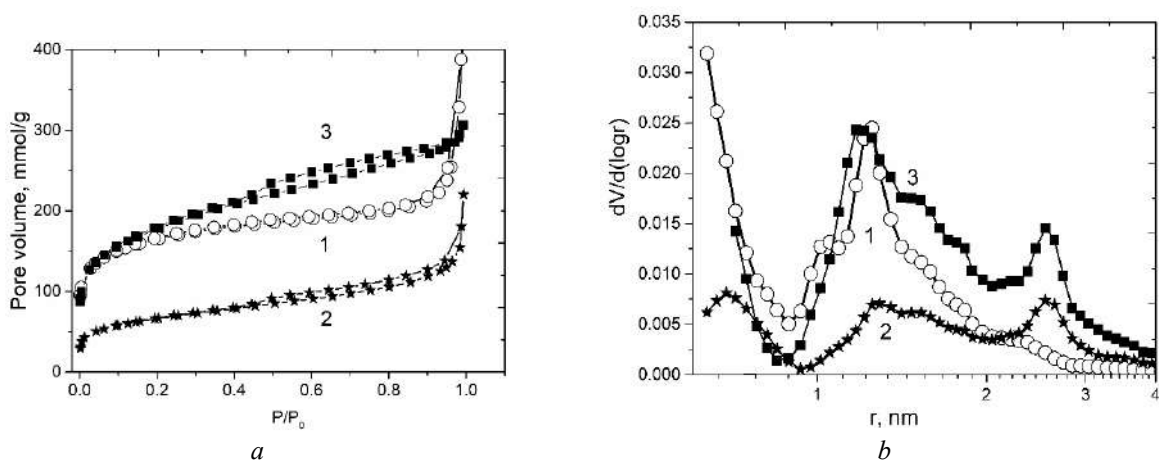


Fig. 3. Nitrogen adsorption/desorption isotherms (a) and pore size distributions obtained in term of DFT method (b) for the magnesium silicates synthesized by precipitation MgSi-1 (1), hydrothermal MgSi-2 (2) and sol gel methods MgSi-3 (3)

Table 1. Porosity data for magnesium silicates synthesized by three methods

Samples	MgSi-1	MgSi-2	MgSi-3
BET surface area, S_{total} (m ² /g)	597	117	640
Total pore volume, V_{total} (cm ³ /g)	0.64	0.25	0.48
Mesopore volume, V_{meso} (cm ³ /g)	0.39	0.21	0.22
Micropore volume, V_{micro} (cm ³ /g)	0.25	0.04	0.26
Mesopore radius, R_{meso} (nm)	1.7	1.7	1.9
Average pore radius, R_{pore} (nm)	2.1	4.3	1.5

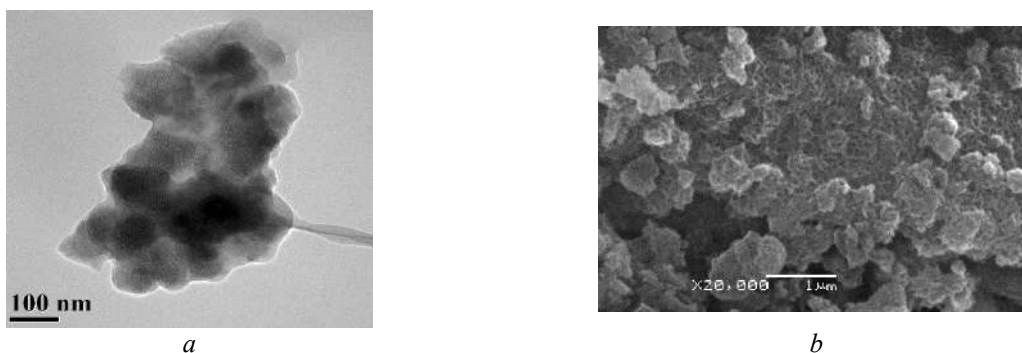


Fig. 4. TEM image of magnesium silicates obtained by precipitation MgSi-1 (a) and SEM image of sample obtained by sol gel method MgSi-3 (b)

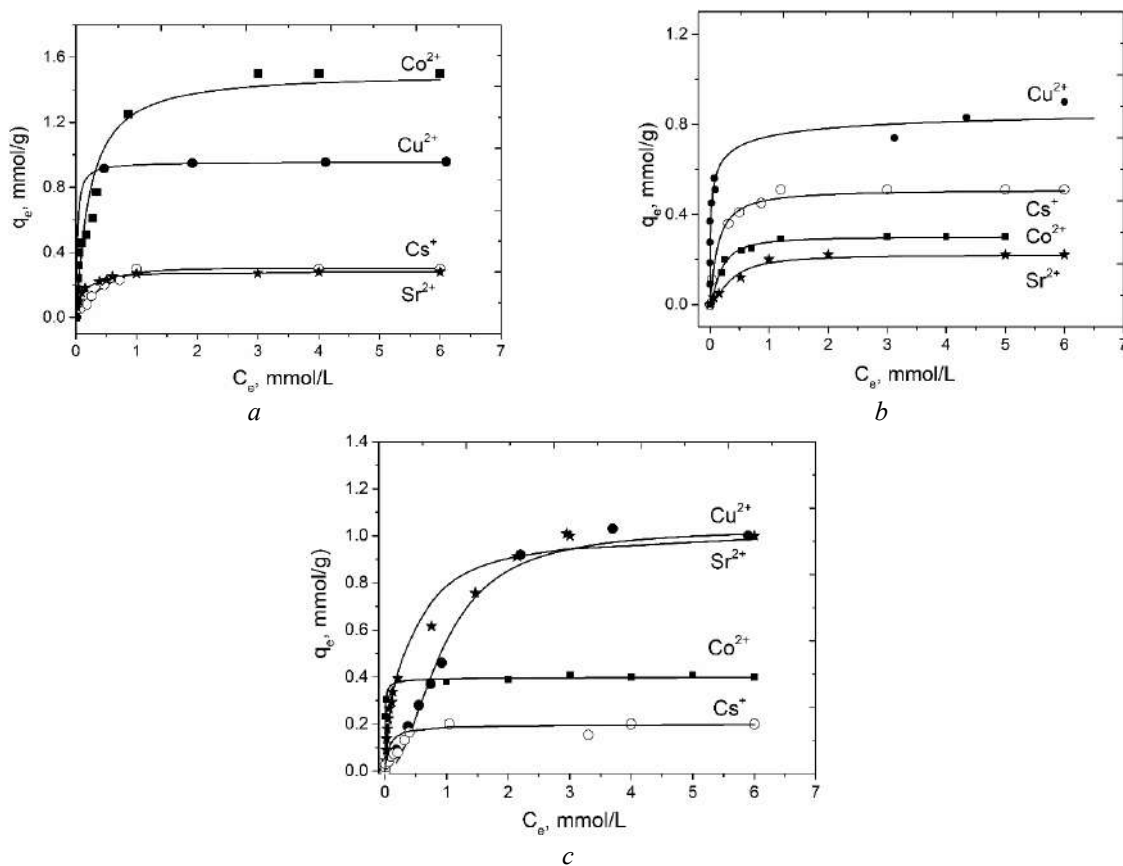


Fig. 5. Sorption isotherms for Cs^+ , Sr^{2+} , Cu^{2+} and Co^{2+} adsorbed onto the samples MgSi-1 (a), MgSi-2 (b) and MgSi-3 (c)

Table 2. Langmuir isotherm constants for the adsorption of cesium, strontium, copper and cobalt ions onto magnesium silicates

Samples	MgSi-1	MgSi-2	MgSi-3
Cs⁺			
Q_0 (mmol/g)	0.32	0.52	0.20
K_L (L/mmol)	2.92	8.74	15.2
R^2	0.99	0.98	0.93
Sr²⁺			
Q_0 (mmol/g)	0.28	0.24	1.03
K_L (L/mmol)	10.5	2.49	3.73
R^2	0.99	0.98	0.97
Cu²⁺			
Q_0 (mmol/g)	0.96	0.86	0.98
K_L (L/mmol)	42.5	19.4	1.90
R^2	0.99	0.98	0.99
Co²⁺			
Q_0 (mmol/g)	1.56	0.31	0.40
K_L (L/mmol)	8.01	6.32	32.1
R^2	0.94	0.99	0.98

Chemical stability studies of materials have indicated that sample MgSi-3 obtained by sol gel method is stable at investigated pH = 7, when the other materials are partially soluble (0.9 and 1.2 mmol/L, respectively).

Fig. 5 *a, b, c* and Table 2 summarize the results of Cs⁺, Sr²⁺, Cu²⁺ and Co²⁺ adsorption by sorbents based on the magnesium silicates. Data show that magnesium silicate synthesized by precipitation method has the higher capacity toward the heavy metal cations compared to the radionuclides (1.56 and 0.96 mmol/g for cobalt and copper, respectively). This sequence is in the good accordance with the hydrated radii of the exchanging ions [22]: the ions with smaller hydrated radii (Cu²⁺ – 0.73 Å, Co²⁺ – 0.74 Å) easily enter the pores of the exchanger, resulting in the higher adsorption. Possible mechanism for the higher selectivity of Cu²⁺ and Co²⁺ ions compared to other metal ions is interpreted by authors [3] considering the great similarity between the ionic radii of copper, cobalt and Mg²⁺ (0.72 Å) leads to including of copper or cobalt ions in the part of the site of magnesium. It can be seen from Fig. 5 and Table 2 that the removal rates of metals investigated decrease in the following order: Co²⁺ > Cu²⁺ > Cs⁺ = Sr²⁺ (MgSi-1), Cu²⁺ > Cs⁺ > Co²⁺ > Sr²⁺ (MgSi-2), Cu²⁺ > Sr²⁺ > Co²⁺ > Cs⁺ (MgSi-3). It should be noted that two samples with the pores of 2.6 nm radii (Fig. 3) show the greater adsorption capacity towards bigger ions of Cs⁺ (1.91 Å) and Sr²⁺ (1.28 Å) than the first one (MgSi-1).

CONCLUSIONS

Amorphous layer-structure magnesium silicates were obtained by precipitation, hydrothermal and sol gel methods from magnesium sulphate and sodium metasilicate solutions with equimolar ratio of components. It was found that magnesium silicates obtained by precipitation and sol gel method have the biggest BET specific surface area (597 and 640 m²/g, respectively) and volume of micropore (~0.25 cm³/g), unlike the porous structure of material synthesized by hydrothermal method. Morphology studies showed that the sol-gel method obtained sample has the smallest particles size (~20 nm), the most homogenous structure among samples investigated and the honeycomb-like surface. Investigation of adsorption properties of sorbents determined that magnesium silicate synthesized by precipitation method has the higher capacity toward the heavy metal cations compared to the radionuclides that is explained by a good accordance with hydrated radii of the exchanging ions and magnesium ions. For two samples with the pores of 2.6 nm radii the increasing adsorption capacity was fixed towards the bigger ions of Cs⁺ and Sr²⁺. Sorption isotherms for all cations were investigated and the data showed the applicability of Langmuir isotherm for all cases. Analyzing the data of adsorption studies, it was noted that all materials obtained can be used in adsorption technology for purification of water from heavy metal ions and radionuclides.

Порувата структура та адсорбційні властивості силікатів магнію, синтезованих трьома методами

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Метою представленої роботи був синтез безпечних для навколишнього середовища сорбентів на основі силікатів магнію з використанням недорогих реагентів та простих методів, таких як осадження, гідротермальний та золь-гель технологія. Морфологію отриманих матеріалів було досліджено за допомогою термогравіметричного аналізу (ТГ-ДТА), методу низькотемпературної адсорбції/десорбції азоту, скануючої та трансмісійної електронної мікроскопії (СЕМ та ТЕМ). Встановлено, що всі сорбенти є аморфними шаруватими силікатами магнію з мікро- та мезопоруватою структурою. На підставі ізотерм адсорбції азоту було встановлено, що питома площа поверхні та об'єм мікропор, розраховані за методами Брануєра-Еммета-Теллера (БЕТ) і Барретта-Джонсона-Галенди (БДГ), є найбільшими для зразка, одержаного золь-гель методом ($S_{\text{БЕТ}} = 640 \text{ м}^2/\text{г}$, $V_{\text{мікро}} = 0,26 \text{ см}^3/\text{г}$), тоді як зразок, синтезований методом осадження, має найбільший об'єм мезопор ($V_{\text{мезо}} = 0,39 \text{ см}^3/\text{г}$). Згідно мікрофотографій, одержаним методом ТЕМ, усі зразки складаються з частинок розміром від 10 до 50 нм, найбільш однорідну структуру мав зразок, синтезований золь-гель методом (MgSi-3). Визначено іонообмінну ємність одержаних сорбентів у процесах видалення Cs^+ , Sr^{2+} , Cu^{2+} та Co^{2+} з водних розчинів та встановлено, що ці властивості значно залежать від методу синтезу матеріалів. Виявлено, що силікат магнію, синтезований методом осадження, має найвищу адсорбційну ємність по відношенню до катіонів важких металів (1.56 і 0.96 ммоль/г для кобальту та міді, відповідно), на відміну від радіонуклідів. Для двох зразків, синтезованих гідротермальним та золь-гель методами, було зафіксовано значне зростання ємності по відношенню до іонів цезію та стронцію, що можна пояснити значною кількістю пор радіусом 2.6 нм, виявлених у їхній структурі. Для обробки експериментальних даних було використано теоретичну модель Ленгмюра. Аналізуючи результати досліджень, можна зазначити, що всі одержані матеріали можуть бути використані як сорбенти для очищення води від іонів важких металів та радіонуклідів.

Ключові слова: силікати магнію, морфологія, адсорбція, важкі метали, вплив методу синтезу

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