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Article

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INCREASING THE STRENGTH OF BUILDING CERAMICS MADE ON THE BASIS OF LOW-MELTING CLAYS

The object of the study is the physical and chemical processes of formation of the structure and properties, intensification of sintering of ceramic masses based on local raw materials (Krynichanska low-melting clay raw materials of the Kyiv region, Ukraine) by regulating the chemical and mineralogical composition and technological regimes. When choosing clay raw materials for specific ceramic technologies, it is necessary to be guided by a comprehensive assessment of the physicochemical properties of clay rock. These can be the granulometric and material composition, including the chemical and mineralogical composition of the clay and impurity components, the presence of amorphous material. The state of order in the structure of clay-forming minerals is also important, the knowledge of which makes it possible to determine the ways of regulating the basic technological properties of clay rock in order to bring them to the required level.

Among physical and mechanical parameters, mechanical strength is one of the main criteria for determining the suitability of raw materials for the production of building ceramics. The conducted studies have shown that with an increase in the amount of rotten stone additive introduced from 10 to 20 % of low-melting clay, the mechanical strength of ceramic samples in compression and bending increases. The introduction of rotten stone additive provides an increase in the coefficient of sensitivity to drying low-melting clay raw materials, which has a positive effect on the crack resistance of raw bricks when drying clay rock in order to bring them to the required level.

The use of silica materials in the composition of ceramic masses based on low-melting clays as an additive to improve the physical and mechanical characteristics of the finished product has shown its effectiveness. This can be explained by the fact that the nature of the interaction of silica additives, which was used as rotten stone, differs from the interaction of clay minerals present in ceramic raw materials with water. Since silica exists in rotten stone in the form of amorphous silica gel, it helps to improve the structure of clay, makes it monolithic, increasing mechanical strength. The impurities of low-melting oxides, which are part of the rotten stone, contribute to the formation of low-melting eutectic, reducing the refractoriness of amorphous silica and have a positive effect on the sintering process, forming a glass phase.

Keywords: *ceramic materials, clay raw materials, mechanical strength, silica materials, building ceramics, rotten stone additive.*

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1. Introduction

The rapid growth of capital construction needs to reduce costs at all stages. A significant net cost in housing construction falls on building materials. Reducing the cost of their production is the most important technological challenge. Almost all performance characteristics of ceramic building materials are largely determined by their mineral composition. At the same time, due to the shortage of high-quality raw materials, it is necessary to use cheap local raw materials to a large extent.

It is known that in order to improve the physical and mechanical characteristics of the finished product, 10–20 % quartz sand is added to the mass. Thanks to the use of modern high-quality equipment, the processes of preparing the ceramic mass, drying and firing are programmed and adjusted to the maximum [1]. This makes it possible to use siliceous materials, such as rotten stone or diatomite, as a non-deficient additive to low-melting clay. Their characteristic feature is active amorphous silicic acid and a finely dispersed structure, lightness, as well as environmental safety when used as natural raw materials. The nature of the interaction with water of amorphous silica, which is part of rotten stone and diatomite, differs from the interaction of clay minerals with water, which leads to an improvement in physical and mechanical properties. Therefore, their use as an additive in the production of building ceramics, which provides the possibility of using local raw materials for efficient and environmentally friendly production, is relevant.

2. The object of research and its technological audit

The object of research is the physical and chemical processes of formation of the structure and properties, intensification of sintering of ceramic masses based on local raw materials (Krynichanska low-melting clay raw materials of the Kyiv region, Ukraine) by regulating the chemical and mineralogical composition and technological regimes. To solve the tasks set, modern physicochemical and physico-technological methods for studying raw materials and masses based on them were used, which made it possible to evaluate the features of the structure formation of ceramic materials, as well as a set of ceramic-technological methods for studying the performance properties of finished products.

The subject of research is a comprehensive study of low-melting clay of the Kyiv region for the manufacture of building ceramics, which, after firing, form an insufficiently dense shard, which does not provide the necessary strength and frost resistance of the finished product.

The addition of silica-containing materials to the raw mass based on low-melting clay improves the physical and mechanical properties of finished products, contributing to the formation of the structure, which increases the strength and thermal insulation properties of the finished product.

3. The aim and objectives of research

The aim of research is to obtain a ceramic material based on local low-melting clay using rotten stone as a non-deficient natural additive, the strength of which will meet the requirements of building ceramics.

To achieve the aim, the following tasks must be completed:

1. To study the chemical and mineralogical compositions of the studied local low-melting clay.
2. To investigate the physical and mechanical properties of products based on local low-melting clay and masses with a silica-containing additive of rotten stone.
3. To work out the technological modes of firing ceramic masses.

4. Research of existing solutions to the problem

Siliceous rocks, which are dominated by opal, cristobalite and their varieties, are divided into two groups according to petrographic features. The first group includes diatomites, spongolites, radiolarites, and silicoflagellites; the second group includes flasks and rotten stones. Industrial value among them should be diatomites, flasks and rotten stones, to a lesser extent – spongoliths.

Rotten stone is the thinnest (0.005–0.020 mm) loosely bound grains of amorphous silica (heating) of volcanic origin (contains impurities of clay and iron oxides), which is used for the manufacture of thermal insulation products.

Diatomite is a loose porous sedimentary rock formed from the remains of microscopic shells of diatomite algae and radiolarian skeletons, containing 75–90 % SiO_2, up to 5 % $A1_2O_3$, up to 7 % Fe_2O_3 and up to 7 % alkali metal oxides. This is a finely dispersed, low-melting and low-strength material that has the same application as rotten stone [2].

The work [3] considers the location of deposits of silicate rocks, namely: rotten stone, diatomite, crystalline quartz, ground sand, etc. Their physico-chemical parameters, areas of application, cost and economic value in the production of ceramics are given. No attention is paid to the technological aspects of the use of siliceous rocks and changes in the properties of samples obtained with such additives.

It is known to use diatomite by its acid leaching in a hot solution (75 °C and 5 M HCl) for 1 h in order to increase the specific surface area at a constant SiO_2 content [4]. The leached material has an amorphous phase for a relatively short time, however, the effect of the percentage of this additive on the physicochemical properties of the material, including the strength of finished products, is not considered in the work. Powders based on diatomite have an increased porosity and specific surface: sintering at a temperature of 1300 °C achieves a porosity of about 48 %.

According to the study [5], as a result of adding oil-contaminated diatomite waste, galvanic sludge and glass production waste to the composition of red ceramic brick from 25 to 35 %, from 20 to 25 % and from 5 to 20 %, respectively, the properties of the samples change. The mixtures of the starting components were hydrated, pressed, dried, and burned out at various temperatures (950 °C, 1000 °C, 1050 °C, and 1100 °C) for 6 hours.

As a result of the study, high values of stability, low values of thermal expansion, water absorption, solubility and leaching of heavy metals were obtained. The high bending resistance (up to 22.5 MPa) of the new ceramics can be explained by the high-temperature chemical interaction of the components of the initial mixture and the synthesis of basic new amorphous vitreous structures. The paper does not indicate a direct dependence of the properties of ceramic products on an increase in the content of waste and with a change in the technological regime.

In the study [6], a change in the properties of the obtained ceramic materials and their phase composition was observed in accordance with an increase in the firing temperature. A lower content of the amorphous phase in the material makes it possible to obtain brighter products with a lower proportion of yellow. With increasing temperature, the total water absorption and total open porosity decrease, and the total linear shrinkage increases as a result of the progressive sintering process. The bending strength increases for materials composed of the studied clay. The addition of dolomite resulted in new pores in the material, resulting in an increase in flexural strength at lower firing temperatures and a decrease in flexural strength at higher firing temperatures. The influence of temperature has been investigated, but the influence of the quantitative addition of silicon-containing additives has not been considered.

In [7], the interdependence between the stages of firing cycles and structural parameters was experimentally determined. By applying the empirical equations of the obtained interdependencies, it is possible to control the technological process and obtain ceramics with different properties, although with the same composition of the shaping mixtures, but by varying the firing cycles (temperature increase, holding at the maximum temperature, temperature decrease). But the influence of the mineralogical composition on the parameters of the obtained samples has not been studied.

In [8], the effect of heating temperature and fly ash particles on the characteristics of porous ceramics with the addition of waste ash and diatomite (0–20 %) was studied. For a sintering process lasting 2 hours and pressure of

5 MPa, the sintering temperature of 1000–1270 °C is the initial one. This study does not highlight the effect of increasing the content of diatomaceous earth additive on the quality of the material, but indicates that the addition of coal fly ash to porous ceramics increases the compressive strength, thereby improving the quality of the products.

It is shown in [9] that the addition of low-grade diatomite and oyster shell improves the physical and mechanical properties of porous low-ceramic ceramics at a ratio of 70 wt. % and 30 % wt. % at a sintering temperature of 800 °C. Open porosity, bulk density and compressive strength were optimized to 50.20 %, 1.26 g/cm^3 and 18.8 MPa, respectively, but the dependence of the effect of temperature on physical and chemical properties was not studied.

In [10], the production of amorphous silica from waste or by-products of various industries and the agricultural sector (rice husk and its ash) is considered. It is concluded that such wastes are a potential alternative to conventional sources of silica (for example, quartz) for the production of value-added ceramics for practical use, but the technological regimes of firing and the required content of additives to obtain ceramics, which, in terms of properties, will meet the required standards, are not considered.

In [11], the optimal parameters for the production of diatomite ceramics with a visible porosity of 50–56 % are studied. At sintering temperatures ranging from 900 °C to 1000 °C, porous diatomaceous earth exhibits typical bimodal porosity, high imaginary porosity and high bending strength.

Rotten stone and diatomite are similar in chemical composition and physical properties. From a chemical point of view, they are earthy varieties of heating and are loose formations of natural silica hydrates, the composition of which can be expressed by the general formula $mSiO_2 \cdot nH_2O$. These rocks contain different amounts of water, ranging from 3 to 13 %, they also contain organic matter and impurities Na_2O, K_2O, CaO, MgO, Al_2O_3, Fe_2O_3. Their composition includes up to 20 % clay, which, with its uniform distribution, is a binding additive in the production of ceramic products. On the territory of Ukraine in Kharkiv, Vinnytsia, Lviv, Dnipropetrovsk regions there are reserves of these silicate materials.

Therefore, based on the analysis of literature data, it can be concluded that in order to select the optimal technological mode for the manufacture of building ceramics, it is necessary to investigate the effect of the silica-containing additive of rotten stone on the physical and mechanical properties of ceramic products based on local low-melting raw materials at optimal firing temperatures.

5. Methods of research

To solve the tasks set, modern physical and chemical methods were used – X-ray phase, differential thermal, chemical methods for studying raw materials and masses based on them, which made it possible to evaluate the features of the structure formation of ceramic materials. To determine the suitability of local low-melting clay for the manufacture of ceramic products, systematic studies were carried out to develop mass compositions, as well as the main characteristics of the manufactured products.

Let's use a plastic method for preparing the raw mixture and forming samples. When developing ceramic masses for the manufacture of building ceramics, it is necessary to take into account their sensitivity to drying, the change in the linear dimensions of the samples during firing, and the ultimate compressive and bending strength.

In order to study the effect of rotten stone additive on the technological properties of ceramic masses based on low-melting raw materials, the following charges for the production of building ceramics were studied, shown in Table 1.

Table 1

The compositions of the studied masses

Components	Content of components in mixtures, wt. %			
	1	2	3	4
Clay	100	90	85	80
Rotten stone	–	10	15	20

Ceramic samples for research were prepared by plastic molding, using a technological regime as close as possible to the process of manufacturing ceramic products for construction purposes. To do this, the clay was first crushed and then ground in fine grinding rollers and runners. Grinding rotten stone was carried out in ball mills, and then added to the original clay in the amount of 10, 15 and 20 %. The prepared components were sifted through a sieve, mixed and covered with water to normal molding moisture. After aging for a day, samples of 50×50×50 mm in size and beams of 60×15×10 mm in size were formed by plastic pressing at a moisture content of 18–22 %. Drying was carried out in an oven at 100 °C to constant weight. The firing was carried out in an electric furnace at 1000, 1050, 1100, and 1150 °C.

6. Research results

At the first stage of research, the chemical and mineralogical composition of low-melting clay was analyzed. The results of a chemical study of low-melting clay are given in Table 2.

As can be seen from Table 2, clay is composed of various oxides, free and chemically bound water, and organic impurities. According to the results of chemical analysis, according to the amount of Al_2O_3 of clay raw materials, it corresponds to the group of acidic clays. According to the amount of Fe_2O_3 and TiO_2, clay is a raw material with a low content of coloring oxides. It also has high carbonate content. The amount of free quartz is 32.92 %. The amount of water-soluble salts in the feedstock was 0.25 mg/eq/100 g of clay, which refers it to the group with a low content. The selected clay belongs to medium plastic. The main clay-forming mineral is hydromica.

Table 2

The chemical composition of Krynichanska clay of the Kyiv region

Deposit	Oxide content, %									
	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	CaO	Na_2O	K_2O	MgO	SO_3	l. o. i.
Krynichanska clay	61.71	11.51	5.01	0.57	8.76	2.19	1.24	1.57	–	7.44

Note: l. o. i. – loss on ignition

According to the results of differential thermal analysis, the heating curve of the studied raw material is characterized by:

– thermal effects related to the processes of release of adsorption water (150 °C) and interpacket water (220 °C);
– oxidation of ferrous iron with the formation of Fe_2O_3;
– removal of structural water from the clay mineral almost to the complete destruction of the lattice (560 °C);
– polymorphic transformation of quartz (580 °C);
– loss of hydroxyl water residues (700 °C);
– dissociation of calcium carbonates (820 °C).

X-ray phase analysis of the studied samples of minerals was carried out on a DRON-2.0 X-ray diffractometer with two Soller slits with filtered CuKα radiation with a nickel filter. The results indicate the presence in the clay:

– quartz (0.137; 0.145; 0.154; 0.166; 0.181; 0.198; 0.212; 0.223; 0.228; 0.245; 0.334, 0.425 nm);
– hydromicas (0.15; 0.256; 0.356; 0.444; 0.493; 1.00 nm);
– kaolinite (0.15; 0.239; 0.256; 0.356; 0.444; 0.714 nm);
– chlorite (0.15; 0.256; 0.356; 0.470; 1.38 nm);
– feldspar (0.284; 0.295; 0.318; 0.370; 0.383; 0.404 nm);
– calcite (0.144; 0.160; 0.162; 0.187; 0.191; 0.209; 0.249; 0.303; 0.386 nm).

Studies of the molding properties of ceramic masses based on low-melting clay have established that with the addition of rotten stone, the plasticity of the mass and molding moisture are reduced. But this did not lead to deterioration in the molding properties of the masses. Also, the addition of rotten stone to the studied clay contributed to a decrease in air shrinkage (Fig. 1).

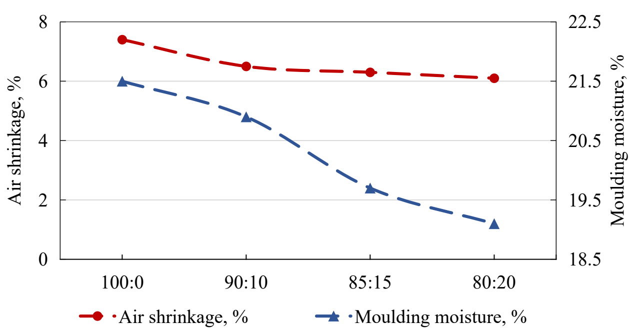

Fig. 1. Dependence of air shrinkage and molding moisture of the studied masses

The plasticity of different compositions of the ceramic mass decreased with an increase in the content of rotten stone in the samples. For clay, it was 14.6 %, for a sample with a ratio of 90:10 – 14.1; for sample 85:15 and for sample 80:20, respectively, 14.8 and 14.1 %. A suitable relationship was also observed for the drying sensitivity, which varied from 58 s for clay to 51 s (sample 90:10), 49 s (85:15) and 41 s (sample 80:20).

Among physical and mechanical parameters, mechanical strength is one of the main criteria for determining the suitability of raw materials for the production of building ceramics. Studies have shown that with an increase in the amount of rotten stone additive from 10 to 20 wt. % to low-melting clay, the mechanical strength of ceramic specimens in compression and bending increases.

Also, a visual inspection of the studied samples for the presence of cracks and distortions was carried out, since the quality of firing affects not only the mechanical strength, but also the water absorption and frost resistance of ceramics. The results of determining the ultimate compressive strength and bending of the studied masses are shown in Fig. 2, 3.

Mechanical strength also depends on the firing temperature. The maximum values of compressive and bending strength were achieved at a firing temperature of 1100 °C and the addition of rotten stone in the amount of 10–20 %. All fired at a temperature of 1000–1150 °C are characterized by a dense, strong ceramic shard. The color intensity does not depend on the amount of additive introduced into the mass, but is determined by the firing temperature. The color of the samples changed with increasing temperature from red-brown to dark brown.

The curves shown in the following figures are not an approximation, but only a visualization of the possible type of dependence.

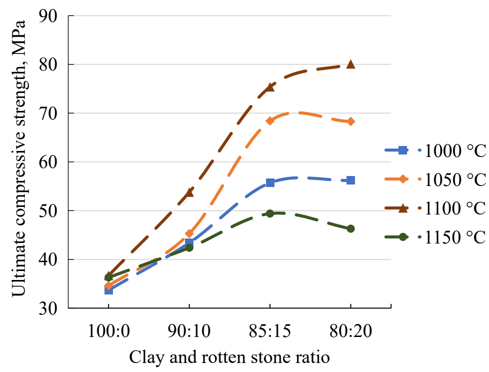

Fig. 2. Dependence of the ultimate compressive strength of the studied masses, scorched at different temperatures, on the ratio of clay and rotten stone additive

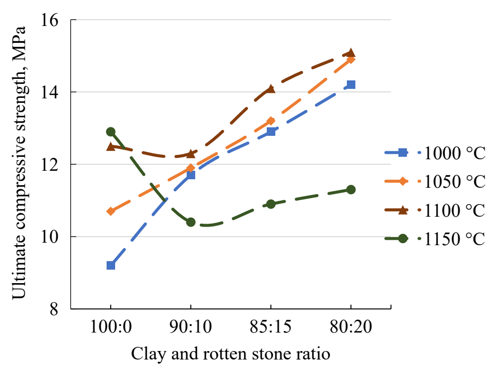

Fig. 3. Dependence of the ultimate compressive strength of the studied masses, scorched at different temperatures, on the ratio of clay and rotten stone additive

In rotten stone, silica exists in the form of an amorphous silicate gel, which helps to improve the structure of low-melting raw materials, makes it monolithic and increases mechanical strength. The presence of iron compounds contributes to the formation of low-melting eutectics, reduces the refractoriness of amorphous silica and has a positive effect on the sintering process, forming a glass phase. The sintering of ceramics occurs with the participation of the liquid phase and is accompanied by a complex of physicochemical processes of the interaction of amorphous silica with oxides that are part of the ceramic mass.

7. SWOT analysis of research results

Strengths. Among the strengths of this study, it should be determined that, due to the limitation of high-quality clay raw materials in many regions, the possibility of using low-melting clays for the production of building ceramics has been shown. Usually, raw materials are used, which include impurities that reduce the molding properties of the masses. To expand the use of low-melting clays, it is necessary to introduce impurities, one of which can be silica-containing materials that do not impair the molding of products.

The use of rotten stone as a raw material additive reduces the cost of production, since the source is local deposits and improves the physical and chemical properties of the resulting products.

This additive contributes to the formation of a monolithic structure due to the formation of a glass phase, which increases the mechanical strength and thermal insulation properties of the finished product. Air shrinkage and molding moisture are also reduced.

A very important factor is that this additive does not change the color of the sample, which is of great importance for facing and household ceramics.

Weaknesses. The weaknesses of this study are related to the fact that the developments are based on the need to use an elevated firing temperature, namely 1000 °C and higher, in order to achieve the appropriate strength of materials. And this increases energy costs.

Also, when rotten stone is added to the raw mass, the sensitivity to drying slightly decreases due to the appearance of thermal stresses.

Opportunities. In the future, it is very important to consider the best methods for preparing the rotten stone additive (purification, enrichment, etc.), which can have a significant impact on product quality and a higher price and greater demand.

It is also necessary to study and select the production parameters and the composition of raw materials in such a way as to reduce energy costs and use them rationally and energy-efficiently. This will reduce the cost and increase the environmental friendliness of production.

It is important to investigate the effect of components that reduce the firing temperature without compromising the strength of the material. The creation and selection of the optimal improved recipe is always a promising area of research in production, which will reduce energy costs, the amount of deficiency and improve quality.

This work may be of interest to foreign manufacturers due to the low cost of low-melting clay in the Kyiv region and in Ukraine as a whole.

Threats. The threat of this study is the presence of an analogue additive, which will provide the best physicochemical properties of the samples and lose rotten stone in cost. The world is also considering the use of diatomite, galvanic sludge, glass production waste, ash and oyster shell as part of the ceramic mass. Each of these additives can be promising when considered in the technology of ceramic production, but so far there is no obvious competitor that dominates in terms of properties.

8. Conclusions

1. When assessing the suitability of raw materials for the manufacture of a particular type of building materials, the chemical and mineralogical compositions of the studied local low-melting clay were studied. The selected clay belongs to medium plastic. The main clay-forming mineral is hydromica.

2. The conducted studies have shown that the addition of silica-containing materials, the deposits of which are located in many regions of Ukraine, can be used for the production of building ceramics (bricks). A slight decrease in molding moisture and plasticity did not adversely affect the molding properties of the ceramic mass.

3. The introduction of an admixture of rotten stone up to 20 wt. % made it possible to ensure maximum strength in compression and bending, as well as to expand the area of using local raw materials for the production of ceramic products.

The maximum compressive and bending strength parameters for the proposed recipes were achieved at a firing temperature of 1100 °C and are characterized by a dense, durable ceramic shard. The color intensity does not depend on the amount of rotten stone addition to the ceramic mass, but is determined by the firing temperature. The color of the samples changed with increasing temperature.

Conflict of interests

The authors declare that there is no conflict of interest regarding this study, including financial, personal nature, authorship or other nature that could affect the research and its results presented in this article.

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Data availability

The manuscript has no associated data.

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ANALYSIS OF METAMORPHISM AND TENDENCY OF COAL SEAMS AND THEIR HAZARDOUS PROPERTIES

The object of this study is coals of different stages of metamorphism. Currently, a situation has arisen when indicators developed to establish the consumer qualities of coal are used to predict the manifestation of hazardous properties of mine seams during mining operations. The need to consider the fuel for its working condition is due to different end goals between the establishment of consumer qualities of coal and the manifestation of the hazardous properties of mine seams. The condition and quality of coal after its preparation for use is significantly different from the condition in the mining area. Appropriate sample preparation changes the physical and chemical properties of coals, which determine the manifestation of the hazardous properties of mine layers during mining. To eliminate such a discrepancy, the quality indicators of coals were recalculated for their working condition, taking into account the yield of ash and moisture content for the mined mine seams, followed by an analysis of changes in the correlations be-tween the indicators. The indicators of the manifestation of the hazardous proper-ties of mine layers are borrowed from the characteristics of the quality of the fuel, reduced to a dry, ash-free state. Using these indicators, additional errors are introduced in advance into the accuracy of the prediction of the manifestation of hazardous properties during mining operations. The values of the indicators of the organic (combustible) part of the fuel serve as general reliable characteristics of its quality for the entire set of mine seams, but they cannot be used to predict the hazardous properties of a particular mine seam due to a decrease in the accuracy of their determination due to the unpredictable content of mineral impurities and moisture. The initial experimental data, which have been accumulated over several decades based on the experience of using coal for industrial purposes, are analyzed. On the basis of the conducted researches the peculiarities of the choice of indicators of metamorphic transformations of coal, which are used in parallel respectively to establish the quality of fuel and forecast the dangerous properties of coal seams, have been established. The discrepancies between the indicators of the degree of metamorphism used in the current regulatory framework for the safe conduct of mining operations, the state of fuel during mining operations in underground conditions.

Keywords: *physico-chemical properties of coal, hazardous properties of mine seams, coal quality, ash yield, moisture content.*

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1. Introduction

Coal fires pose great threats to valuable energy resources [1], the environment [2], and human health and safety [3]. They occur in numerous countries in the world. Not only do coal seam fires [4, 5] harm the environment and human health by burning uncontrolled, they also deplete non-renewable resources. Estimates of coal lost