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АСПЕКТЫ ПРОБЛЕМ ПЕРЕРАБОТКИ ОТХОДОВ ХРИЗОТИЛ-АСБЕСТОВОЙ ОТРАСЛИ

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Аннотация: Проведен патентно-ананалитический обзор различных технологий в области применения серпентинитов и переработки отходов хризотил-асбестовой отрасли. В частности, рассмотрена научная литература, описывающая перспективы новых направлений применения серпентинитов и отходов хризотил-асбестовой отрасли в сельском хозяйстве, керамике, катализе, электрохимии, а также в качестве адсорбента, добавки в полимерах, минерала для улавливания углекислого газа. Рассмотрен ряд методов и способов использования и переработки серпентинитов и отходов хризотил-асбестовой отрасли в разные годы различными научными коллективами. В ходе проведенного анализа выявлены преимущества и недостатки имеющихся на современном этапе научных достижений по использованияю и переработке серпентинитов и техногенных отходов хризотил-асбестовой отрасли. Приведены и проанализированы технолого-экономические и экологические аспекты проблем утилизации отходов хризотил-асбестовой отрасли при помощи их кислотной переработки, решение которых могут открыть новые возможности в диверсификации основных производств хризотилдобывающих компаний стран Содружества Независимыз Государств (СНГ), и в первую очередь таких стран, как Российская Федерация и Республика Казахстан. По результатам исследований сформулировано заключение о поиске технологических решений применения и утилизации серпентинитов и отходов хризотил-асбестовой отрасли путем их комплексной переработки в качестве вторичного минерального сырья.

Ключевые слова: серпентинит, отходы хризотил-асбестовой отрасли, кислотные методы, переработка, окружающая среда, утилизация отходов, экологическая ситуация, комплексная переработка.

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Aspects of waste recycling problems in chrysotile asbestos industry

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Abstract: In this article, research has been conducted using a patent-analytical review of various technologies in the field of serpentinite application and recycling of chrysotile asbestos industry waste. In particular, the scientific literature describes the prospects for new applications of serpentinites and waste from the chrysotile asbestos industry in agriculture, ceramics, catalysis, electrochemistry, as well as as an adsorbent, an additive in polymers, and a mineral for carbon dioxide capture is considered. A number of methods and technologies of using and processing serpentinites and waste from the chrysotile asbestos industry in different years by various scientific teams are considered. The analysis reveals the advantages and disadvantages of the scientific achievements available today for the use and processing of serpentinites and man-made waste from the chrysotile asbestos industry. Based on the review, the techno-economic and environmental aspects of the problems connected with recycling of chrysotile asbestos industry waste through acid processing are presented and analyzed. The solution of these problems can open up new opportunities in diversifying production at chrysotile mining companies of the Commonwealth of Independent States (CIS), and primarily in such countries as the Russian Federation and the Republic of Kazakhstan. Based on the results of the research, a conclusion is formulated on the search for technological solutions for the use and recycling of serpentinites and waste from the chrysotile asbestos industry through their integrated processing as secondary mineral raw materials.

Key words: serpentinite, chrysotile asbestos industry waste, acidic methods, recycling, environment, waste disposal, environmental situation, integrated recycling

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Introduction

Extraction and processing of chrysotile has been a part of the mining industry for a long time. Over more than 100 years of exploitation of mineral deposits, large volumes of mining waste have been generated. Operating chrysotile asbestos quarries in Russia and in Kazakhstan use the open pit mining. A huge amount of waste is being accumulated, including residues of chrysotile fibers, which constitute an environmental danger. Many representatives of the business and scientific community recognize that waste from extraction and processing of chrysotile is a manmade raw material for the production of many magnesium and silicon-containing commercial-value products.

Meanwhile, little attention has been paid to their potential use in Russia and Kazakhstan. In this regard, the work draws attention to the results of the related studies conducted around the world. Currently, several areas of application of serpentinites are known, which are presented in the scientific and technical literature.

The purpose of this work is to analyze and summarize the research in the field of application of serpentinites, and to identify aspects of the problems of technology for processing waste from the chrysotile asbestos industry.

Results and discussion

Research on Serpentinite Applications

The use of serpentinite in ceramics is primarily due to its mechanical properties. The ceramic industry is distinguished by the possibility of recycling waste after processing of serpentinites, since large volumes of them are formed here. The favorable physicochemical characteristics of serpentinites as ceramic raw materials and the features of ceramic technology make this sector one of the best options for processing serpentine-containing waste [1].

In modern conditions, owing to chemical and mineralogical composition, industrial waste can act as a secondary mineral raw material for a number of industries [2—7].

Serpentinites have several interesting features for agricultural applications and can be used as a source of magnesium, silica and micronutrients such as Cr, Ni, etc. It can be used to increase the pH of typical acidic soils due to the presence of brucite [Mg] in its structure $(OH)_2$] layer [8—10].

One of the new and promising areas of application of serpentinites is their use as an aggregate in polymeric composites, which leads to a significant improvement in the mechanical and thermomechanical stability of the latter. It can also be used in high and low density polyethylene. The presence of serpentinite in an amount of 20—40 wt.% led to a significant increase in various mechanical properties of polyethylene. It is noteworthy that most of these studies use very fine tailings of serpentinite-bearing ore mining.

The peculiarities of the physicochemical properties of the surface layers and the molecular structural structure allow considering serpentinites as apromising material with high adsorption properties. The surface of the serpentinite mineral contains a large number of groups — OH of the water surface. These groups are responsible for properties associated with high adsorption capacity which is directly related to the surface charge. According to [11], in serpentinite minerals, charges can arise due to surface exposure, which occurs due to the rupture of hydrogen bonds connecting tetrahedral-octahedral units. These two open surfaces are formed by oxygen atoms and protons saturating the other negative surface. The acid–base characteristics of serpentinites in water can be attributed to acidic silicate layers and basic brucite-like layers.

The practical application of serpentinite as an adsorbent was studied in [12], where the adsorption of Cd^{2+} was tested on serpentinite, showing a value of 7.7 mg/g. After thermal activation, the adsorption capacity increased significantly, reaching 15.2 mg/g. Serpentinite can also be used to remove hexavalent chromium. This ion is commonly present in industrial wastewater generated from electroplating, pigmentation, chrome plating, leather tanning, and metal finishing. Hexavalent chromium has two main forms, i.e. chromate (CrO_4^{2-}) and dichromate ($Cr_2O_7^{2-}$); both forms are highly toxic in nature [13]. According to [14], chromium removal can be associated with two main effects: adsorption and reduction. A decrease of approximately 50% was observed at pH 5; at pH 3—7 adsorption was constant.

Serpentinite has also been proposed as a selective adsorbent for the removal of copper (Cu^{2+}) present in solution with other metals such as Ni^{2+}, Mn^{2+}, Zn^{2+} and Cd^{2+} [15].

Adsorption of organic dyes (Congo red and methylene blue) was studied in [13]. Removal of the cationic dye (methylene blue) was superior to the adsorption capacity of the anionic dye (Congo red). Adsorption of anionic dyes (Procyon red MX-58, Procyon orange and Ramozol black) was also assessed in [16]. Removal rates above 98% were achieved, however, relatively high doses of adsorbent were required to remove anionic dyes.

An interesting application of serpentinite [17] is the synthesis of electrodes. Electrochemistry of a carbon paste electrode containing serpentinite and graphite powder was investigated in an electrode cell to determine its electrocatalytic and sensing properties. The results show that the hydroxyls of the mineral structure are responsible for the main interactions between them and the solution. The electrode modified with serpentinite showed the best voltammetric response. This suggests that electron transfer occurs faster. The results of [18] also indicate that, due to its structure, surface groups, morphology and composition, serpentinite exhibits good electrocatalytic activity.

Another interesting and very promising direction is the study of using serpentinites to absorb anthropogenic carbon dioxide (CO_2). The absorption/trapping of CO_2 by serpentinites has been assessed by several researchers from around the world [18, 19]. Direct carbonization with the participation of serpentinite rocks is proposed [20, 21]. In this case, CO_2 dissolves in solution to form carbonic acid. This causes a decrease in pH in the medium, allowing the dissolution of magnesium silicate and subsequent carbonation [22].

To increase the efficiency of CO_2 sequestration by serpentinites, the work [22] proposed the use of magnesium silicate with another metal, for example Li^+, obtained as a result of the reaction between serpentinite and lithium hydroxide:

$$Mg_3SiO_5(OH)_4 + 8LiOH \rightarrow \\ \rightarrow 2Li_4SiO_4 + 3MgO + 6H_2O, \quad (1)$$

$$Li_4SiO_4 + 2MgO + 2CO_2 \rightarrow \\ \rightarrow Li_2MgSiO_4 + Li_2CO_3 + MgCO_3. \quad (2)$$

The introduction of lithium into serpentinite minerals leads to the formation of Li_4MgSiO_4 and MgO as the main phase. These phases are considered potential CO_2-scavenging agents due to their high thermal stability and selectivity. The experimental data obtained show that this material can capture approximately 25 wt% of CO_2 in a dynamic cycle and can reach up to 36% at 500 ºC. The stability of this material was also assessed during cycles of use and the results showed a loss of only 6% of its effectiveness.

Carbonation of natural silicate minerals is an interesting alternative to geological reservoirs for CO_2 storage, as silicate mineral resources have the highest capacity and longest CO_2 storage time of any known storage option. Since serpentinite contains a large amount of MgO (35–43%), it is considered as one of the main minerals for reducing atmospheric emissions from human activities. However, the natural carbonation of serpentinites and other silicate minerals is very slow, which means the carbonation reaction must be greatly accelerated to become a viable large-scale method for storing captured CO_2. The most effective processes proposed for carbonation involve leaching or dissolving serpentinite in liquid media and precipitating magnesium as carbonates or hydroxides for subsequent carbonation.

Serpentinite rock can act as a source of magnesium for the carbonate precipitation reaction. It is of interest as a feedstock for carbonation reactions because it leaches quickly in acidic conditions and is available on the Earth's surface. Therefore, among the strategies being developed for sequestering geological storage of CO_2 in carbonate minerals, serpentinite is of particular interest. In this case, the production of magnesium-rich solutions could be obtained from serpentinite-bearing tailings, providing an important resource for the development of technological strategies to reduce carbon emissions. This process represents a win-win scenario for the management of serpentinite and chrysotile-containing wastes in which the hazardous material is converted into a geologically stable mineral sink for CO_2.

Although these strategies are effective for rapid CO_2 sequestration in carbonate minerals, their technological and economic aspects are still poorly understood, and experimental data on these processes are scarce and sometimes contradictory. Therefore, at this stage of the research, it can be said that it is not yet practical for large-scale use due to the financial and energy resources required (for crushing, grinding or preheating, etc.). The search for alternative ways to use serpentinites in this direction continues.

There are also other applications of serpentinite in the literature [23, 24], for example, composites using chrysotile formed in the form of mesh threads and granules [25—29] to obtain various filter materials [30, 31]. Due to their amphiphilic nature, these composites have demonstrated a unique ability to form and break oil/water emulsions [30]. A special feature was the treatment of biodiesel wastewater to remove dispersed oil.

Serpentinites are also used as a precursor and catalyst support for the production of magnetic amphiphilic composites by chemical vapor deposition (CVD) [30—32].

The resulting composites showed very interesting properties, such as magnetic properties due to the nuclei of metallic iron inside the carbon structures and the carbon surface in combination with the hydrophilic surface of magnesium and silicon oxides. These composites demonstrated good results as magnetic adsorbents for the hormone ethinyl estradiol [31].

Another current trend is the use of serpentinite to produce silica for geopolymers. Geopolymers are new composite materials with a polymer structure that have very high strength. Despite the fact that cement is the most common material in construction, geopolymers can compete with it. They have technical advantages: they are more resistant to high temperatures and chemicals and do not contain hydration products that dissolve under the influence of acids and other aggressive substances.

The above areas of application show that serpentinites have very interesting physicochemical properties and great potential for technological applications.

Although serpentinite, as shown above, can be used in various fields, due to its physicochemical properties, one of the pressing issues of the chrysotile-asbestos industry is the processing of serpentinite and serpentinite-containing waste from chrysotile mining and enrichment.

Scientific, technological and economic aspects of problems of processing waste from chrysotile asbestos industry

Chrysotile enterprises in Russia and Kazakhstan are currently actively looking for solutions for resource conservation, integrated processing and diversification of their main production. The goal is obviously to search for scientifically, technically and economically sound technological schemes for extracting useful components from man-made waste of the chrysotile-asbestos industry in the form of goods and products with market values. Serpentinite rocks from the deposits of Russia (Kiembaevskoe) and Kazakhstan (Zhitikarinskoe) contain on average up to 43 wt.% MgO and up to 45 wt.% SiO_2, as well as other useful elements such as Fe, Ni, Co, Cr, etc. There are rocks containing nickel 0.8—1.0 wt.%. Useful components from the point of view of industrial importance in the composition of waste are considered to be MgO, SiO_2, and Fe, Ni, Cr and potential products that can be obtained from them: $MgSO_4$, $MgCl_2$, $Mg(NO_3)_2$, $Mg(OH)_2$, MgO, SiO_2 (amorphous), concentrates of Fe, Ni, Cr as well as other magnesium-silicon-containing composites and materials as a result of processing.

Despite the large number and versatility of research conducted in the world, including Russia and Kazakhstan, there are still no industrially acceptable technologies for practical application for obtaining the above useful products from waste from the chrysotile asbestos industry. There are many methods for processing serpentinites (including chrysotile production waste) [24—30], however, along with the advantages of each method, they also have disadvantages that limit their use. Basically, this applies to both technological and economic aspects. If we highlight and point out the general shortcomings of the above listed patents selected on the topic, then they can be summed up in the following phrases: 1) "the processing process is lengthy, labor-intensive with high material and energy costs"; 2) "the processing is inappropriate from the economic and environmental point of view"; 3) "all valuable components obtained by this method are of limited use due to their purity."

If serpentinite or serpentinite waste is used to produce magnesium and its compounds, acid leaching is usually the first stage of the entire hydrometallurgical process. When treated with an acid, a soluble magnesium salt is formed, which accumulates in the solution and is then separated from the insoluble residue, then the solution is cleared of by-product metal ions, which are leached along with the magnesium. The given stages of the entire hydrometallurgical leaching process have been studied in many works, where acids of various natures, mainly sulfuric acid [32] and hydrochloric acid [33—35], were used as leaching reagents. However, almost all works either superficially touch upon or lack information at all about the problems of acid leaching of serpentinite caused by the formation of polysilicic acid gels. This phenomenon is especially relevant when high concentrations of H_2SO_4 and HCl are used as a leaching reagent in order to more completely extract magnesium from serpentinites or serpentinite waste. The formation of colloidal silica particles, as known, is one of the main factors leading to complications in subsequent stages–accumulation of magnesium salt and its separation from the insoluble residue. Insufficient development of this stage of the process, which affects the manufacturability of subsequent stages, is found in almost all studies conducted. Apparently, the processes of this stage have not been sufficiently studied from the point of view of a fundamental study of the patterns of transformation of the silicate components of serpentinites into silica when using high concentration acids for leaching serpentinites. One way or another, the scientific and technological aspects of the problems of this stage still remain a task awaiting solution.

Regarding the technological and economic aspects of the problems of acid methods for waste processing, researchers, when determining the viability of the technologies being developed, often mention the high cost and volumes of reagents (NaOH, NH_4Cl, MgO) consumed in carrying out the processes of neutralization and purification of productive magnesium solutions from by-products, which ultimately hinders the achievement of a positive economic balance.

And, finally, it should be noted that despite the presence of large reserves of serpentinite-bearing ores in Russia and Kazakhstan, very little research has been carried to solve pressing issues of the use of serpentinites and processing of serpentinite-containing waste from chrysotile production.

Conclusions

Thus, based on the research conducted using a patent analytical review, the following conclusions and conclusions can be drawn:

• serpentinites and serpentinite-containing waste from mining and processing have very interesting physical and chemical properties, chemical composition, great potential for technological applications and in the production of various industrially important magnesium compounds, magnesium- and silicon-containing composites;

• the noted technological and economic aspects of the problems of recycling waste from the chrysotile asbestos industry show that research in this area should provide innovative approaches that minimize the use of external energy, technological risks and reagents, thereby helping to improve the technical and economic indicators of all stages;

• an integrated approach to the research covering the entire hydrometallurgical process, including aspects of leaching, purification and extraction, up to economic feasibility may have the prospect of its practical use in the disposal and processing of serpentinite-containing waste from mining and enrichment;

• solving the current aspects of technological and environmental problems discovered during the analysis of the research conducted around the world, in the future, may open up great opportunities in the diversification of the main production of chrysotile mining enterprises in the Russian Federation and in the Republic of Kazakhstan.

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