ENVIRONMENTALLY FRIENDLY AND RESOURCE-SAVING TECHNOLOGY FOR DISPOSAL OF DUSTY ASBESTOS-CONTAINING WASTES AND PRODUCTION OF MAGNESIUM SALTS

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ABSTRACT

This paper described a scheme for disposal of dusty asbestos-containing wastes (DAW), mainly containing serpentine minerals such as antigorite, chrysotile and lizardite with the grade of MgO and SiO_2 of 32-38 wt. %, respectively, is proposed in the work on the basis of conditions and modes of acid treatment of DAW determined experimentally. Conditions and modes of acid treatment of DAW are proposed in order to produce magnesium salts ($MgSO_4$, $MgCl_2$ и $Mg(NO_3)_2$) using sulfuric, hydrochloric and nitric acids to leach magnesium from DAW. It has been noted that the quality of produced magnesium salts is consistent with the currently valid quality standards, and a thick layer of polysilicic acid is formed on the surface of undissolved residues that changes composition and physical-and-chemical properties of surface layers of chrysotile-asbestos, including its adverse environmental impact. An environmentally friendly and resource-saving technology for disposal of industrial dusty asbestos-containing wastes produced from mining and beneficiation of chrysotile-asbestos has been proposed on the basis of the research results.

Keywords: chrysotile-asbestos, dusty asbestos-containing wastes (DAW), magnesium sulfate, magnesium chloride, magnesium nitrate, and environment.

INTRODUCTION

Dusty asbestos-containing wastes (DAW) are produced as a result of three-stage crushing (coarse, medium, small) during chrysotile-asbestos (CA) beneficiation processes. Cyclone filters and dust-collection systems with filter socks are used at crushing stages to catch/collect the main portion of DAW. They mainly contain serpentinite mineral ($Mg_3[Si_2O_5](OH)_4$) that can be found in three minerals: antigorite, chrysotile and lizardite. They normally contain 32-38% of MgO and 35-40% of SiO_2 with minor amounts of Fe, Al, Ca, Cr, and Ni. According to mining companies, average annual volume of produced DAW (containing up to 1.0% of chrysotile-asbestos) is over one thousand tonne.

DAW is the main chrysotile-asbestos mining and beneficiation waste that contains asbestos; thus, it is strictly monitored by local environmental departments in accordance with the Environmental Code.

Notwithstanding numerous researches on disposal of asbestos-containing wastes [1-3] and minerals, there are no commercially viable schemes for disposal of asbestos-containing industrial wastes and production of magnesium compounds. Therefore, this research was focused on development of methods for mitigation of DAW environmental impact and production of magnesium salts in compliance with valid quality standards.

EXPERIMENTAL PART

Dusty asbestos-containing wastes (DAW). A sample of DAW used in this research was provided by Kostanayskiye mineral JSC (Kazakhstan) being a mining company producing chrysotile-asbestos. Technically saying, it is called a multipurpose mineral powder being one of the most environmentally dangerous industrial waste produced as a result of chrysotile-asbestos mining. We received 10 kg of product for research purposes. 100-250 μ fraction was produced by screening. This fraction was chemically analyzed after being dried at 100ºC during 2 hours.

Chemicals: All chemicals used in the research were chemically pure. Concentrated sulfur and nitric acids were diluted as needed and used for research purposes and measurements when considering the interaction of DAW-acid (H_2SO_4, HCl, HNO_3) systems.

Methods of chemical analysis. Complete chemical elemental analyses of DAW and products of interaction of PAW and acids were carried out using screening electron microscope (JSM-6490LV, JEOL, Japan) complete with energy-disperse microanalyzer INCA Energy 350 systems.

X-ray analysis was carried out using X-ray diffraction meter DRON-3 by radiating copper cathode Cu K and nickel filter. X-ray diffraction patterns were made using powder method with ionizing recording of diffraction peaks at ambient temperature with the goniometer speed of two degrees a minute. ASTM card register was used to read X-ray reflections.

Sieve analysis was used to determined granulometric composition of initial wastes and siliceous residues after being acid treated [4].

Solution pH values were measured using pH-340 device.

The method used for studying the interaction in DAW-sulfuric acid system. The decomposition DAW

VOL. 16, NO. 9, MAY 2021 ISSN 1819-6608
ARPN Journal of Engineering and Applied Sciences

process and its studying were carried out according to the following method. A flask reactor c/w a reflux condenser was placed in a thermostat with glycerin as the working liquid (temperature fluctuation ±1°C). Before running experiment, a calculated amount of DAW and water was placed into the reactor, and then started the propeller stirrer [5]. Upon reaching the calculated temperature in the reactor that was a bit lower than the set temperature, a calculated amount of acid was added into the rotation zone of the stirrer. Once the initial components were loaded, the required leaching temperature was maintained for a certain period of time. Upon completion of the experiment, solid phase sedimentation rate, sediment moisture content, and degree of extraction (n, %) of magnesium ions from DAW were determined. Thereafter, the suspension was separated by vacuum filtration. The resulting solid residue enriched in silica was washed with water by displacement method until obtaining of neutral pH value. After washing, the solid residue was dried in a drying oven at 100-105°C. The solid precipitate was analyzed using a SEM (JSM-6490LV, JEOL, Japan) complete with the systems of INCA Energy 350 energy dispersive microanalyzer.

RESULTS AND DISCUSSIONS

Optimized Technology Modes for Production of Magnesium Salts from DAW

Production of magnesium chloride. Hydrochloric acid treatment. 400 g DAW is initially treated in a round-bottom flask with 0.4 L of water, then add 30-40 ml of HCl using a dropping funnel at intensive stirring; the reaction mixture is heated up to 80-85°C and maintained during 1.0-1,5 hours by thermostating. The liquid/solid ratio (W/S) is 3.14. The reaction mixture is a thick red-yellow suspension. 0.5 hours later, 2.0-2.5 L of water is added to the suspension; the pH value is 1.1-1.2. The required amount of neutralizing agent is then added while stirring until achievement of pH=8. In this case, the W/T is 3.25. After settling, the suspension is filtered. The characteristic of the filtrate (salt solution - magnesium chloride): a colorless transparent liquid, weight 2800-3105 g, density (d) is 1.14 g/cm^3, pH=7.8-8.0.

Table-1. Chemical composition of DAW.

Fraction size	Content of oxides, wt.%						
	SiO_2	MgO	Fe_2O_3	$A1_20_3$	CaO	K_2O	Other
DAW	37,77	44,05	5,55	0,44	0,10	-	16,46

Production of magnesium sulfate. Unlike other acids, when treated with sulfuric acid, the temperature of the reaction mixture rises up to 100°C, thus, there is no need for additional heating during the entire leaching process. The reaction mixture is a thick blue-gray suspension, W/S=3.41. Upon completion of neutralization and filtration, the solution is a colorless liquid, pH=8.0, concentration of magnesium sulfate is 175 g/l, density (d) is 1.12 g/cm^3, degree of magnesium leaching is 40-45%.

Production of magnesium nitrate. The treatment technique is the same as with hydrochloric and sulfuric acids. In contrast to them, treatment (synthesis) is carried out at a temperature of 45-55°C, W/T=3.2. Initially, the reaction mixture is a thick blue-green suspension. After adding water, a neutralizing reagent and filtration, the leach solution has a slightly yellowish color, pH=8.0, density (d) is 1.11 g/cm^3, and the degree of magnesium leaching is 40-45%.

Treatment of magnesium-containing solutions to produce magnesium and determination of their quality indicators.

Dry salts: $MgCl_2$, $MgSO_4$ and $Mg(NO_3)_2$ are produced by evaporation of leaching solutions (with appropriate acids) in a water bath (90°C) and drying in an oven (at 100°C).

The above-described method of DAW treatment with sulfuric, hydrochloric and nitric acids was used to produce experimental batches of magnesium salts: $MgSO_4$, $MgCl_2$ and $Mg(NO_3)_2$, 1.0 kg of each type of salt.

The processing of the results of the chemical analysis of the produced salts showed that the salts are practically free from any impurity elements (Fe, Ni, Cr, Si, Al, Mn, etc.), except for calcium (0.29 ÷ 0.62%):

- magnesium sulfate contains 99.29% of the basic substance in the form of $MgSO_4 \cdot 1.8H2O$ and 0.71% of $CaSO_4$.
- magnesium chloride contains 98.77% of the basic substance in the form of $MgCl^2 \cdot 3.5H2O$ and 1.23% of $CaCl^2$.
- magnesium nitrate contains 98.73% of the main substance in the form of $Mg(NO^3)^2 \cdot 1.3H2O$ and 1.27% of $Ca(NO^3)^2$.

The quality of salts is primarily determined by their purity, i.e. the content of impurity elements in their composition. Chemical analysis of magnesium salts $MgCl^2$, $MgSO^4$ and $Mg(NO^3)^2$ produced by treating DAW with appropriate acids: HCl, H^2SO^4 and HNO^3 under the experimentally established conditions and modes has showed that the produced salts, in terms of quality indicators and content of impurity elements, are practically consistent with the requirements of state standards (GOST) for magnesium salts.

According to the results of the survey carried out in the accredited laboratory of the testing center Sapa of the Auyezov SKSU, in terms of their physical and chemical properties, the batches of crystalline magnesium sulfate are consistent with requirements of the current

VOL. 16, NO. 9, MAY 2021 ISSN 1819-6608
ARPN Journal of Engineering and Applied Sciences

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regulatory documents (GOST 4523-77, TU 2141-016-32496445-00, TU 400069905.043-2012) (Table-2).

Table-2. Comparative characteristics of the pilot batch of magnesium sulfate.

Parameters	**TU 2141-016-32496445-00**	**TU 400069905.043-2012**	**Pilot batch**
Appearance	Crystalline powder	White crystalline powder	White crystalline powder
Weight percentage of magnesium sulfate ($MgSO_4$), %	Not less than 483	Not less than 48.3	48.3
Weight percentage of residue undissolved in water, %	Not more than 0.4	Not more than 0.4	0.35
Weight percentage of iron (Fe), %	Not more than 0.01	Not rated	0.0075
Weight percentage of chlorides (Cl), %	Not more than 0.2	Not rated	0.13
Weight percentage of sodium in terms of sodium oxide (Na_2O), %	Not more than 0.1	Not rated	0.08
Weight percentage of manganese (Mn), %	Not more than 0.01	Not rated	0.01
Weight percentage of calcium oxide (CaO), %	Not rated	Not rated	0.069
pH of 1% solution	Not rated	Not rated	6.5

The results of laboratory tests correlate with the data of laboratory studies and support the possibility of industrial implementation of the developed method for DAW treatment to produce magnesium sulfate for various purposes, including for development of efficient technologies for capturing carbon dioxide (CO_2) and formation of thermodynamically stable mineral $MgCO_3$.

Study of Changes in the Composition and Physical and Chemical Properties of the Surface of Acid Leaching Residue

Earlier, we determined [6] that the interaction of chrysotile-asbestos and sulfuric acid leads to a complete destruction of the morphology and structure of the surface layers of CA and the formation on the surface of enriched layer of polysilicic acid $SiO_2 \cdot nH_2O$. A relatively dense amorphous layer formed on the surface of CA leads to a change of its physical and chemical properties, including biological and congeneric activity, i.e. transforms them into an environmentally friendly state.

Scheme of DAW Disposal and Production of Magnesium Salts and Reduction of its Harmful Environment Impact

The recommended scheme for elimination of DAW environmental impact was developed on the basis of the results of studies of the processes of acid decomposition of DAW, as well as the quantitative and qualitative changes in the composition of the surface layers of chrysotile-asbestos during acid treatment.

DAW treatment according to the developed scheme (Figure-1) makes it possible to produce the following magnesium-containing products with stable competitive properties (in terms of quality and cost): magnesium sulfate, magnesium nitrate, and magnesium chloride corresponding to the requirements of GOSTs. Physical and chemical properties of DAW surface after acid treatment and washing with water, which is a predominantly amorphized layer consisting of $SiO_2 \cdot H_2O$, is not environmentally hazardous due to the complete change of the initial properties, including biological and carcinogenic activity.

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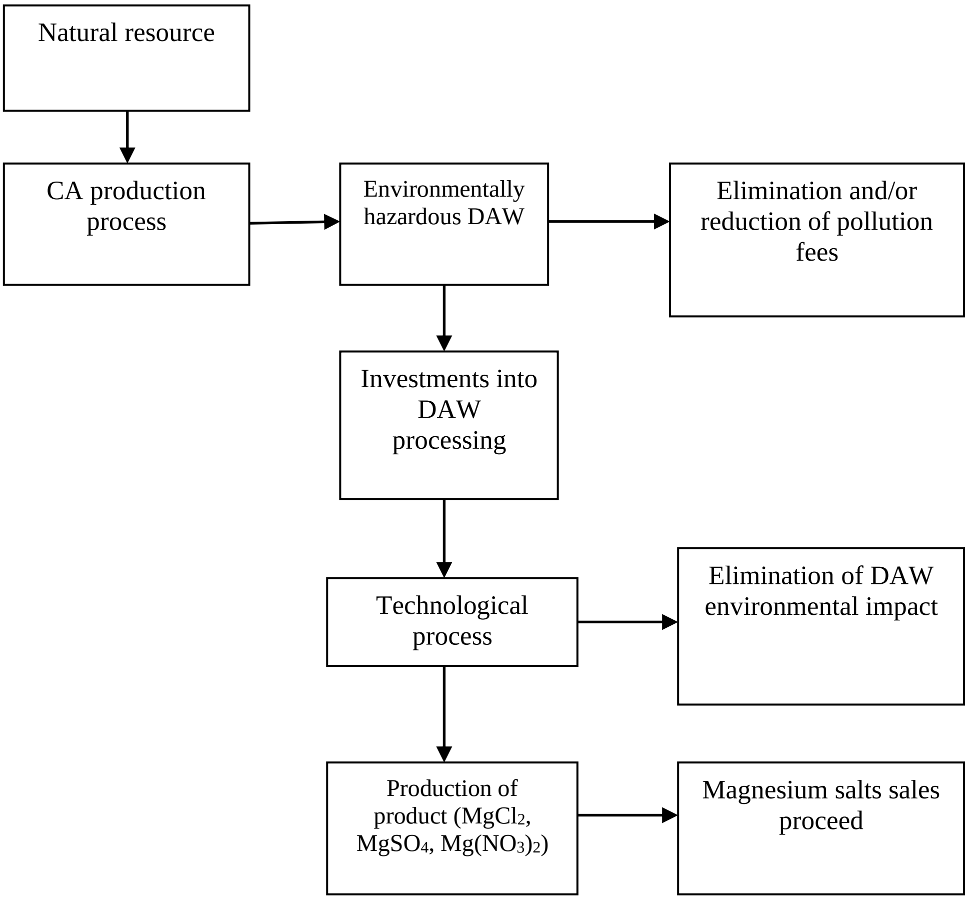

Figure-1. DAW disposal and production of magnesium salts and minimization of environmental impact.

Magnesium salts ($MgSO_4$, $Mg(NO_3)_2$ and $MgCl_2$) produced under the scheme of DAW treatment are initial substances used for production of industrial compounds - magnesium oxide, magnesium hydroxide, magnesium fertilizers and metallic magnesium. The consumption of these magnesium-containing compounds is an integral part of the chemical industry, for the production of which natural resources are used. Therefore, this DAW disposal scheme is the most promising for processing of this large-tonnage environmentally hazardous man-made waste containing 40-45% MgO in terms of environmental issues, i.e. environmentally resource-saving, which increases its investment attractiveness.

The proposed DAW disposal scheme can be used to produce magnesium salts ($MgSO_4$, $MgCl_2$, and $Mg(NO_3)_2$) with high qualitative parameters being consistent with the applicable quality standards and transform CA residue after acid leaching into an environmentally safe substance.

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