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A thermodynamic model of CO_2 sequestration in aqueous solutions of selected waste

Introduction

Mineral sequestration of CO_2 applying waste is an interesting way of reducing anthropogenic emissions of carbon dioxide. Research on attempts to use waste for the needs of CO_2 sequestration concentrates on the kinds of waste which contain calcium compounds reacting with CO_2 and which alkalize the environment in the presence of water (high pH of the solution). In addition, aqueous solutions of waste should have an adequately high buffer capacity to ensure permanent binding without the risk of starting the undesirable reaction of creating calcium bicarbonate. Permanent binding of carbon dioxide is also ensured by the pozzolanic qualities of waste in connection with a high content of free CaO. In this way, as a result of a chain of simple (portlandite) and more complicated (silicates and aluminosilicates) reactions the final capability of this waste to sequester carbon dioxide is shaped. Solubility is another important feature of waste, relevant for the sequestration process. For example at a ratio ash: water 1:10 ca. 100% of the soluble part is transferred to the solution, whereas at a ratio of 2:1 it is 2% up to a maximum of 50% (Łączny et al. 1995).

Mineral waste which can be used for mineral sequestration of CO_2 must have a high content of CaO. Among others such content have first of all: fly ashes, fluidized ashes, steel slag, cement kilns dust.

A particularly interesting material for CO_2 sequestration are ashes from fluidized bed combustion, whose economic applications are restricted.

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Mineral carbonation in waste is mostly conducted as direct carbonation applying waste-water suspensions with different water to waste ratios (among others: Stolaroff et al. 2005; Huijgen and Comans 2005; Huijgen 2007; Uliasz-Bocheńczyk et al. 2007; Uliasz-Bocheńczyk 2009).

Products of the hydration of ashes, which are above all subject to carbonation, are: $Ca(OH)_2$, hydrated calcium silicates C-S-H ($CaO \cdot nSiO_2 \cdot mH_2O$) and ettringite ($3CaO \cdot Al_2O_3 \cdot 3CaSO4 \cdot 32H_2O$) (Kurdowski 2010). Carbonized products of hydration also include hydrated hexagonal calcium aluminates.

Carbonation is an exothermic reaction. The conformity of chemical and physical methods is deliberately stressed here, as both in the first and the second case it is free calcium oxide which is responsible for the result of the process. It is also essential to take into account such parameters as: heat of wetting and heat of hydration, hydrophilicity coefficient, specific surface, the total quantity of CaO and the quantity of bound CaO, the amount of insoluble matter, etc.

Mineral sequestration of CO_2 is a complex process, which is divided in case of waste-water suspensions into the following stages (Sun, Simons 2008; Fernandez Bertos et al. 2004; Rendek et al. 2006):

1. Diffusion of CO_2 into the suspension.
2. Solvation of $CO_2(g)$ into $CO_2(aq)$.
3. Creation of H_2CO_3.
4. Dissociation of H_2CO_3 into H^+, HCO_3^-, CO_3^{2-}.
5. Migration of Ca^{2+} ions from the sorbent's phases into the solution.
6. Nucleation of carbonates.
7. Precipitation of carbonates.

The article presents the results of research and a computer simulation prepared in the programme Phreeqc Interactive, aiming at analyzing the individual stages of the process of mineral sequestration of CO_2 applying fluidized ashes from the Tychy heat and power plant. The suspensions selected for research and analysis are noted for high absorption of CO_2: 5.32 g of CO_2/100 g of ashes (Uliasz-Bocheńczyk 2009).

1. Description of the ashes used in research

Ashes from fluidized beds differ from conventional ashes in their phase content and grain development. They consist mainly of irregular grains of dehydrated and dehydroxylated minerals of gangue, quartz, anhydrite, free CaO, $Ca(OH)_2$ and unburnt coal. The ashes may also contain: sodium and potassium chlorides, sodium and potassium sulphates, sodium and potassium nitrates coming from the stream of flue gas and resorbing on the surface of solid components (Roszczczynialski, Gawlicki 2004).

Ashes obtained from fluidized beds are distinctive in that they don't containt any amorphous phase or mullite (Gawlicki 2007; Giergiczny 2006). Due to their characteristic composition, the ashes obtained from biomass co-firing should contain some amorphous phase, because fluxes (sodium and potassium compounds) significantly reduce the melting temperature of the fuel's mineral part. Table 1 presents a simulation confirming the above-mentioned assumptions.

The ashes used in the research were obtained in CYMIC fluidized beds of the Tychy power plant from the combustion of bituminous coal with a maximum of 5% additional biofuel (Maryamchik, Wietzke 2000).

TABLE 1

Calculated ranges of viscosity and surface tension in ashes obtained from fluidized beds (Wałek 2006)

TABELA 1

Symulacja dotycząca popiołów z kotłów fluidalnych

Fly ash	Description	Comments
Fly ash obtained from the combustion of bituminous coal (A)	Dynamic viscosity – optimal for the temperature range of 1346–1447°C Surface tension – optimal for the temperature range of 500 and 872°C	No common range
Fly ash obtained from the combustion of common osier woodchip (B)	Dynamic viscosity – optimal for the temperature range of 714–805°C Surface tension – optimal for the temperature range of 500–838°C	The ranges coincide for temperatures 714–805°C
80% of A + 20% of B	Dynamic viscosity – optimal for the temperature range of 1199–1301°C Surface tension – optimal for the temperature range of 500–861°C	No common range

TABLE 2

Oxide composition of the ashes from Tychy Power plant [%]

TABELA 2

Skład tlenkowy popiołu Ec. Tychy [%]

Oxide	Content	Oxide	Content
SiO_2	24.82	K_2O	1.58
Al_2O_3	14.47	SO_3	6.18
Fe_2O_3	4.07	TiO_2	0.38
CaO	25.26	P_2O_5	0.55
MgO	2.77	BaO	0.13
Na_2O	0.46	ZnO	not observed

Source: author's study

The main phases observed in aqueous suspensions of fly ashes from Tychy power plant are: calcite, quartz, anhydrite, portlandite, ettringite and calcium silicates (Uliasz-Bocheńczyk 2009).

Table 3 presents the composition and pH of an aqueous extract obtained from mixing ash with water at a weight ratio 1:10.

TABLE 3

Chemical composition and pH of an aqueous extract obtained from mixing ash with water at a weight ratio 1:10 (experimental data) [mg/dm^3]

TABELA 3

Skład chemiczny wyciągu wodnego oraz jego pH uzyskanego po zmieszaniu popiołu z wodą w stosunku 1:10 (dane eksperymentalne) [mg/dm^3]

Element	Content
S as SO_4^{2-}	1 932.5
Cl	196.4
Na	18.5
K	10.8
Ca	1 757.4
Al	15.0
Si	35.0
pH	12.0

Source: author's study

2. Thermodynamic model

The elaborated model is an attempt at identifying the kind and order of chemical reactions leading to permanent binding of carbon dioxide observed in aqueous ash suspensions as exemplified by waste from the Tychy power plant. The phase composition of the ashes allows us to formulate the thesis that there is a possibility of broadening the thermodynamic model to include other waste containing similar crystalline and amorphous solids (steel slags, cement kiln dusts, waste from dry and semi-dry desulphurization of flue gases). Regardless of the number of the phases they have one common feature: the ability to enter reactions of the kind of hydraulic bonds or pozzolana features.

The calculations were conducted in the programme Phreeqc Interactive, which is meant to be used for hydrogeochemical modeling, but which can also be used to solve problems in various other areas. The program lets you conduct a series of calculations and simulations, i.e. speciation and saturation indicators' distribution, reactions of dissolution and precipitation of minerals, ion exchange, surface complexation, kinetic processes, one-dimensional transportation and inverse modelling (Parkhurst, Appelo 1999).

Mineral sequestration of CO_2 is analyzed in literature in relation with brines, fly ashes (obtained from the combustion of household waste, as well as bituminous and lignite coal), steel slags, cement kiln dusts, paper waste, etc. Phreeqc is mainly used here to calculate the saturation coefficient (SI), individual solid phases which can precipitate in the above-mentioned systems and to simulate possible speciations of ions contained in the solution (Gitari et al. 2008; Montes-Hernandezi et al. 2009; Perez-Lopez et al. 2008, 2010). The saturation coefficient specifies the state of the aqueous solution as far as each individual phase is concerned (balance, supersaturation or undersaturation of a particular phase). The assessment of the solution's saturation with a particular (solid or gas) phase consists of comparing the product of activities of the forms taking part in the reaction (in the real solution) with the product of activities of the same forms in equilibrium conditions (with a constant K).

In addition, Phreeqc creates simulations of the influence of the solution's pH, CO_2 pressure and temperature on the effectiveness of the carbonate creation reaction (Soong et al. 2004). Based on literature kinetic data of selected minerals Wigand et al. prepared a time-dependent model describing the changes in the solution's concentration and the mineral phases' content (Wigand et al. 2008).

During the calculations it is necessary to take into consideration certain restrictions of the programme deriving from the equations used to calculate the ions' activity coefficients. The programme uses the Debye-Hűckel equation or Davies equation, which is applied to solutions whose ionic strength does not exceed 0.5. The programme includes also a database for the calculations of ions' activity coefficients by means of the Pitzer method. However, the data are not complete and need to be supplemented with further ions and mineral phases.

The calculations were conducted in the following stages:

- choice of a standard composition of an aqueous suspension for a fly ash,
- simulation of an equilibrium composition of the aqueous ash suspension at a water to ash ratio 1:1,
- simulation of mineral carbonation of the aqueous ash suspension.

On the basis of laboratory research results (Table 2 and 3) the equilibrium composition of the solution after vaporizing 9 parts of water by weight, i.e. 50 moles, was calculated in Phreeqc (database lln.dat). Tables 4 and 5 present the standard composition of the aqueous ash suspension as exemplified by ash from the Tychy heat and power plant.

The following step was to conduct a simulation consisting of gradual adding of CO_2 to the aqueous ash suspension (0.1 moles in 100 steps). Figures 1a and 1b present:

- the change in calcite quantity in relation to the quantity of introduced CO_2,
- the change in ettringite quantity in relation to the quantity of introduced CO_2,
- the change in gypsum quantity in relation to the quantity of introduced CO_2,
- the change in portlandite quantity in relation to the quantity of introduced CO_2,

- the change in wollastonite quantity in relation to the quantity of introduced CO_2,
- the change in calcium ions' concentration in relation to the quantity of introduced CO_2.

Figure 2 presents experimental data on the absorption of CO_2, as well as pressure and temperature changes in the chamber containing the aqueous ash suspension from the Tychy heat and power plant (Uliasz-Bocheńczyk 2009). The research was conducted in the

TABLE 4

Composition and pH of the aqueous ash suspension (1:1) (simulation results)

TABELA 4

Skład oraz pH zawiesiny wodno-popiołowej (1:1) (wyniki symulacji)

Solution composition	
Element/ion	[mol/kg$_w$]
Al	$3.15 \cdot 10^{-7}$
Ca	$1.77 \cdot 10^{-1}$
Cl	$3.26 \cdot 10^{-1}$
K	$2.80 \cdot 10^{-3}$
Na	$8.15 \cdot 10^{-3}$
S	$8.29 \cdot 10^{-3}$
Si	$3.45 \cdot 10^{-7}$
OH^-	$1.34 \cdot 10^{-2}$
H^+	$1.35 \cdot 10^{-12}$
pH	11.97

Source: author's study

TABLE 5

Phase composition of the aqueous ash suspension (1:1) (simulation results)

TABELA 5

Skład fazowy zawiesiny wodno-popiołowej (1:1) (wyniki symulacji)

Mineral phase	[mol]	[g]
Ettringite	$2.780 \cdot 10^{-4}$	0.35
Gypsum	$1.847 \cdot 10^{-2}$	3.18
Portlandite	$5.694 \cdot 10^{-3}$	0.42
Wollastonite	$5.826 \cdot 10^{-4}$	0.09

Source: Author's study

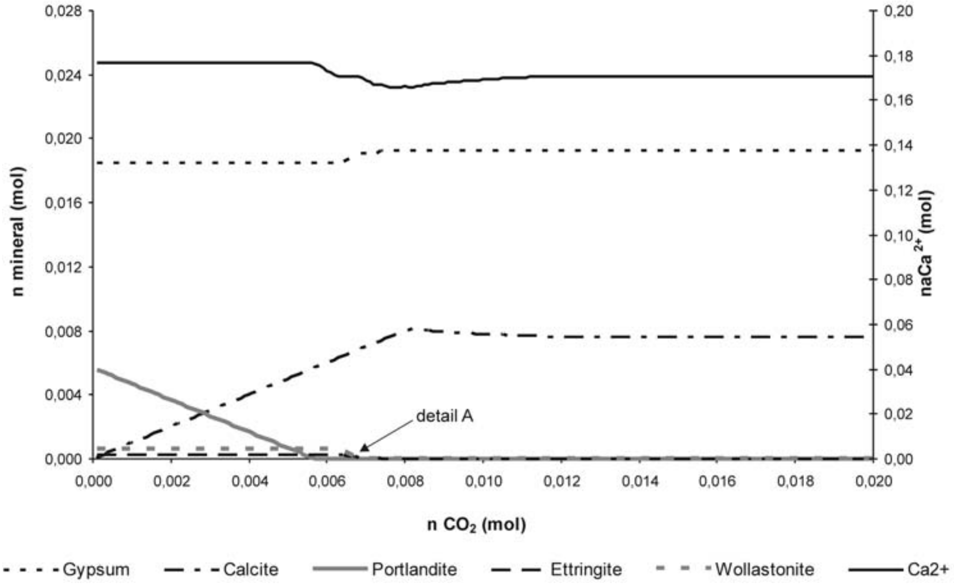

Fig. 1a. Changes in the quantity of selected mineral phases in relation to the introduced CO_2
Source: author's study

Rys. 1a. Zmiana ilości wybranych faz mineralnych w funkcji ilości dodawanego CO_2

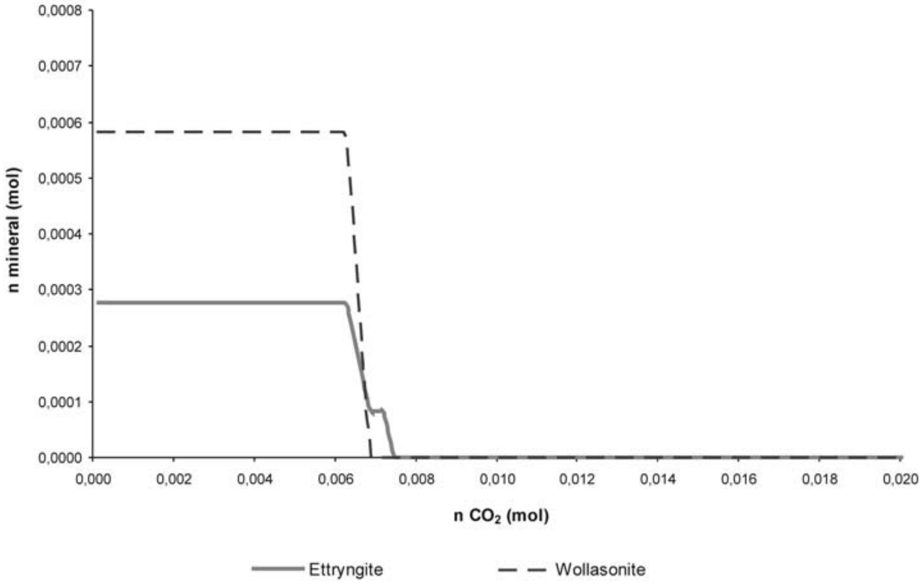

Fig. 1b. Changes in the quantity of ettringite and wollasonite in relation to the introduced CO_2
(detail A of figure 1a)
Source: author's study

Rys. 1b. Zmiana ilości ettryngitu i wollasonitu w funkcji ilości dodawanego CO_2

Department of Environmental Engineering and Mineral Processing of the Faculty of Mining and Geoingineering at the AGH University of Science and Technology (Uliasz-Bocheńczyk et al. 2007; Uliasz-Bocheńczyk 2009). The system consists of two measuring units and each unit comprises two chambers, a logger (registering tool), a gas cylinder and a regulator. Initial preparation of suspensions was conducted by mixing waste and water in a laboratory

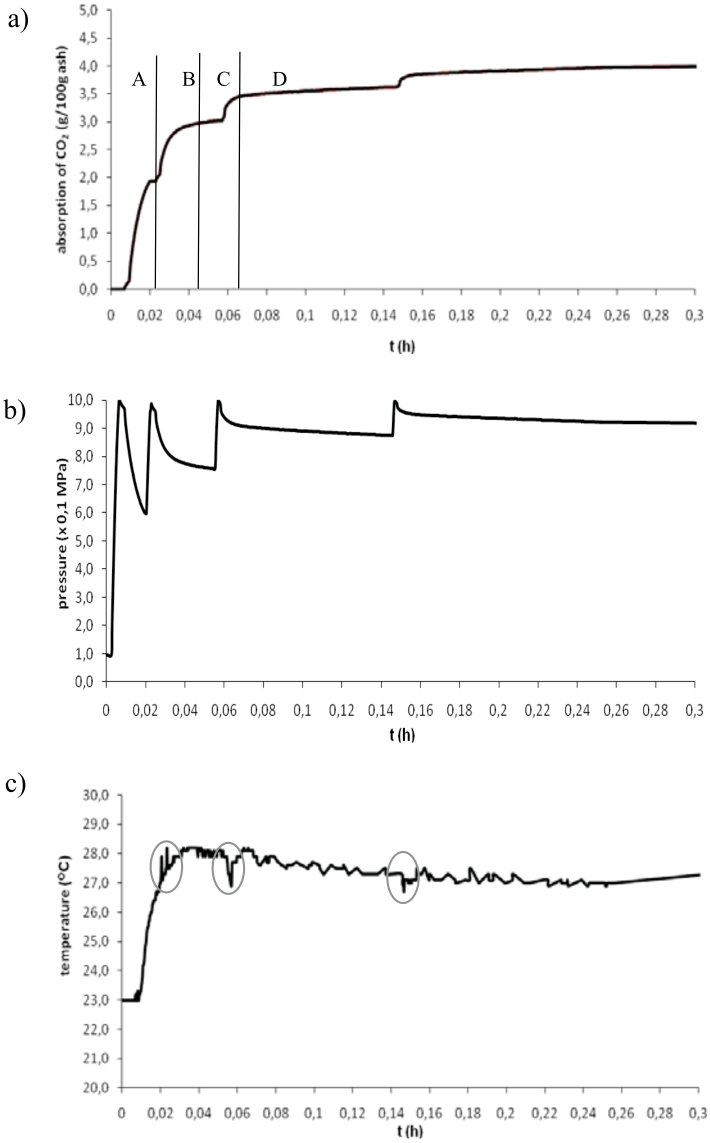

Fig. 2. Absorption of CO_2 (a), changes of pressure (b) and temperature (c) in the chamber containing the aqueous ash suspension in the Tychy heat and power plant (Uliasz-Bocheńczyk 2009)

Rys. 2. Pochłanianie CO_2 (a), zmiana ciśnienia (b) i temperatury (c) w komorze dla zawiesiny wodno-popiołowej Ec. Tychy mixer. Then the suspensions were introduced into the research chambers. On closing the chamber tightly measuring and registering equipment was connected and the inflow of CO_2 was opened. The chambers containing the suspensions were shaken in a rocking shaker. Pressure and temperature in the chamber were measured during the research (Uliasz-Bocheńczyk 2009).

3. Analysis of the experiment and simulation results

Results of the simulation presented in Figure 1a and 1b show which mineral phases react with CO_2 and in which order the above reactions take place in the aqueous ash suspension. The shape of the curves presented in Figure 2 demonstrates the complex character of mineral sequestration of carbon dioxide.

The order of reactions, which have been ascribed to individual stages, may be linked to the value of Gibbs free energy. Table 6 contains the values of Gibbs free energy and the equilibrium constants for the reactions of carbonation of portlandite, ettringite and wollastonite.

TABLE 6

Gibbs free energy and equilibrium constants for selected reactions of carbonation

TABELA 6

Entalpia swobodna oraz stałe równowagi wybranych reakcji karbonizacji
(Kurdowski 2010; Stumm, Morgan 1999)

No.	Chemical equation		G_{298} [kJ/mol]	$\ln K_{298}$
1	Portlandite	$Ca(OH)_2 + CO_2 = CaCO_3 + H_2O$	–74.61	30.11
2	Ettringite	$1/3(3CaO \cdot Al_2O_3 \cdot 3CaSO_4 \cdot 32H_2O) + CO_2 =$ $= CaCO_3 + CaSO_4 \cdot 2H_2O + 2/3Al(OH)_3 + 23/3H_2O$	–42.96	17.34
3	Wollastonite	$CaO \cdot SiO_2 + CO_2 = CaCO_3 + SiO_2$	–41.18	16.62

Area A

On introducing gas CO_2 into the suspension it is gradually dissolved. Most molecules are dissolved in water creating nonpolar molecules, but part of them reacts with water creating a weak carbonic acid, which in its turn undergoes two-stage dissociation:

$$CO_2 + H_2O = H_2CO_3 = H^+ + HCO_3^- = 2H^+ + CO_3^{2-} \qquad (1)$$

Because the aqueous ash suspension has a pH of ca. 12, the predominant form is CO_3^{2-} (Maciaszczyk, Dobrzyński 2007). This stage is represented in graph 2a by a horizontal line. Portlandite dissolves, calcium ions pass from the solid phase into liquid phase and react with carbonate ions:

$$Ca^{2+} + CO_3^{2-} = CaCO_3\downarrow \tag{2}$$

This stage is represented on the graph by a quickly rising curve.

Area B

After the whole portlandite has been dissolved the carbonation process slows down. The horizontal line in the graph corresponds to the "activation" stage of the next mineral phase. The speed of the process rises again only when ettringite calcium ions pass into the solution. The ettringite releases sulphate ions as well, so in given conditions part of calcium ions precipitates as gypsum:

$$1/3\,(3CaO \cdot Al_2O_3 \cdot 3CaSO_4 \cdot 32H_2O) + CO_2 = \tag{3}$$

$$CaCO_3 + CaSO_4 \cdot 2H_2O + 2/3\,Al(OH)_3$$

Area C

In area C takes place the "activation" process of calcium silicate (slow-down) similar to area B, calcium ions are released from the mineral phase to the solution and they react with carbonate ions:

$$CaSiO_3 + 2H^+ = Ca^{2+} + SiO_2 + H_2O \tag{4}$$

$$Ca^{2+} + CO_3^{2-} = CaCO_3\downarrow \tag{5}$$

Area D

In this area the whole available quantity of mineral phases contributing calcium ions to the system has been dissolved. Carbon dioxide in its turn can take part in the undesirable reaction conditioning the dissolution of calcite:

$$CaCO_3 + CO_2 + H_2O = Ca^{2+} + 2HCO_3^- \tag{6}$$

The point where the function (Fig. 1a) reaches its highest value represents the maximum quantity of carbon dioxide which can be introduced into the system without carbonates turning into bicarbonates.

Summary

The elaborated thermodynamic model of CO_2 sequestration in an aqueous ash suspension adequately demonstrates the complexity of the process.

It was demonstrated that by means of the Phreeqc programme it is possible to determine the kind and order of occurrence of reactions responsible for binding carbon dioxide. The model can be used to calculate the kinetics of the process and consequently to calculate the reactor.

In addition, the results of the analysis conducted confirmed the results of laboratory research carried out applying other waste (Uliasz-Bocheńczyk 2009). Finally, they allow for a fuller description of the process.

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TERMODYNAMICZNY MODEL SEKWESTRACJI CO_2 W WODNYCH ROZTWORACH WYBRANYCH ODPADÓW

Słowa kluczowe

Odpady mineralne, mineralna sekwestracja CO_2, popioły z kotłów fluidalnych, modelowanie

Streszczenie

Mineralna sekwestracja przy zastosowaniu odpadów mineralnych jest interesującą próbą połączenia dwóch ważnych problemów ekologicznych: redukcji antropogenicznej emisji CO_2 poprzez jego trwałe wiązanie oraz pełniejszego wykorzystania odpadów o ograniczonym zastosowaniu gospodarczym. Do wiązania CO_2 poprzez mineralną sekwestrację powinny być stosowane odpady o wysokiej zawartości CaO i wolnego CaO. Interesującymi odpadami do sporządzania zawiesin wodnych do zastosowania ich dla mineralnej sekwestracji CO_2 są popioły fluidalne. W artykule przedstawiono wyniki modelowania dotyczące procesów zachodzących w zawiesinach wodnych na przykładzie zawiesin sporządzonych z popiołem fluidalnym z Ec. Tychy poddanych działaniu CO_2. Zawiesiny te wybrano na podstawie wyników wcześniejszych badań. Przedstawiony w artykule model jest próbą identyfikacji rodzaju i kolejności reakcji chemicznych w wodnych zawiesinach popiołowych poddanych działaniu CO_2 prowadzących do trwałego wiązania ditlenku węgla. Wykonane modelowanie potwierdziło złożoność procesu mineralnej sekwestracji oraz wyniki badań innych autorów.

A THERMODYNAMIC MODEL OF CO_2 SEQUESTRATION IN AQUEOUS SOLUTIONS OF SELECTED WASTE

Key words

Mineral waste, mineral sequestration of CO_2, ashes from fluidized beds, modelling

Abstract

Mineral sequestration using mineral waste is an interesting attempt at combining the solving of two important ecological problems: the reduction of anthropogenic emissions of CO_2 by permanent binding and a fuller use of waste with restricted economic applications. The waste used to bind CO_2 by way of mineral sequestration should have a high content of CaO and free CaO. Fly ashes from fluidized beds seem interesting in terms of preparing aqueous suspensions to be used in mineral sequestration of CO_2. The article presents the results of modelling of the processes occuring in aqueous suspensions as exemplified by suspensions prepared on the basis of fluidized ashes from the power plant in Tychy subjected to the effects of CO_2. The selection of these suspensions was based on the results of previous research. The thermodynamic model presented in the article is an attempt to identify the kind and order of occurrence of chemical reactions leading to permanent binding of carbon dioxide observed in aqueous ash suspensions subjected to the effects of CO_2. The elaborated model confirmed both the complexity of the mineral sequestration process in this kind of waste and the results of research conducted by other authors.