IRSTI 31.17.15; 61.31.51 https://doi.org/10.26577/IJBCh202518111

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(Received 14 May 2025; received in revised form 28 May 2025; accepted 7 June 2025)

Resource-efficient technology for the utilization of serpentine technogenic waste with the production of magnesium oxide

Abstract. The article discusses the possibility of using serpentine and serpentine waste from Zhitikara deposit for the production of inorganic magnesium compounds: $MgSO_4$, $Mg(OH)_2$ and MgO. The aim is to produce magnesium sulfate, whose quality characteristics would further enable the production of high-purity magnesium hydroxide and magnesium oxide. A step-by-step process for obtaining magnesium compounds is proposed, initially starting with the purified magnesium sulfate solution, from which magnesium hydroxide and magnesium oxide can be produced. The proposed process differs from other known methods in the choice of reagent for neutralizing and purifying the initial sulfate solution to obtain high-purity magnesium hydroxide and magnesium oxide.

A technical and economic analysis was conducted on the efficiency of producing industrially important inorganic magnesium compounds from serpentine waste derived from the processing of chrysotile ore at Zhitikara deposit. The technology's resource intensity, energy efficiency, and economic feasibility of the new approach to processing serpentine waste for the production of magnesium compounds are shown. The advantages of using an acid method in combination with thermally activated serpentine for neutralizing and purifying the initial sulfate leaching solution are discussed. Based on experimental data obtained from studying magnesium extraction processes from serpentine, as well as the production of magnesium hydroxide and magnesium oxide from the purified magnesium sulfate solution, and the technical and economic analysis of each stage, it is concluded that the proposed process becomes more efficient when the final product is magnesium oxide. The potential for utilizing technogenic waste containing serpentine for sustainable magnesium compound production is considered.
Key words: serpentine, magnesium sulfate, magnesium hydroxide, magnesium oxide, resource efficiency.

Introduction

In many regions of the world, including Kazakhstan, serpentinite is contained in mining dumps. Serpentinite waste processing allows to reduce the cost of production due to the zero cost of raw materials, reduce the volume of technogenic accumulations, implement the principles of "green" technologies, and provide a comprehensive extraction of valuable elements (Mg, Si, Fe, etc.) in the form of usable compounds. These advantages contribute to the formation of waste-free technological processes, which is important from the point of view of environmental sustainability and rational environmental management.

Serpentinite anthropogenic wastes generated during mining and processing of chrysotile raw materials and non-ferrous metal ores (chromium, nickel, cobalt, etc.) have accumulated in the world in the volume of hundreds of millions of tons and represent an environmental hazard. These wastes contain significant amounts of magnesium and silicon, and have good reactivity after thermal and acid activation. The advantages of their use also include their location near mining enterprises, which facilitates recycling, environmental impact reduction and reclamation of anthropogenic landscapes.

Serpentinite is widely distributed in ultramafic rocks of the Earth's crust. Large deposits are located in Russia (Urals), China, Canada, Brazil and other countries. In Kazakhstan, large deposits of serpentinite (chrysotile) raw materials are located in the Kostanay region (Zhitikara deposit). These deposits are often associated with nickel and chromite ores, which in the future allows for the integrated development of deposits [1]. Currently, Zhitikara deposit is developed by Kostanay Minerals JSC. As a result of 65 years of operation some 300-350 million tons of chrysotile raw material processing waste were accumulated here.

It is known that serpentinites belong to complex silicates [2]. Their structure can be represented as a two-dimensional compound of two types of geometric shapes – tetrahedrons and octahedrons (Figure 1). The silica tetrahedrons have silicon atoms in the center, while the octahedral interlayer contains magnesium ions [3]. Typical chemical composition of serpentinite: MgO – 30-45%, SiO_2 – 35-45%, H_2O – 10-13%; FeO, Al_2O_3, CaO and other components are also present in small amounts. The high MgO content makes serpentinite a promising source of magnesium for production of industrially important compounds and magnesium-bearing products.

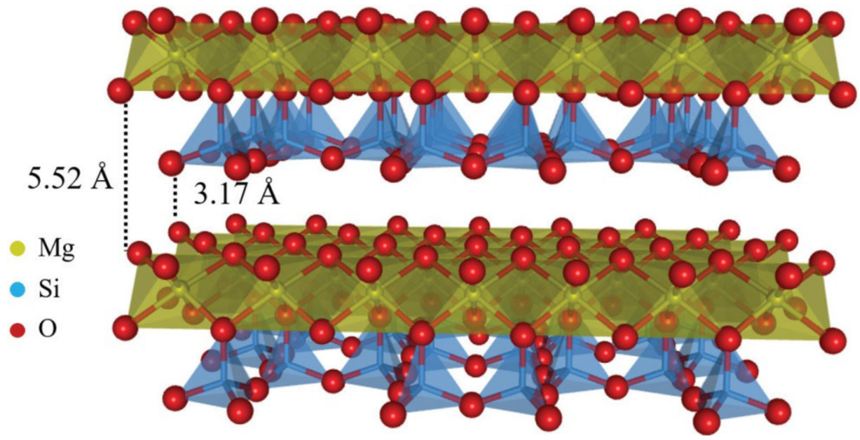

Figure 1 – Basic structure of serpentine [3]

The study of the possibility of using Zhitikara serpentinite or chrysotile raw material processing wastes is relevant, since there are no high-quality deposits of dolomite – traditional industrial raw material for obtaining inorganic magnesium compounds – on the territory of Kazakhstan. In this respect, the development of innovative scientific approaches to the use of accumulated serpentinite wastes of Zhitikara deposit can open new perspectives:

- creation of a processing cluster, development of new industries (production of magnesium compounds, magnesium fertilizers, etc.), transition to a sustainable "green" economy;

- integrated recycling allows to realize the "zero-waste" principle – waste becomes a raw material, and the accompanying by-products can be used in other industries.

Analysis of scientific literature in this area shows that so far there is no information on the forecast technical and economic indicators of potential products derived from serpentinite or serpentinite-containing industrial wastes of Zhitikara deposit using acid processing methods. Such data could be of interest from a practical point of view. Obviously, unsolved technological problems still prevent the transition to the economic evaluation of serpentinite application as a source of magnesium for the production of its compounds. This aspect is particularly relevant in the context of the development of new technological solutions aimed at processing industrial wastes and potential attraction of investments for their industrial implementation.

Various studies of physical and chemical properties and thermo-acid behavior of serpentinite ores, as well as serpentinite wastes of extraction and concentration of chrysotile of Zhitikara deposit [4-7] showed that serpentinites of this deposit can be considered as a promising source of magnesium for the production of inorganic compounds – magnesium sulfate, hydroxide and oxide, which are in demand in various industries of Kazakhstan.

As a first step, the goal was set to obtain magnesium sulfate, the qualitative characteristics of which will further provide the possibility of synthesizing magnesium hydroxide and oxide of high purity. In the study special attention is paid to the processes of leaching of serpentinite with sulfuric acid solution, as well as the selection of reagents for neutralization and purification of the productive solution of magnesium sulfate.

On the basis of experimental data obtained during the study of the processes of $MgSO_4$ extraction and step-by-step synthesis of magnesium compounds – $Mg(OH)_2$ and MgO, a technical and economic analysis was carried out to determine the resource and economic efficiency of their production from waste processing of serpentinite chrysotile raw materials of Zhitikara deposit.

Materials and methods

Dust-like material generated during extraction and concentration of chrysotile raw material of Zhitikara field (Kostanay region), with a particle size of 1-1.25 mm, was used as an anthropogenic waste in the research. Sulfuric acid (H_2SO_4, chemical purity) was used as a leaching reagent, and initial technogenic waste with the same particle size (1-1.25 mm) thermoactivated at 750°C for 1 hour was used as a reagent for neutralization and purification of leaching solution (magnesium sulfate).

The stoichiometric amounts of acid and man-made waste used for the interaction were calculated on the basis of the assumed reaction corresponding to the following equation:

$$Mg_3Si_2O_5(OH)_4 + 3H_2SO_4 = 3MgSO_4 + H_2SiO_3 + H_2SiO_4 + 2H_2O \quad (1)$$

All analytical studies were performed using a JSM-6490LV scanning electron microscope (JEOL, Japan) complete with an INCA Energy 350 energy dispersive microanalyzer system.

Methodology. Preparation of magnesium sulfate included two stages:

1) leaching of technogenic waste with sulfuric acid;

2) neutralization and precipitation of impurity metal ions by increasing the pH of the suspension to 7.8-8.0 using thermally activated serpentinite.

The processes of leaching, neutralization and solution purification were carried out in one reactor – a pyrex flask equipped with a thermometer, dosing device, filter and stirrer (Figure 2).

Figure 2 – Laboratory installation for obtaining magnesium sulfate solution: 1 – reactor, 2 – filter, 3 – thermometer, 4 – doser

Results and discussions

Analysis of modern researches and developments of technologies for obtaining magnesium compounds from serpentinite [8-13] shows that the main method of magnesium extraction in the form of its salts – sulfate, chloride, nitrate, as well as magnesium hydroxide and oxide – is acid leaching. In the process of acid treatment, serpentinite dissolves in solutions of inorganic acids (H_2SO_4, HCl and HNO_3), as a result of which magnesium passes into solution. The generalized reaction scheme is represented as follows:

$$(Mg, Fe)_3Si_2O_5(OH)_4 + 6H^+ \rightarrow Mg^{2+} + 2SiO_2 + Fe^{2+} + 5H_2O \quad (2)$$

Heat treatment at 600-800°C has also been found to activate the serpentinite structure by breaking the Mg-Si bonds [14-16]:

$$[Mg_3Si_2O_5(OH)_4] \xrightarrow{600\text{-}800^\circ C} MgO + SiO_2 + H_2O \quad (3)$$

The above reactions are presented in a simplified form, because in practice both dissolution of serpentinite in acids and its disintegration during calcination are complex processes depending on the type of the source rock and its chemical and mineral composition [17, 18].

The increase of alkaline properties of serpentinite after heat treatment in the range of 725-750°C is an established fact [19], which justifies its use as a reagent for neutralization of acidic solutions.

Taking into account the above-mentioned, within the framework of experimental studies, the sulfuric acid method of serpentinite leaching was used, as well as heat-treated serpentinite for neutralization and purification of the initial sulfate solution. The latter, in turn, was used to obtain magnesium hydroxide and oxide according to the scheme presented in Figure 3.

In accordance with the scheme, laboratory studies of the processes of step-by-step production of purified magnesium sulfate, magnesium hydroxide and magnesium oxide solution were carried out. For each stage, the optimum conditions of the processes were determined. A technical and economic analysis was performed on the basis of scaling of quantitative data (expenditures of raw materials, reagents and thermal energy) obtained during laboratory experiments to assess the resource and economic efficiency of obtaining magnesium compounds from pulverized serpentinite waste generated during the processing of chrysotile raw materials. Tables 1-3 present specific costs, as well as resource and economic efficiency indicators of each stage of obtaining magnesium sulfate, magnesium hydroxide and magnesium oxide, calculated on the basis of fixed costs and material and energy balances.

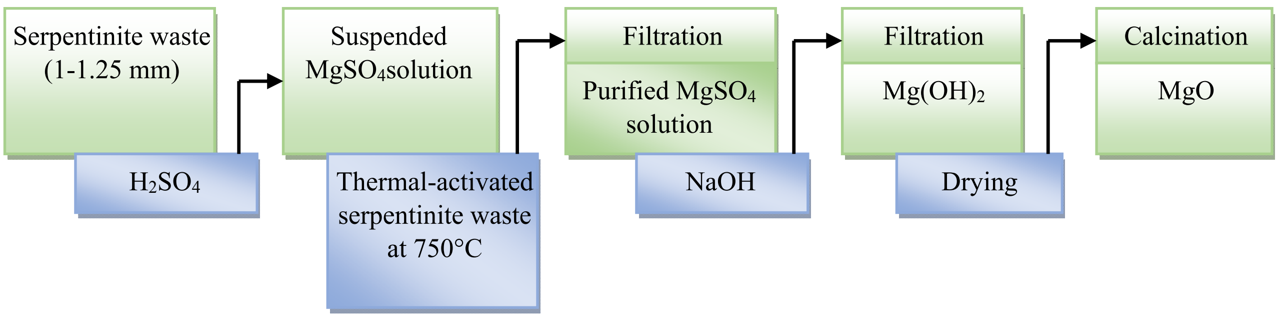

Figure 3 – Scheme of magnesium oxide production from serpentinite waste

Table 1 – Specific consumption of raw materials and energy resources for production of 1 ton of $MgSO_4 \cdot 7H_2O$ and resource and energy

Type of resource	UoM	Specificconsumption	Designation
Specific consumption of raw materials and energy resources for production of 1 t of $MgSO_4 \cdot 7H_2O$			
ITW (industrial and technical waste)	tons	0.400	C_raw
ITW-TA (industrial and technical waste thermoactivated at 750°C, for neutralization of acid slurry)	-	0.400	C_raw
Sulfuric acid (H_2SO_4, density = 1.824 g/cm^3)	tons	0.444	C_reagent 1
Water (technical)	tons	2.800	C_reagent 2
Specific consumption of raw materials and energy resources to produce 0.400 tons of ITW-TA			
ITW	tons	0.462	C_raw
Electricity (calcination)	kWh	92.4	C_energy

Resource and energy efficiency of $MgSO_4 \cdot 7H_2O$ (seven-water magnesium sulfate)				
Description	UoM	Q-ty	Unit price, USD	Cost, USD
Revenues	-	-	-	$175.0
$MgSO_4 \cdot 7H_2O$	tons	1	$175	$175.0
Expenditures				$60.0

Continuation of the table

TVET	tons	0.462	$3	$1.4
FTP-TA	tons	0.400	$3	$1.2
Sulfuric acid (H_2SO_4, density=1.824 g/cm^3)	tons	0.444	$100	$44.4
Water (H_2O, technical)	tons	2.800	$1	$2.8
Electro Energy (firing)	kWh	92.4	$0.07	$6.0
Others	-	-	-	$4.2
Resource and energy efficiency				
Gross profit (margin)	-	-	-	$115.0

Table 2 – Specific consumption of raw materials and energy resources for production of 1 ton of $Mg(OH)_2$ and resource and energy efficiency

Specific consumption of raw materials and energy resources for production of 1 t of $Mg(OH)_2$			
Type of resource	UoM	Specific consumption	Designation
$MgSO_4 \cdot 7H_2O$	tons	4.245	C_raw
	tons	10.6	
NaOH (25% NaOH solution)	tons	1.380	C_reactive
	tons	5.560	
H_2O	tons	4.2	C_ reagent

Resource and energy efficiency of $Mg(OH)_2$ (magnesium hydroxide)				
Description	UoM	Q-ty	Unit price, USD	Cost
Revenues	-	-	-	$1445
$Mg(OH)_2$ (magnesium hydroxide)	tons	1	$1200	$1200
Na_2SO_4 (sodium sulphate)	tons	2.450	$100	$245
Expenditures	-	-	-	$950
$MgSO_4 \cdot 7H_2O$ (seven-water magnesium sulfate)	tons	4.245	$60	$255.8
NaOH (sodium hydroxide)	tons	1.380	$500	$690.0
Water (H_2O, technical)	tons	4.2	$1	$4.20
Resource and energy efficiency				
Gross profit (margin)	-	-	-	+$495.0

Table 3 – Specific consumption of raw materials and energy resources for production of 1 ton of MgO (magnesium oxide) and resource and energy efficiency

Specific consumption of raw materials and energy resources for production of 1 ton of MgO			
Type of resource	UoM	Specificconsumption	Designation
$Mg(OH)_2$ (magnesium hydroxide)	tons	1.470	C_raw
Electricity	kWh	731.3	C_energy

Resource and energy efficiency of MgO				
Description	UoM	Q-ty	Unit price, USD	Cost
Revenues	-	-	-	$5000
MgO (magnesium oxide)	tons	1	$5000	$5000

Continuation of the table

Expenditures	-	-	-	1444.3
$Mg(OH)_2$ (magnesium hydroxide)	tons	1.470	\$950	1397
Electricity	kWh	731.3	\$32.64	47.8
Resource and energy efficiency				
Gross profit (margin)	-	-	-	+\$3555.7

Comparative analysis of the revenues of each stage of the process of obtaining magnesium compounds in the series: magnesium sulfate solution ($MgSO_4$) → $Mg(OH)_2$ → MgO shows that the use of serpentinite (including technogenic) resources becomes more resource- and energy-efficient in the deep processing of technogenic waste, when the end product is magnesium oxide (MgO). This is an important factor motivating further scientific and technological research in this direction.

Conclusion

The use of serpentinite as a source of magnesium to obtain industrially important compounds – magnesium sulfate, hydroxide and oxide – is a promising direction that combines technological, economic and environmental sustainability. The most effective methods are acid leaching and application of thermoactivated serpentinite at the stages of neutralization and purification of sulfate solution, which provides obtaining of magnesium hydroxide and oxide with high quality characteristics. The development of these technologies allows to obtain in-demand inorganic magnesium compounds in various industries of Kazakhstan, because the volume of serpentinite technogenic raw materials allows to organize their production in any necessary quantity.

Acknowledgments

This study was funded by the Committee of Science and the Ministry of Education and Science of the Republic of Kazakhstan (Grant No. BR21882242).

Conflict of interest

All authors are aware of the article's content and declare no conflict of interest.

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