



Thermal Decomposition of Asbestos Fiber from Asbestos Cement Wastes

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1. Introduction

Asbestos was widely used as a result of the industrial revolution of the last 100 years. Most of it was used for the production of asbestos cement products (Maciołek et al. 2012). Chrysotile asbestos constitutes from 90% to 95% of all asbestos used all over the world (Frank & Joshi 2014). It was added about 15% by weight to the total weight of cement-asbestos slates (Kusiorowski et al. 2014). The theoretical composition of chrysotile is: 13% H₂O, 33.65% MgO, 43.55% SiO₂ (Bolewski & Manecki 1993). Despite the fact that asbestos has been widely used in our everyday life, studies have shown for many years that it is not indifferent to human health. The greatest risk is release of fibers from products containing asbestos, as they penetrate the respiratory system, causing various diseases. Asbestosis, lung cancer, larynx, ovaries, mesothelioma (Więcek 2004) are some of the diseases that are caused by asbestosis. The World Health Organization (WHO) and the Agency for Research on Cancer (IARC) recommend phasing out asbestos due to its carcinogenic effects. It is estimated that occupational exposure to asbestos causes an estimated 107,000 deaths of people worldwide each year (Takahashi & Landigran 2016). As a result, a ban on the use, production and marketing of asbestos products has been introduced or is in the implementation process in many countries. Since 2005, the ban on asbestos has been in force in all EU countries (Commission Directive 1999/77/EC). Asbestos is banned in 60 countries around the world. Unfortunately, there are still countries where asbestos products are on the market. According to data published by the United States Geological Survey (USGS), asbestos production worldwide in 2015 was approximately 1.4 million Mg. The largest percentages were Russia, China, Brazil and Kazakhstan. In addition, asbestos products are still sold or used in many

countries, including India, Indonesia, Bangladesh, Kyrgyzstan, Colombia (Kazan-Allen 2017). Currently, India is the world's largest importer of asbestos (Minerals.usgs.gov 2018).

Due to the harmfulness of asbestos, it is important to control its disposal and use in various products. Asbestos products should also be safely disposed, and the waste containing asbestos should be neutralized.

Currently, the dominant method of managing asbestos waste in the world is its storage or the use of solidification, which involves covering asbestos waste with a protective layer. These methods do not lead to the complete neutralization of hazardous fibers contained in asbestos waste, which can still be released into the environment. The European Parliament in its resolution on asbestos of 14 March 2013. (2012/2065(INI)) points out that asbestos is the preferred method of inertising when disposing of asbestos rather than storing it. Therefore, it is very important to look for and study technologies that would permanently eliminate the problem of fibrous structure of the asbestos. There are several dozen patented ways of disposing asbestos. These processes can also be divided into two groups: thermal, chemical and mechanochemical treatment.

Among thermal methods, the so-called vitrification is used at temperatures above 1000°C, where in various furnaces an inert silica material without fibers is obtained. Then, thermal methods based on the melting process, here at temperatures of 1350-1550°C, serpentine fibers are converted into silicate minerals and glass. Additionally, e.g. kaolin clay can be used in these processes in order to lower the melting point and obtain construction materials after the process (Belardi et al. 1998, Paolini et al. 2018, Poniatowska 2008). Attempts have been made in Germany and Italy to dispose of asbestos waste using a cement kiln. As a result of the process, glassy, amorphous clinker balls were obtained, and no harmful chrysotile asbestos fibers were found (Ambrosius et al. 1996, Italcementi 1992, Paolini et al. 2018). Thermal disposal of asbestos-containing waste can also be carried out in plasma-based furnaces. As a result of neutralization in the plasma oven, a black, amorphous mass is formed, similar to glass, which can be reused in road construction and soil improvement (Klimas 1998, Paolini et al. 2018). In Poland, a technology using microwaves to neutralize asbestos has been developed. This process consists in heating the asbestos waste, which was previously crushed in a hermetic crusher, and then soaking it with additives, which improve wave absorption and reduce the temperature of the process up to 900-1000°C. Asbestos fibers are destroyed during processing (Paolini et al. 2018, Pawluk 2010).

Chemical methods use chemical reactions to convert asbestos into harmless compounds. Among others, strong acids, alkaline solutions, reducing factors such as metals in the elementary state are used. An interesting solution

here is the technology using whey from the dairy industry, at high temperature and pressure, in the presence of a chelating agent, such as oxalic acid, lactic acid (Balducci et al. 2012, Paolini et al. 2018).

In case of mechanochemical methods, fibers are destroyed by mechanical grinding. There are used various mills in which crystalline networks and molecular bonds present in asbestos are destroyed. High energy milling is also based on this process (Deng et al. 2009, Paolini et al. 2018).

On the basis of available literature, it can be concluded that only methods affecting the transformation of asbestos fibers, e.g. chemical, thermal, cause that the waste produced as a result of their use is processed and inert for health. Therefore, these methods should be regarded as preferable for the disposal of asbestos waste.

2. Materials and methods

The aim of this paper was to evaluate the influence of temperature and time on asbestos decomposition and determine if the asbestos fiber structure is damaged at high temperatures.

The following test methods were used to achieve the study objective:

- Thermal analysis using a high-sensitivity balance, a furnace for heating the sample and a system for dosing the sample washed gases (derivatograph);
- Thermal treatment of waste in the furnace with electronically programmable temperature regulator;
- Observation of samples after calcination with the use of scanning electron microscope with EDS probe (Energy Dispersive Spectrometry) for analysis of qualitative composition of materials;
- Analysis of phase composition on X-ray diffractometer.

Tested samples of asbestos cement waste with a weight of 510 mg was heated at a rate of 7.5°C/min, from 20 to 1400°C for about 190 minutes in an air atmosphere using a 1500 D derivatograph, MOM Budapest.

Thermal treatment of asbestos cement waste was carried out by calcination in a Nabertherm furnace with electronically programmable temperature regulator, increasing the temperature at 10°C/min to 1000-1500°C, for 30 and 120 minutes.

In order to identify asbestos fibers in asbestos cement waste, a scanning electron microscope LEO type 1430 with EDS microanalyzer by Oxford Analytical type ISIS 300 was used to analyze the chemical composition in the micro-area.

Phase analysis was conducted on Philips X'Pert PW 3020 X-ray diffractometer for samples after calcination at 1250 and 1400°C for 120 minutes.

3. Research results and discussion

Figure 1 shows thermal analysis of cement-asbestos slates samples, DTA curve (thermal differential analysis) and DTG/TG curves (thermogravimetry) were obtained. Three effects correlated with specimen weight loss are visible in the obtained record on DTG curve. The first effect at a temperature of 120-160°C was related to the loss of adsorbed water and decomposition of gypsum being a slight admixture of cement slurry. The second maximum at the temperature of about 480-520°C is related to the dehydroxylation processes of chrysotile and hydrated silicates and aluminosilicates of calcium. The third maximum at the temperature of about 820°C is the effect of both the final dehydroxylation phases of the components of cement slurry and chrysotile (Paolini et al. 2018). The record of TG curve indicates a loss in mass of the sample equal to 33% in the range up to 500°C and 58% up to 820°C. It can be assumed that up to 820°C the main decrease in mass of the whole sample occurs, because the subsequent changes are already very small, there is a 5% decrease in mass. A total loss in mass was 61%. Therefore, the loss in mass of sample is related to water removal processes associated with Portland cement components. The image of changes in TG and DTG curves is also reflected in DTA curve. Effects on DTA curve are endothermic peaks associated with weight loss. At about 120-160°C this is associated with decomposition of the gypsum structure ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) which is an additive (retardant bond), a cement from which asbestos cement was made, leading to the formation of anhydrite (CaSO_4). The endothermic effect at a temperature of about 480-520°C results from chrysotile structure breakdown and initial phases of calcium silicates crystallization. The endothermic effect with a maximum of about 820°C results from calcite decomposition (Kusiorowski et al. 2015). At about 1320°C another endothermic effect is recorded, related to decomposition of calcium carbonates and polymorphic transformation of dicalcium silicate (belite) of γ to β (Bielankin et al. 1957).

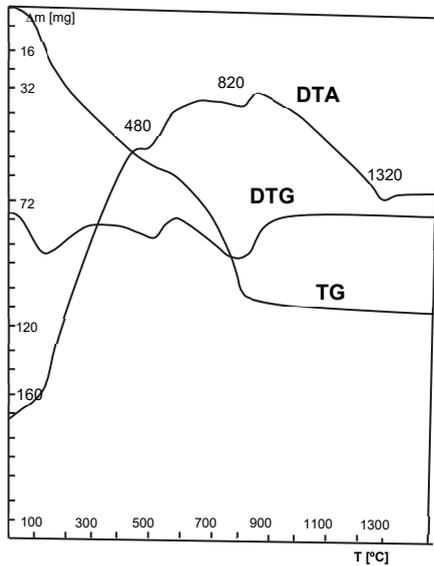


Fig. 1. DTA, DTG, TG curves of cement-asbestos slates after calcination at 1400°C for 190 minutes (Poniatowska 2008)

Tests conducted on a scanning electron microscope for raw samples show characteristic beams of chrysotile asbestos fibers. Cement minerals inlay asbestos fibers as a result of mixing with cement mass (Fig. 2a).

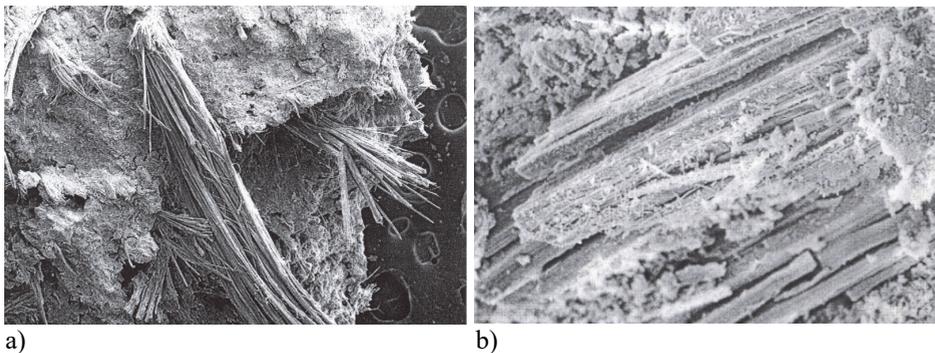


Fig. 2. SEM images of the sample of cement-asbestos slates a) and sample of cement-asbestos slates, in temperature of 1000°C for 30 minutes b) (Poniatowska 2008)

In case of samples after calcination at 1000°C, for 30 minutes (Fig. 2b), the first changes of asbestos in cement-asbestos slates are visible. Numerous packets and individual asbestos fibers are visible. Asbestos melting is at an early stages, and the packets are still prone to split into individual fibers.

Figures of samples after calcination at 1250°C for 120 minutes show the progress of calcination process in a form of increasing size of neogenic silicates contained in the cement binder, as well as their recrystallization along the asbestos fibers, which leads to their agglomeration and chemical transformation. Calcinated packet of chrysotile asbestos fibers creates characteristic rosettes. The number of visible fibrous packets in relation to samples after calcination at lower temperatures significantly decreases with the use of scanning electron microscope (Fig. 3a), and in the visible asbestos fibers are wrapped by calcium-magnesium silicates and the change of structure to granular.

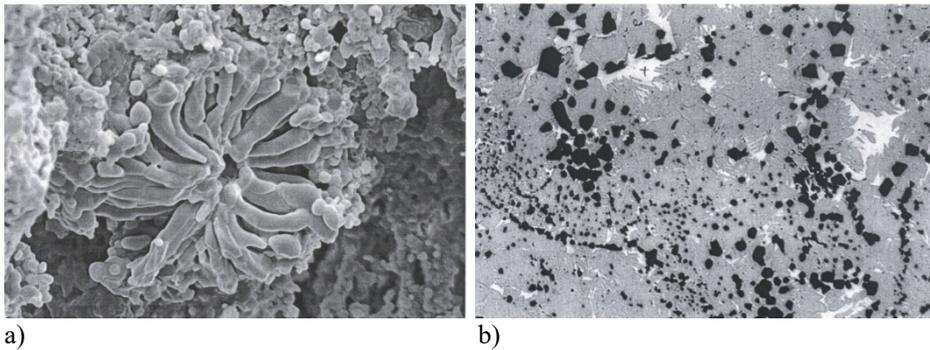


Fig. 3. SEM images of sample of cement-asbestos slates in temperature of: a) 1250°C for 120 minutes, b) 1400°C for 120 minutes (Poniatowska 2008)

As a result of calcination process carried out for a longer period, an increase in the dimensions of crystallites of all components is obtained (Fig. 3b). Asbestos packets are completely transformed and converted into calcium silicates and magnesium oxide (periclase). Figure shows total calcination of asbestos in the sample. Black fields correspond to magnesium oxides, while light poles and needles are silicates and calcium aluminosilicates.

This is also confirmed by X-ray phase analysis (Fig. 4a), where in case of the sample after calcination at 1250°C for 2 hours, the presence of larnite and periclase as the main components, small amounts of wollastonite and trace amounts of chrysotile was observed. This type of composition indicates a significant sample transformation. As a result of dehydration of components, the tobermorite diminishes, a part of which is transformed into larnite (Kusiorowski et al.

2015). Larnite is also produced as a result of the synthesis reaction of calcium oxide and silica – components resulting from decomposition of calcite, as well as antigorite and chrysotile. Periclase is produced during decomposition of antigorite and chrysotile. Due to the excess of calcium over magnesium at 1250°C, the formation of calcium silicates will be the preferred process, and therefore magnesium oxide is present in the sample as a periclase.

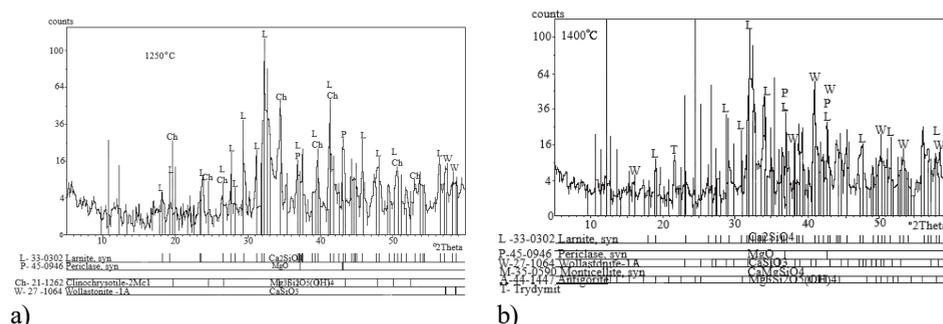


Fig. 4. X-ray diffraction patterns of cement-asbestos slates after calcination at: a) 1250°C for 120 minutes, b) 1400°C for 120 minutes (Poniatowska 2008)

In case of sample after calcination at 1400°C (Fig. 4b), larnite, periclase is dominant, wollastonite is likely, and monticellite and tridymite, antigorite and chrysotile were not found. The presence of tridymite should be associated with a far-reaching decomposition of antigorite and chrysotile. Monticellite is the effect of synthesis of calcium silicates with magnesium oxides. This type of composition confirms the process of asbestos disintegration from the serpentine group (antigorite and chrysotile).

Fig. 5a for samples after calcination at 1500°C for 30 minutes shows that the original structure of sample is transformed, barren silicates and calcium aluminates are formed and as a result of decomposition of chrysotile asbestos, magnesium oxides and di-calcium silicate are formed.

Heating the sample at 1500°C for 2 hours completely eliminates asbestos from cement-asbestos slates (Fig. 5b). The process of chemical transformation due to high temperatures led to the formation of coarse-crystalline calcium silicates, while the magnesium contained in chrysotile asbestos was transformed into an oxide phase in the form of periclase.

Three types of analytical spectra were obtained for this sample during the composition studies in the micro-area: for periclase (Fig. 6b), i.e. magnesium oxide, calcium silicates (Fig. 6c) and calcium-iron aluminates (Fig. 6a), formed as a result of calcination of asbestos cement components.

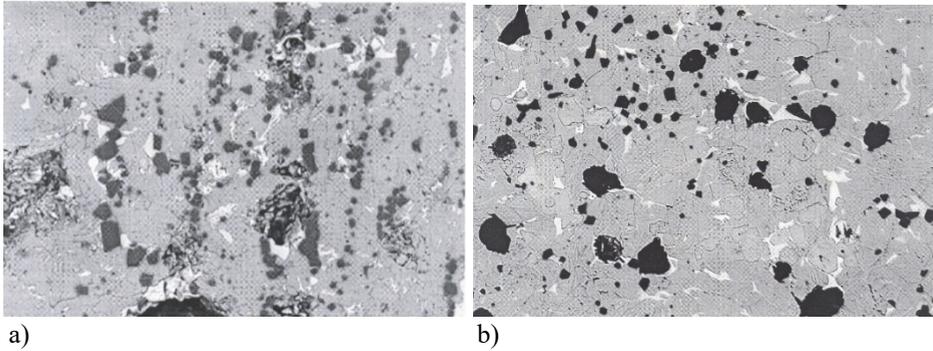


Fig. 5. SEM images of sample of cement-asbestos slates in temperature of: a) 1500°C for 30 minutes, b) 1500°C for 120 minutes (Poniatowska 2008)

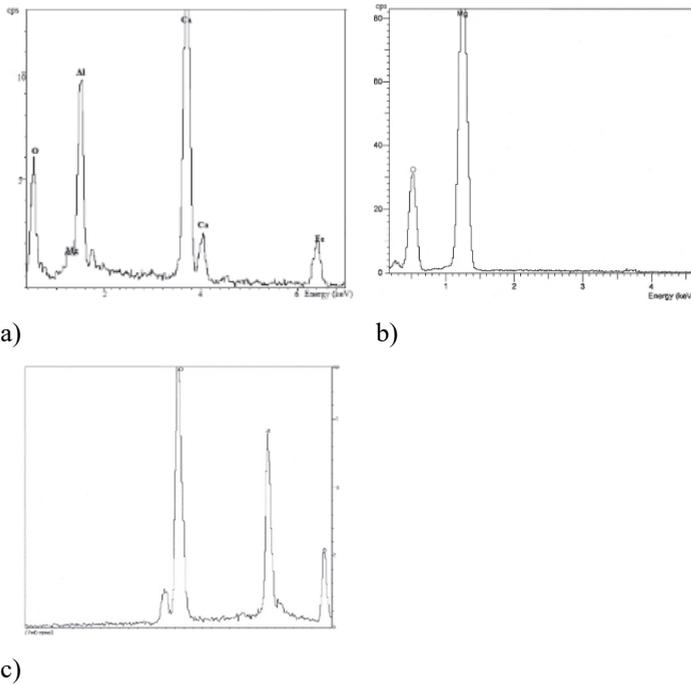


Fig. 6. Analytical spectrums of cement-asbestos slates, in temperature of 1500°C for 120 minutes, a) calcium and iron clay, b) periclase, c) calcium silicates (Poniatowska 2008)

Decomposition of chrysotile causes the release of silica, which leads to a preferential bonding with calcium oxide (resulting from the decarboxylation of CaCO_3 present in the old cement), resulting in the formation of dicalcium silicate, i.e. larnite. The calcination process leads to secondary clinkerisation, the main component of which is larnite, in cement terminology called belite and periclase resulting from the decomposition of chrysotile asbestos.

4. Conclusion

Based on the conducted research it can be established that thermal treatment of asbestos containing waste causes changes in the fibrous structure of the material. With the increase in temperature asbestos is transforming into neomorphic crystalline structures. In samples sintered in the temperature of 1250°C for 120 minutes trace amounts of chrysotile were found. When the temperature was increased to 1400°C , based on X-ray diffraction patterns, there were no traces of chrysotile. In this sample a significant recrystallization of alloy components and the beginning of periclase crystallization occurred, which is confirmed by photos of scanning electron microscope, analysis of the sample composition in the micro-area and the results of the phase composition analysis. When the temperature was increased to 1500°C , chrysotile contained in asbestos tile releases silica during decomposition, which binds to the calcium oxide from the cement phase. This produces dicalcium silicate (larnite), brownmillerite and periclase as a product of chrysotile decomposition. In this way, periclase is produced, magnesium oxide does not bind in silicate as excess calcium has a preference for secondary silicate formation. The calcination process leads to a second clinkerisation of the sinter components, the main components of which are larnite and periclase resulting from the decomposition of chrysotile asbestos. This composition confirms disintegration of asbestos in tested material and the formation of completely new, harmless phases. In case of samples sintered in the temperature of 1500°C for 30 minutes, phase composition studies were not carried out and thus it cannot be unequivocally stated that chrysotile asbestos has been eliminated. However, SEM image shows that the structure of the sample has transformed. Dark asbestos fiber residues are visible, which are probably magnesium oxides and dicalcium silicate. Extension of sintering time from 30 to 120 minutes for a temperature of 1500°C significantly changed the image of the SEM sample: there are no traces of asbestos fiber residue. Coarse crystalline calcium silicates and periclases are visible. To sum up, on the basis of thermal decomposition of cement-asbestos slates, it can be concluded that sintering of cement-asbestos slates at a temperature of $1400\text{-}1500^\circ\text{C}$ causes a remold of the fibrous structure of chrysotile asbestos it contains. With the use of a method enabling such

temperatures to eliminate the emission of asbestos fibers into the air (e.g. during cement production), subjecting it to heat treatment, we permanently change its structure and thus it becomes a mineral neutral to human health. Moreover, the material obtained gives potential uses for cement production (Witek et. al. 2012). This paper may provide a basis for further research on the possibility of thermal processing of asbestos-containing waste, in which the addition of fluxes to the process may also be considered, which reduce the decomposition asbestos temperature in asbestos cement (Poniatowska 2008).

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Abstract

Negative impact of products including asbestos and asbestos wastes on human health comes from asbestos' needle-like, fibrous structure. To terminate its negative influence on the environment, including human health, processes aimed at its destruction should be conducted. Basing on available literature one can ascertain that only chemical and thermal methods of asbestos fibers translation cause the ensuing waste to be health neutral. The aim of this paper was to evaluate the influence of temperature and time on asbestos decomposition and determine if the asbestos fiber structure is damaged at high temperatures. In the first stage of heating cement-asbestos slates, the physicochemical processes occurring in the material under investigation will involve dewatering, i.e. removal of adsorptive, constitutional and crystallizing water, followed by breaking down the chrysotile structure and destruction of its brucate layers, finally crystallization of the forsterite and later its disintegration into silica and periclase. During the sintering of asbestos cement we have an excess of calcium oxide coming from cement components. Chrysotile contained in asbestos cement exorcises during its dissolution silica which merges with calcium oxide from the cement phase. Thus arises bicalcium silicate (larnite), brownmillerite and periclase as a product of chrysotile dissolution. Periclase is therefore formed, magnesium oxide does not bind in silicate because the excess of calcium has the preference for the secondary formation of silicates. The process of sintering causes its contents to clinkering again, the main component of which is larnite and periclase resulting from the decay of chrysotile asbestos. It causes potential possibilities of using the products of this method to produce cement.

During the tests, in the temperature rises to 1400°C, there is a significant recrystallization of components and the beginning of the crystallization of periclase, which is confirmed by scanning tests, analysis of sample composition in the micro-area and results of phase composition analysis. To sum up, based on the conducted studies of the thermal decomposition of cement-asbestos slates, it can be concluded that sintering it at a temperature of 1400-1500°C leads to the transformation of the fibrous structure of the chrysotile

asbestos contained therein. With the use of a method that allows the operation of such temperatures, eliminating the emission of asbestos fibers into the air, subjecting it to thermal treatment, we permanently change its structure and in this way it becomes a mineral indifferent to human health. In addition, the obtained material gives the potential to use products of this method of neutralizing cement-asbestos slates, e.g. in construction. This paper might be the basis for further research of the possibilities of thermal processing of asbestos-containing waste, in which the addition of fluxes to the process, affecting the decomposition temperature of asbestos in cement-asbestos slates can also be considered.

Keywords:

asbestos, asbestos wastes, asbestos cement, thermal decomposition of chrysotile asbestos

Termiczny rozkład włókien azbestu występującego w odpadach eternitu**Streszczenie**

Negatywny wpływ wyrobów i odpadów zawierających azbest na zdrowie ludzkie wynika z igłowej, cienko włóknistej struktury azbestu. Dlatego w celu zlikwidowania jego negatywnego oddziaływania na środowisko, w tym zdrowie ludzi, należy prowadzić procesy prowadzące do zniszczenia jego struktury. Na podstawie dostępnej literatury można stwierdzić, że jedynie metody chemiczne i termiczne, mające wpływ na przekształcenie włókien azbestowych powodują, że powstały w wyniku ich zastosowania odpad jest objęty dla zdrowia. Celem niniejszej pracy była ocena wpływu temperatury i czasu na rozkład azbestu i określenie, czy włóknista struktura azbestu ulega zniszczeniu w wysokich temperaturach. W pierwszym etapie ogrzewania eternitu procesy fizykochemiczne zachodzące w badanym materiale wiązały się z odwadnianiem, czyli usuwaniem wody adsorpcyjnej, konstytucyjnej i krystalizacyjnej, a następnie z rozbijaniem struktury chryzotyłu i niszczeniem jego warstw brucytowych, w końcu krystalizacją, powstałego w trakcie narastania temperatury, forsterytu oraz późniejszym jego rozpadem na krzemionkę i peryklaz. W procesie spiekania eternitu mamy do czynienia z nadmiarem tlenu wapnia pochodzącego ze składników cementu. Chryzotyl zawarty w eternicie uwalnia podczas rozpadu krzemionkę, która łączy się z tlenkiem wapnia z fazy cementowej. W ten sposób powstaje krzemian dwuwapniowy (larnit), brownmilleryt, a także peryklaz, jako produkt rozpadu chryzotyłu. Peryklaz powstaje w związku z tym, tlenek magnezu nie wiąże się w krzemian, ponieważ nadmiar wapnia posiada preferencje, przy wtórnym tworzeniu krzemianów. Proces spiekania prowadzi do powtórnej klinkieryzacji składników spieku, której głównym składnikiem jest larnit oraz peryklaz powstający z rozpadu azbestu chryzotylowego. Taki skład potwierdza dezintegrację azbestu w badanym materiale i powstanie całkiem nowych, nieszkodliwych dla środowiska faz. Stwarza to potencjalne możliwości wykorzystania produktów tej metody unieszkodliwiania eternitu do produkcji cementu. Jak stwierdzono w trakcie badań, dopiero przy wzroście temperatury do 1400°C następuje znacząca rekrytalizacja składników stopu i początek krystalizacji peryklazu, co potwierdzają badania skaningowe, analiza składu próbek w mikroobszarze oraz wyniki analizy składu fazowego. Reasumując na podstawie przeprowadzonych badań termicznego rozkładu eternitu można stwierdzić, że spiekanie go w temperaturze 1400-1500°C prowadzi do przekształcenia

struktury włóknistej zawartego w nim azbestu chryzotylowego. Przy zastosowaniu metody umożliwiającej działanie takich temperatur, eliminującej emisję włókien azbestowych do powietrza, poddając go obróbce termicznej trwale zmieniamy jego budowę i w ten sposób staje się on minerałem obojętnym dla zdrowia ludzkiego. Ponadto uzyskany materiał, daje potencjalne możliwości wykorzystania produktów tej metody unieszkodliwiania eternitu np. w budownictwie. Niniejsza praca może być podstawą do dalszych badań nad możliwością termicznego przetwarzania odpadów zawierających azbest, w których można rozważyć także dodawanie topników do procesu, wpływających na obniżanie temperatury rozkładu azbestu w eternicie.

Słowa kluczowe:

azbest, odpady azbestowe, eternit, termiczny rozkład azbestu chryzotylowego