

ТЕХНОЛОГІЯ НЕОРГАНІЧНИХ РЕЧОВИН ТА СИЛІКАТНИХ МАТЕРІАЛІВ

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CURRENT TRENDS IN THE USE OF NATURAL AND SYNTHETIC WOLLASTONITE. REVIEW

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The crystalline phases of the CaO-SiO₂ system and polymorphic modifications of calcium metasilicate (wollastonite) are analysed. The physical, crystallographic characteristics and structural parameters of the wollastonite crystal lattice are presented. The features of the use of natural and synthetic wollastonite in various industries are considered. The role of wollastonite as a reinforcing material, which depends on the morphology of its crystals, is highlighted. The prospects of developing innovative technologies for the synthesis of wollastonite, in particular, by hydrothermal synthesis and high-temperature sintering, are substantiated.

Keywords: chemical technologies, wollastonite, tobermoryte, solid-phase sintering, hydrothermal synthesis, ceramic tiles, technical ceramics, composite materials.

Introduction

Natural and artificially synthesised wollastonite is one of the most important raw materials, widely used across various engineering and industrial sectors, particularly in the production of silicate and finishing materials [1–4]. This mineral is characterised by a unique combination of physical and chemical properties, such as high thermal resistance, strength, chemical inertness, and stability at elevated temperatures. These properties make it a valuable component in fields such as electrical and radio engineering, metallurgy, construction, and technical ceramics, as well as in dry building mixes, insulation materials, decorative coatings, and as a filler in the production of polymers, paper, paints, and more. Wollastonite enhances the mechanical properties of materials, including their flexural strength, impact resistance, and resilience to fluctuating temperatures and harsh environments.

Thanks to its environmental friendliness, wollastonite is gradually replacing traditional fillers such as talc and asbestos, which have several limitations, particularly regarding their impact on human health and the environment. Wollastonite is environmentally safe, as it contains no harmful

elements and can be recycled without negatively affecting ecosystems. In this regard, its use is crucial for the development of eco-friendly construction and technical products that meet modern safety and sustainability standards. Due to its versatile properties, wollastonite can undoubtedly become the foundation for new high-performance materials used in industries where a combination of high strength, thermal resistance, and environmental safety is essential.

The purpose of the work is to conduct a comprehensive analysis of the crystal modifications and properties of wollastonite, the main principles of its reinforcing and mineralising action in the ceramic matrix, technological aspects of its industrial production, and areas of application, based on literature sources.

Wollastonite: properties and applications

Theoretical and practical knowledge on the synthesis and application of materials based on refractory non-metallic compounds is grounded in a detailed qualitative and quantitative analysis of phase transformations in systems at various temperatures, an investigation of the nature of physical and chemical processes, and theoretical calculations of

material properties based on a given composition, among other factors.

The fundamental theoretical basis for the development of wollastonite-containing materials is the phase diagram of the CaO–SiO₂ binary system (Fig. 1). This system produces four chemical compounds: CaO·SiO₂ (SS), 3CaO·2SiO₂ (C₃S₂), 2CaO·SiO₂ (C₂S), and 3CaO·SiO₂ (C₃S).

According to the CaO–SiO₂ phase diagram, calcium metasilicate CaO·SiO₂ (CS) melts without

decomposition at a temperature of 1544 °C; it exists in two modifications:

- α-CS – pseudo-wollastonite – a high-temperature modification that melts without decomposition at 1544 °C, the structure of which is not precisely determined;
- β-CS – wollastonite – a low-temperature modification that reversibly transforms into α-CS at a temperature of 1125 °C.

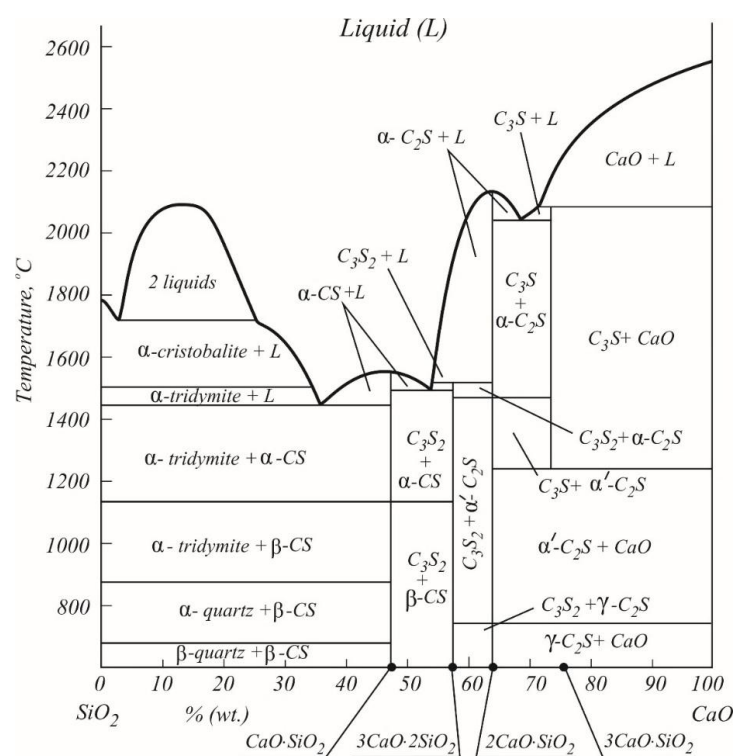


Fig. 1. Phase diagram of the CaO–SiO₂ system

Calcium metasilicate is characterised by distinct crystallographic individuality, forming tabular (β-CaSiO₃) or prismatic (α-CaSiO₃) crystals, which differ in terms of birefringence, extinction

angles, 2V, and other optical properties. The physical and crystallographic characteristics of the calcium metasilicate modifications are presented in Table 1.

Table 1

Physical and crystallographic characteristics of wollastonite CaSiO₃

Indicator	Polymorphic modification	
	low-temperature β	high-temperature α
1	2	3
Structural formula	Ca ₃ [Si ₃ O ₉] [‡]	Ca ₃ [Si ₃ O ₉] [‡]
Symmetry class	triclinic	triclinic
Space group	P1	–
Formula unit Z	6	8

Continuation Table 1

1	2	3
Unit cell parameters *		
– linear, nm		
<i>a</i>	0.794	0.690
<i>b</i>	0.732	1.178
<i>c</i>	0.707	1.965
– angular		
α	90°	90°
β	90°16'	90°48'
γ	–	90°
Crystal habit	tabular	Prismatic
Cleavage	perfect along (100), (102) and (001)	perfect along (100) and (001)
Melting temperature, °C	1125**	1544
Density, kg/m ³	2915	2905
Hardness on the Mohs scale	4.5–5	5
Coefficient of thermal linear expansion, K ⁻¹	6.5·10 ⁻⁶	11.8·10 ⁻⁶
Refractive index:		
<i>N_g</i>	1.634	1.654
<i>N_m</i>	1.632	1.611
<i>N_p</i>	1.620	1.610
2 <i>V</i>	39°	37°
Optical sign	(–)	(+)

* The crystal structure of wollastonite is shown in Fig. 2.

** The temperature of the polymorphic transformation of the β -form (wollastonite) into the α -form (pseudo-wollastonite).

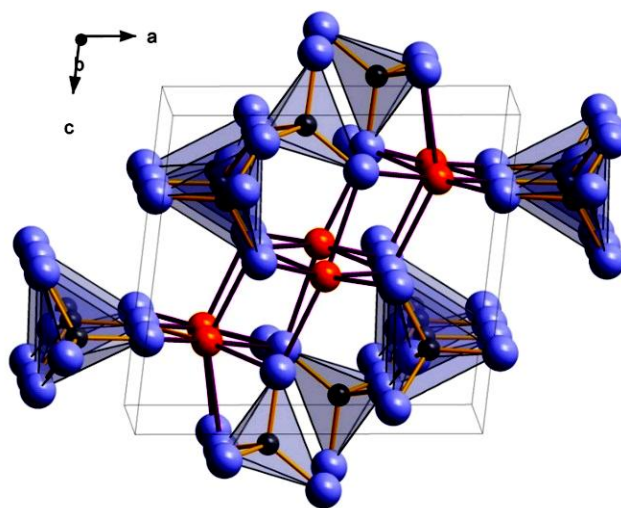
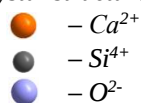


Fig. 2. Crystal structure of wollastonite [12]:



Natural wollastonite is characterised by a coarse crystalline needle-like fibrous structure with a length-to-diameter ratio ranging from 3 : 1

to 20 : 1, whereas synthetically produced wollastonite exhibits lower values for this parameter (Fig. 3).

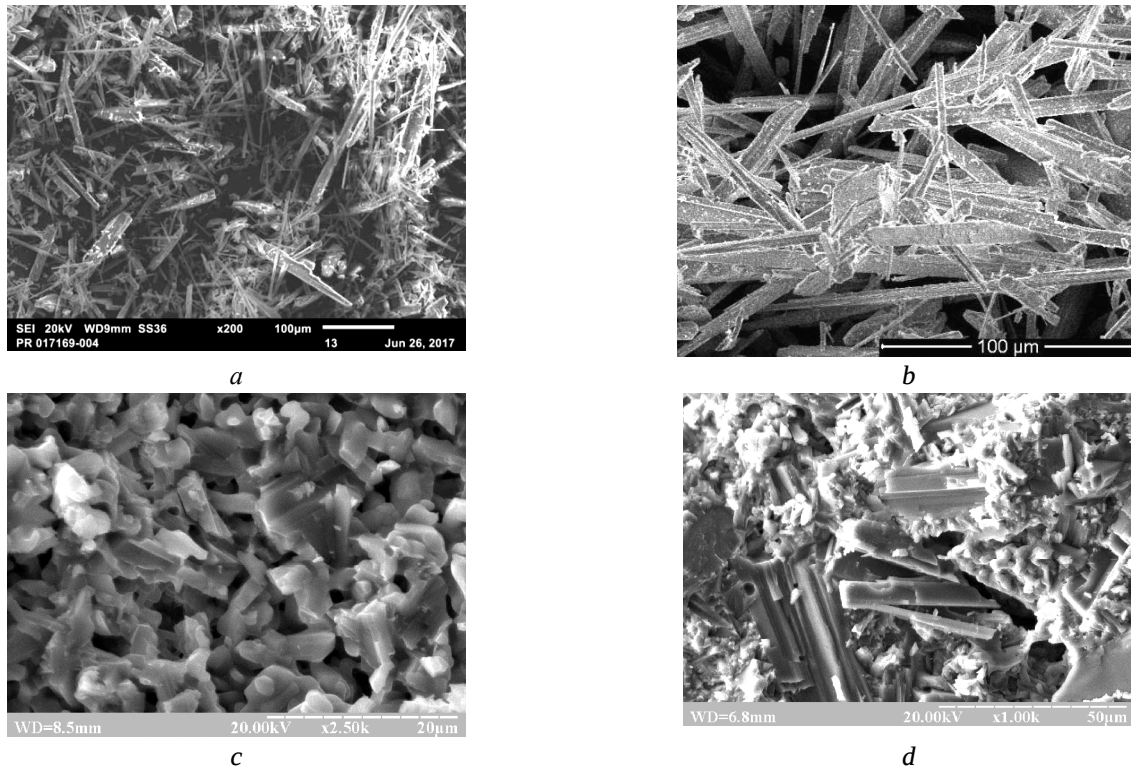


Fig. 3. Electron micrographs of wollastonite: *a, b* – natural [13, 14]; *c, d* – synthetically produced [15]

The rock extracted from the quarry contains between 25 % and 40 % wollastonite, whereas the enriched wollastonite concentrate consists of 80–85 % of this mineral. Currently, wollastonite deposits in Europe are largely depleted, and industrial reserves of this mineral are concentrated in countries such as the USA, China, India, Kazakhstan, and Uzbekistan.

Both natural and synthetically produced wollastonite are used in industry. Special attention is given to synthetic calcium metasilicate, which is currently obtained by firing a mixture of components containing CaO and SiO₂. Unlike the natural form, synthetic wollastonite has a fine-grained structure [15], is chemically purer, and contains a higher percentage of the CaSiO₃ mineral. The properties of synthetic wollastonite depend on the type of raw materials, the synthesis method, and the technological parameters of its production. The melting temperature of synthetically produced wollastonite ranges from 1350 to 1440 °C, which is 100–200 °C lower than that of the natural form (1540–1550 °C).

The range of applications for both natural and synthetic wollastonite is largely determined by its properties, as well as the nature of the transformations that occur during heating and cooling. The

content of wollastonite as a filler in ceramic compositions depends on its origin and properties and ranges from 15 % to 65 % [16–17].

In the technology of ceramic tile production, the incorporation of wollastonite into the composition of the material improves deformation resistance, bending strength, as well as thermal and frost resistance of the products. The effect of wollastonite on the physicomaterial properties of the products is due to its involvement in the sintering and phase formation processes of the ceramic material. During the firing of wollastonite-containing bodies at temperatures around 1000 °C and above, the formation of new crystalline phases is possible, including anorthite (CaO·Al₂O₃·2SiO₂) [18]. The density of anorthite (2.76 g/cm³) is somewhat lower than that of wollastonite (2.9 g/cm³), which leads to a reduction in the true density of the crystalline phases and an increase in their volume. This, in turn, reduces the shrinkage of ceramic bodies. At temperatures above 1050 °C, partial dissolution of fine-grained quartz and wollastonite occurs. Furthermore, the addition of wollastonite reduces the temperature range for the formation of mullite, an important crystalline phase of the ceramic material, and shifts its formation to lower temperatures.

The incorporation of natural or synthetic wollastonite into ceramic bodies significantly reduces the hygroscopic expansion of the material. Ceramics containing synthetic wollastonite exhibit somewhat lower values of hygroscopic expansion compared to ceramics containing the natural mineral [17]. This can be explained by the saturation of the glass phase with alkaline earth metal ions. The low hygroscopic expansion and high strength of the ceramic material, which contains synthetic wollastonite, contribute to a significant improvement in frost resistance, which is a crucial condition for the production of façade and wall ceramics.

Wollastonite is also used in the production of low-fire porcelain. The incorporation of 25 % wollastonite into the material composition allows for a reduction in firing temperature of such porcelain to 1150–1200 °C and shortens the dwell time at the maximum temperature compared to traditional fine ceramic products. In terms of physicomachanical properties, wollastonite-containing porcelain is not significantly inferior to conventional porcelain [19–21].

Wollastonite is widely used for the production of various types of glazes and coloured pigments. Wollastonite-containing glazes are characterised by good flow, gloss, fewer defects, and high whiteness [16].

Wollastonite-containing ceramics are characterised by high specific electrical resistance, which is an important property in the development of electronic equipment. Electrical insulating wollastonite-containing porcelain is used in electronic devices, the production of aviation ignition plugs, and more. The combination of lead silicate and wollastonite allows the creation of materials with ultra-high electrical resistance. The use of wollastonite in the production of electrical and radio ceramics significantly reduces the cost of products [22, 23].

The hardness and high natural whiteness of the wollastonite mineral make it an effective filler for paints and coatings [24]. Moreover, wollastonite imparts increased corrosion and abrasion resistance to paint coatings, as well as the ability to maintain their optical and colour characteristics under the influence of atmospheric factors.

At the current stage of materials science development, synthetic wollastonite, due to its high bioactivity and biocompatibility, is actively used in medical technologies for the regeneration and restoration of damaged human bone tissue [25–27].

The mechanism of the regenerative action of wollastonite is that the Ca^{2+} and Si^{4+} ions present in its structure actively participate in the formation of a hydroxyapatite layer, which facilitates the creation of a strong bond between the material and the bone tissue. This promotes the rapid regeneration of bone structures [28–29].

Overall, both natural and synthetic wollastonite are characterised by a range of valuable properties that positively influence the quality of products for various functional purposes, improving their performance and durability [30–32]. As a result, wollastonite is considered a multifunctional material and a promising industrial raw material for the Ukrainian market.

Reinforcing properties of wollastonite

The multifunctional applications of wollastonite are primarily due to its needle-like morphology, relatively low coefficient of thermal linear expansion, and high refractive index and whiteness. The needle-shaped crystals of wollastonite make it an ideal reinforcing filler for a wide variety of composite materials [33–34]. Reinforcing a matrix with needle-like crystalline phases enhances the product's resistance to thermal shock and increases its strength. This is because the fibrous filler amplifies the matrix's fracture resistance, promotes the formation of microcracks at the filler-matrix interface, and alters their propagation paths, thereby slowing the spread of cracks throughout the structure of the product.

The effectiveness of reinforcing a material with wollastonite fibers depends on the length-to-diameter ratio (l/d) of the crystals, the volume content, and the strength of the bond with the matrix [35]. An increase in the l/d ratio contributes to an improvement in all types of material strength. When reinforcing a dense ceramic matrix with needle-like wollastonite crystals, an optimal fiber volume content of approximately 20 % is considered ideal [36].

If the reinforcing fibers do not have a strong bond with the matrix, crack propagation occurs through a 'fiber pull-out' mechanism [37]. An example of a composite material with low fiber-matrix adhesion is asbestos cement. Due to the minimal interaction between the asbestos fibers and the matrix, they are mechanically pulled out during specimen fracture [33–34]. However, if the microstructure of the cement paste is modified with

needle-like wollastonite crystals, a significant number of hydrated new formations are localized on its surface, providing a high degree of bonding between the matrix and the reinforcing filler. In this case, the physico-chemical processes of new hydrate phase formation and increased adhesion between the filler and matrix occur through dissolution and crystallization in an aqueous environment.

In ceramic products, the interaction of wollastonite fibers with the matrix and the achievement of strong adhesion between them occur due to the rare-phase sintering of the material. This ensures a high degree of interaction between the introduced mineral and the base mass [38]. The amount of glass phase (8–15 %) depends on both the impurities present in the wollastonite mineral and the degree of interaction of the wollastonite particles with the aluminosilicate matrix of the ceramic. The nature and features of the microstructure of the reinforced matrix, as well as its mechanical properties, are determined by the chemical and mineralogical composition of the raw material, the amount of wollastonite additive, the dispersion of raw materials, the conditions and parameters of shaping the products, and the temperature-time firing regime, among other factors.

Overall, the nature and intensity of the reinforcing effect of wollastonite in the ceramic matrix structure are determined by:

- the content of wollastonite in the mass;
- the dispersion of the mineral (particle size and specific surface area);
- the ratio of l/d .

The reinforcing effect of the mineral in the ceramic composition is observed even with a relatively small content of wollastonite in the mass and gradually increases with the amount of additive. The optimal wollastonite content for various ceramic products, which ensures their high physical and mechanical properties, ranges from 15 to 65 % [16–17]. Due to their fibrous-needle-like morphology, the wollastonite crystals significantly improve the strength properties of the products, primarily bending strength, such that the bending strength of wollastonite-containing ceramics is only slightly lower than their compressive strength.

Another positive effect of wollastonite is that during firing at temperatures above 1000 °C, it acts as a flux due to the calcium oxide and participates in the rare-phase sintering of the ceramic matrix. At the same time, its fibrous structure enhances the matrix's resistance to deformation.

It is also important to highlight the positive reinforcing effect of wollastonite during the shaping and drying stages of the products. The addition of up to 50 % wollastonite to the mixture significantly reduces air shrinkage of the products due to the formation of a spatial framework.

Synthesis of synthetic wollastonite

Global ceramics manufacturers' practices have shown that the incorporation of both natural and synthetic wollastonite into the composition significantly improves the operational characteristics of the products and enhances their quality [39–41].

The traditional synthesis technology for artificial wollastonite is based on the solid-phase sintering of silica and calcium-containing components [42–43]. The focus is placed on intensifying the synthesis process through increasing the grinding fineness of the raw materials, replacing crystalline quartz with amorphous SiO_2 forms, adjusting the $\text{CaO} : \text{SiO}_2$ ratio in the mixture, and selecting optimal temperature-time parameters for firing. Calcium-containing components such as calcium carbonate or silica-limestone are recommended as the calcium source.

In ceramic production, synthetic wollastonite is obtained using silica-containing raw materials that include various modifications of SiO_2 . Amorphous forms of silica, such as diatomite, tripolite, opal, silicon dioxide, and others, exhibit a higher reactivity with calcium oxide compared to quartz or other crystalline forms of SiO_2 [44–45].

The most significant parameters in the synthesis process are firing temperature, the fineness of quartz grinding, and the duration of holding at the maximum temperature. The presence of iron oxide, magnesium oxide, and other compounds in the mixture has a minimal effect on the wollastonite synthesis process. Additionally, the presence of soda positively influences the synthesis of wollastonite.

At the current stage of solid-state sintering technology, to ensure a more complete synthesis of wollastonite, crystalline quartz is gradually being replaced by active amorphous forms of silica. This significantly improves the efficiency of the process and accelerates the interaction between silica and calcium oxide [46].

Due to the limited deposits of natural wollastonite worldwide, the technology for producing

synthetic calcium metasilicate by sintering a quartz component with calcium carbonate has become an important source of its production. However, a significant disadvantage of this method is the high energy consumption required for the high-temperature synthesis of the mineral. An alternative method, which involves lower energy costs, is the two-stage technology for the production of synthetic wollastonite [47], which includes the following stages:

- hydrothermal synthesis of calcium hydrosilicate with tobermorite structure from a mixture of silicon oxide and calcium hydroxide;
- sintering of tobermorite followed by the production of calcium metasilicate (wollastonite).

The main technological factors that determine the completeness of the physicochemical processes of formation and crystallization of tobermorite in the $\text{CaO-SiO}_2\text{-H}_2\text{O}$ system during the hydrothermal synthesis stage are:

- the degree of order (crystallinity) of the silicon oxide structure;
- the water-to-dry ratio of the initial raw material mixture before autoclaving;
- technological parameters of autoclaving (duration, excess pressure in the autoclave).

The structure of silicon dioxide has a significant influence on the kinetics and mechanisms of the tobermorite synthesis processes. Studies [17, 44–45] have shown that when using a mixture of silica with slaked lime based on crystalline quartz, even after 10 hours of hydrothermal treatment, maximum interaction between silica and lime does not occur, and the content of unreacted Ca(OH)_2 in the final product is approximately one-third (31 %) of the amount initially introduced into the reaction mixture.

The duration of calcium silicate synthesis depends on the technological parameters of the process and is mainly determined experimentally (the composition of solid phases is monitored using DTA and XRD). A significant factor that can greatly intensify the processes of calcium silicate formation during hydrothermal treatment and its subsequent transformation into wollastonite during firing is the addition of small amounts of mineralizers – compounds of boron and fluorine – to the raw mix.

The second stage of this technology involves the firing of the synthesized calcium hydrosilicate to β -wollastonite at temperatures of 900–1000 °C, which is 150–250 °C lower compared to the conventional solid-phase sintering process of crystalline silica with calcium carbonate. Given the

increasing demand for wollastonite across various industrial sectors and the need for energy- and resource-efficient technologies, the two-stage synthesis scheme for calcium silicate holds significant potential for further improvement and implementation in production.

Conclusions

Based on literature sources, the crystalline phases formed in the CaO-SiO_2 system have been analysed, and the main crystallographic and physical properties of calcium metasilicate (wollastonite) are presented. The applications of both natural and synthetic calcium metasilicate in various fields of engineering and industry are described. It has been established that the use of wollastonite in the ceramics industry significantly improves the operational characteristics of products by enabling the directed regulation of the ceramic material sintering processes during firing. The incorporation of this mineral into the raw material mixture reduces shrinkage during drying and firing, increases product strength, lowers the sintering temperature and the temperature for glass phase formation, and allows the production of ceramics with specified operational properties. The determining factor for the multifunctional application of wollastonite is its reinforcing ability, which depends on the morphological characteristics of the crystals (particularly the length-to-diameter ratio, l/d) as well as the nature of the interaction with the matrix.

Given the limited industrial deposits of natural wollastonite worldwide, the development of innovative technologies for its artificial production has become highly relevant. The traditional method for producing wollastonite involves high-temperature solid-phase sintering of a silica-lime mixture; however, a great deal of interest is focused on the two-stage hydrothermal synthesis technology of the lime-silica mixture, followed by firing of the intermediate product to recrystallise it into wollastonite. The promising multifunctional applications of this material in various industries highlight the need for the development of new effective methods and innovative technologies for synthesising artificial wollastonite that meet the requirements of modern production and environmental safety.

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СУЧАСНІ ТЕНДЕНЦІЇ В ЗАСТОСУВАННІ ПРИРОДНОГО ТА СИНТЕТИЧНОГО ВОЛАСТОНІТУ. ОГЛЯД

Проаналізовано кристалічні фази системи CaO-SiO_2 та поліморфні модифікації метасилікату кальцію (воластоніту). Наведено фізичні, кристалографічні характеристики та структурні параметри кристалічної ґратки воластоніту. Розглянуто особливості застосування природного і синтетичного воластоніту в різних галузях промисловості. Висвітлено роль воластоніту як армувального матеріалу, що залежить від морфології його кристалів. Обґрунтовано перспективність розроблення інноваційних технологій синтезу воластоніту, зокрема методами гідротермального синтезу та високотемпературного спікання.

Ключові слова: хімічні технології, воластоніт, тоберморит, твердофазове спікання, гідротермальний синтез, керамічні плитки, технічна кераміка, композиційні матеріали.