

Determination of Thermodynamic Function Parameters of Tedzami Clinoptilolite

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(Presented by Academy Member Ramaz Khurodze)

Abstract. The thermodynamic function parameters of the Tedzami clinoptilolite, including its dehydration and carbonation by CO₂ sorption, as well as rehydrolime pozzolanization of cement have not yet been sufficiently studied. This lack of data limits its application in industrial and environmental projects aimed at reducing the amount and footprint of CO₂ emitted into the atmosphere during cement production. The study involved predicting the thermodynamic function parameters of raw and dehydrated Tedzami clinoptilolite and the possibility of calcium hydroaluminosilicate formation and during cement hardening using approximate methods of calculation. The research findings revealed the following properties of Tedzami clinoptilolite: transition from a hydrated to an anhydrous state; physicochemical sorption of CO₂ generated during technological process; participation in rehydrogenation pozzolanization and carbonization of cement to reduce or eliminate the CO₂ footprint and contribution to solving environmental issues through ecologization of cement technology. © 2026 Bull. Natl. Acad. Sci. Georg.

Keywords: aluminosilicate, capture, enthalpy, modification, portlandite

Introduction

At the beginning of the 21st century, more than 40 natural zeolites (ZEO_{nat}), were known, differing from each other in crystal structure, cation content, and *Si/Al* ratio. Among them, tuffs (TF) containing the mineral clinoptilolite (CPT) are characterized by their stable composition and particularly high CPT content (70-95%) (Tsitsishvili et al., 1992).

Dehydrated CPT exists in a metastable, nonequilibrium state, which contributes to its physicochemical, especially sorption activity (Smith, 1999; Navrotsky and Tian, 2001). The temperature at which dehydration begins, in addition to the rate and duration of heating, is reflected in the thermodynamic (THD) function parameters (Wendlandt, 2024). Due to the gas separation ability of CPT, it is widely used as gas/water filters and in environmental technologies (Skhvitaridze et al., 2024).

With the flue gases of the clinker kiln of a cement plant, ≤ 890 kg of CO₂ is emitted into the atmosphere per 1 ton of clinker produced, which represents the corresponding carbon footprint =(CFP). CO₂ is

considered the main greenhouse gas responsible for climate change. Accordingly, numerous technological methods have been developed to prevent it. In projects such as Carbon Capture, Utilization and Storage (CCUS), CO₂ is captured in a FF containing the zeolite mineral – CPT. In this direction, positive results have been obtained using the CPT-TF of the Khekordzula deposit in Georgia (Skhvitaridze et al., 2024). The “green technology” for cement production “CO₂ ZEOCEM” was developed, and selected as one of the 19 finalists out of 101 participants in the global competition for innovative technologies of the “Global Cement and Concrete Association” (GCCA).

Studies have substantiated the use of natural hydrous aluminosilicates modified by calcination as effective mineral additives to cement, as well as the possibility of using CPT-TF as a sorbent to reduce emissions and traces of CO₂ into the environment (Skhvitaridze et al., 2024). For the adsorption of CO₂ in CPT-TF, it is practically necessary to dehydrate it to a temperature of $\leq 400^\circ\text{C}$, while thermodynamically (as THD) the onset of dehydration is expected at $t = 79.75^\circ\text{C}$ and $P = 684 \text{ mm Hg Torr}$. It is assumed that during the cyclic process of CO₂ sorption-regeneration, CO₂ forms “adduct compounds” with alkaline (Na¹⁺, K¹⁺) and alkaline earth (Ca²⁺, Mg²⁺) metals CPT (Davarpanah et al., 2020).

Using the above-mentioned assumptions, adding mineral additives into cement contributes to the reduction of CO₂ emissions into the atmosphere and carbon footprint formed by flue gases.

Materials and Methods

Research materials are as follows: 1. TZCPT mineral in quantity $\leq 95\text{mass}\%$ in 0-30 mm clastic tuff of Tedzami deposit, located in Georgia, where the remaining 5 mass% is quartz and traces of other minerals. TZCPT together with Khekordzula CPT-TF (Skhvitaridze et al., 2024) has been periodically utilized in Georgian cement production since 2000.

The research methods include the prediction of the potential formation of calcium hydrosilicate and aluminosilicate. Determination of the functions Thermodynamic (THD) function parameters of TZCPT were determined, including the enthalpy of formation $\Delta_f H_{298}^0$, Gibbs free energy ($\Delta_f G_{298}^0$), and entropy $\Delta_f S_{298}^0$. In addition, key parameters such as the temperature of the onset of dehydration and partial pressure according to the guidelines set out in]. In the context of participation in CCUS processes, the physicochemical (CO₂) sorption parameters and Ca(OH)₂ sorption parameters in TZCPT were determined.

The research objects are as follows;a) dehydration of TZCPT and absorption of CO₂ – processes of capture of T (K) or t ($^\circ\text{C}$) and P ; b) TZCPT_{deh}, with CO₂ (ad)sorption into carbonate pozzolan (CARP) and Ca(OH)₂ (ab)sorption into rehydro-lime pozzolan (Rehydro-liming Pozzolna=RHLP), respectively, the possibility of modifying TZCPT_{carp} and TZCPT_{RHLP}.

The research employed thermal (DTA) and X-ray diffraction (XRD) methods of solid, powdery materials. Mathematical analysis, including a thermodynamic approach was applied for the evaluation of cement testing methods. Laboratory dryers and muffle furnaces were used for sample preparation.

Table 1. Coefficients a, b, c CPT_{nat} and TZCPT_{deh}, $\Delta_r C_p = f(T) \cdot \text{K}^{-1} \cdot \text{mol}^{-1}$

Title	<i>a</i>	<i>b</i>	<i>c</i>
$\Sigma (a,b,c) \text{ TZCPT}_{\text{nat}} (a,b,c) =$	290.90	135.26	- 53.03
$\Sigma (a,b,c) \text{ TZCPT}_{\text{deh}} (a,b,c) =$	228.58	134.64	- 57.42

First of all, a search was conducted for chemical and THD constants of the materials used in the studies. These data, previously reported in Skhvitaridze et al., (2024)., are summarized in Table 1. The Table

presents the designations of the compounds participating and formed in the process of hydration and hardening of cement, the CCN process, as well as their data $\Delta_f G^\circ_{298}$ kJ/mol.

The X-ray phase analysis was performed on a DRON 4.0 diffractometer (scanning speed 10/20 = 1 g/s. Anode - Cu, radiation - K^α , I = 12 ml, filter - Ni, V = 30 kV), natural, 300°C and 700°C after heating for 1 hour. The characteristic peaks of the TZCPT according to the X-ray phase analysis data: [(2.97; 3.34; 3.95; 7.97; 8.792) (Fig. 1 and (Skhvitaridze et al., 2024)), 300°C and 700°C after 1 hour of calcination are repeated and do not change in magnitude. Thermal properties of TZCPT were determined on a NETZSCH derivatograph [STA-2500 analyzer, REGULUS thermogravimetric (TG), differential thermogravimetric (DTG) and differential thermal analyzer (DTA). TZCPT was heated to 1000°C at a rate of 10°C/min (Fig. 2).

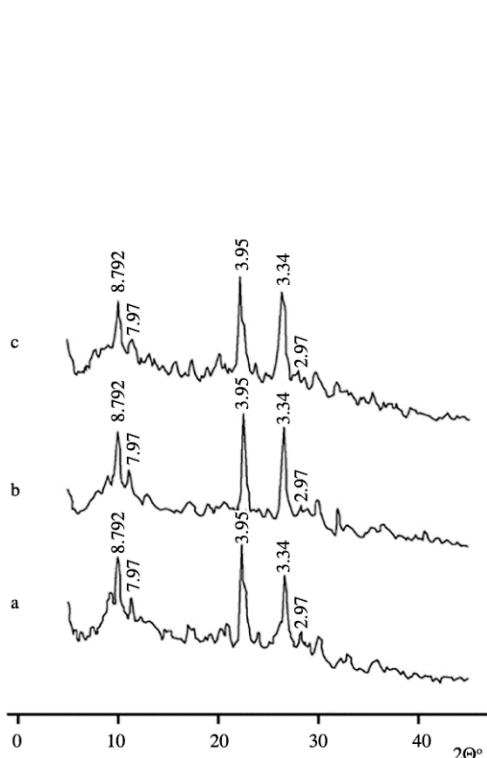


Fig. 1. Diffractogram of of CPT-TF.

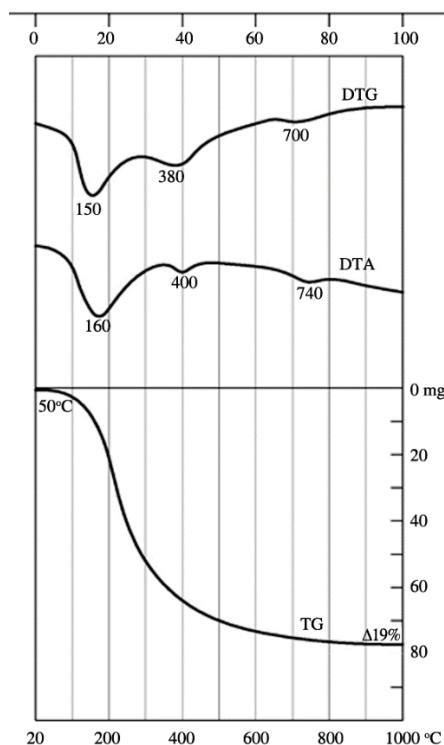


Fig. 2. Derivatogram of CPT-TF.

Table 2. Complete chemical analysis of TZCPT

Name	Chemical composition		Name	Chemical composition	
	Mass. %	EU of ion		Mass. %	EU of ion
Na ₂ O	4.23	Na 3.71	Fe ₂ O ₃	1.63	Fe 0,55
K ₂ O	1.52	K 0.88	SiO ₂	60.10	Si 28.02
CaO	3.90	Ca 1.89	TiO ₂	0.28	Ti 0.11
MgO	0.90	Mg 0.61	O(g)	-	Og 72.00
Al ₂ O ₃	13.95	Al 7.88	H ₂ (l)	13.49	H ₂ O.20.27

Using the method of chemical-oxidative analysis (Table 2) and according to Tsitsishvili et al. (1992), the summation method, the generalized stoichiometric composition of the unit cell (UC) of zeolites was determined:

$$\zeta = (M', M''_{0.5})x(H_2O)_n[Al_xSi_{1-x}O_2], x \in (0;1). \tag{1}$$

Here M', M" (non-framework compounds) are cations of alkali and alkaline earth metals (sometimes Fe+2, etc.). Aluminosilicate frame elements are enclosed in square brackets "; $x \in (0;1)$ – degree of aluminization of the silicate framework, where \in – membership index within the range TZTCPT $x = Al / (Al + Si) (0;1) = 0.22$.

$$\zeta = Na_{3,71}K_{0,88}Ca_{1,89}Mg_{0,61}[(Al_{7,88}Fe_{0,55})(Si_{28,02}Ti_{0,11})O_{72}] \cdot 20,27 = 1,855Na_2O \cdot 0,44K_2O \cdot 189CaO \cdot 0,61 MgO \cdot 3,94Al_2O_3 \cdot 0,275Fe_2O_3 \cdot 28,02\beta SiO_2 \cdot 0,11TiO_2 \cdot 1,725 \cdot O_{(g)} \cdot 20,27H_2O (i) \quad (2)$$

The determination of the THD function parameters of the TZCPT is problematic due to their experimental complexity, therefore the enthalpy of the TZCPT $\Delta_f H^0$, the Gibbs free energy $\Delta_f G^0$, the entropy S_{298}^0 , as in (Skhvitaridze et al., 2024), were determined by an approximate method: according to the principle of structural analogy of the constituent compounds and summing up the values:

$$\Delta_f TC(\zeta) = \sum_i y_i t_i, \quad (3)$$

where TC of any THD (H, G, S) is constant; ζ is the stoichiometric composition; i is the number of compounds in the zeolite; y_i is the component of the chemical compound in the zeolite; t_i is the constant:

$$\Delta_f C_{Pi} = (\Delta a_i + \Delta b_i + \Delta c_i). \quad (4)$$

The molecular mass of TZCPT was calculated using empirical stoichiometry, formulas (2) and (4) and the data in Table 1:

$$M_{TZCPT} = \sum_i y_i m_i. \quad (5)$$

In the formula (Skhvitaridze et al., 2024), m_i is the molecular mass of the chemical compound, $M_{TZCPT} = 2819.08 \text{ g mol}^{-1}$; This mass of 20.27 water molecules – $M_{H_2O} = 365.27 \text{ g mol}^{-1}$.

During dehydration, voids are formed in the TZCPT, the total volume of which is equal to the water content:

$$V_{TZCPT} = MH_2O / \rho_{H_2O} = 365.27 / 0.99705 = 366.35 \text{ cm}^3. \quad (6)$$

The density of CO_2 under normal conditions at $25^\circ C$ (298 K) = 1.8083 g/cm^3 , the volume of 1 mole of CO_2 will be: $V_{CO_2} = M / P = 44.01 / 1.8089 = 24.34 \text{ cm}^3 / \text{mol}$. Accordingly, the voids formed in the TZCPT can absorb: $V_{TZCPT} / V_{CO_2} = 366.35 / 24.34 = 15.01 \text{ mol } CO_2$. The density of $Ca(OH)_2$ under p normal conditions ($25^\circ C$) = 2.211 g/cm^3 . Then: $V_m = 74.093 \text{ g mol}^{-1} / 2.211 \text{ g/cm}^3 = 33.51108 \text{ cm}^3 \text{ mol}^{-1}$; In the void of 366.35 cm^3 volume, formed during dehydration in Tezami zeolite, not water is (absorbed), but an "aqueous solution of lime": $366.35 \text{ cm}^3 / 33.51108 \text{ cm}^3 = 10.93 = 11.0 \text{ mol } Ca(OH)_2$.

The enthalpy of formation of TZCPTnat, $\Delta_f H_{298}^0$ was calculated using formula (2) and the data from (Skhvitaridze et al., 2024):

$$\Delta_f H_{298}^0 TZCPTnat = -40740.454 \text{ kJ} \cdot \text{mol}^{-1}. \quad (7)$$

Gibbs free energy of formation of TZCPTnat. $\Delta_f G_{298}^0$ TZCPTnat under standard conditions is calculated using formula (4) and the data in Table 1:

$$\Delta_f G_{298}^0 TZCPTnat = -37643.783 \text{ kJ} \cdot \text{mol}^{-1}. \quad (8)$$

The entropy of TZCPT. From the Gibbs-Helmholtz equation $\Delta_f G_{298}^0 = \Delta_f H_{298}^0 - T\Delta S_{298}^0$ obtained entropy value is: $T\Delta S_{298}^0 = \Delta_f H_{298}^0 - \Delta_f G_{298}^0 = -3096.671 \text{ kJ} \cdot \text{mol}^{-1}$.

The quantities calculated above in TZCPTdeh will be as follows:

$$\Delta_f H_{298}^0 TZCPT_{deh} = \Delta_f H_{298}^0 TZCPTnat - 20.27 \Delta_f H_{298}^0 H_2O_L = -34946.68 \text{ kJ mol}^{-1} - 2199858 \text{ J} / \text{mol}.$$

$\Delta_f G^0 T = f(T)$ from the Gibbs-Helmholts equation to obtain the physicochemical values at any desired temperature it is necessary to integrate it:

$$\Delta_f G^0 T = \Delta_f H_f^0 - \Delta a T \ln T - 1/2 \Delta b T^2 - 1/2 \Delta c T^{-1} + y T . \quad (9)$$

In the range of 25-100°C (298-373 K) of heating of TZCPT, dehydration will begin, i.e.: TZCPTnat – 20.27 H₂O. In fact, the right part of equation (2) represents zeolite water release. The temperature of the onset of dehydration of the TZCPT (Fig. 2) and the pressure were determined using the calculation method as follows:

$$\Delta_f G^0 T = -707738 - 596.78 \cdot T \cdot \ln T - 107.675 \cdot 10^{-3} \cdot T^2 - 5.16 \cdot 10^5 \cdot T^{-1} + 1645,1011 T , \quad (10)$$

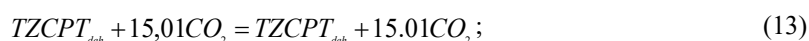
$$\Delta_f G^0_{298} = +2199858 J / mol ; \Delta G^0_{f, 368} = -352 J / mol ; \Delta_f G^0_{367.85} = 0 , \quad (11)$$

$$\Delta_f G^0_{367.85} = 0 ; T = 367.85(K) = 94.7^\circ C , \quad (12)$$

$$LgK = -\Delta_f G^0 T / 4.576 = -0.2185 ; K = 0.6046 atm ; P = 459.5 mm Hg , Torr .$$

The possibility of physical sorption in the dispersion system "TZCPT– CO₂ was evaluated.

Physical sorption of gaseous CO₂ in solid, fragmented TZCPT can be conditionally carried out according to the following equation:



CO₂ is physically sorbed (and modified) TZCPT- CO₂PHSOR :

$$\text{Enthalpy } \Delta_f G^0_{298 TZCPTdeh} + 15,01 \Delta_f H^0_{298} CO_2 = -40853.28 kJmol^{-1} , \quad (14)$$

$$\text{Gibbs free energy- } \Delta_f G^0_{298 TZCPTdeh} + 15,01 \Delta_f G^0_{298} CO_2 = -38756.25 kJmol^{-1} , \quad (15)$$

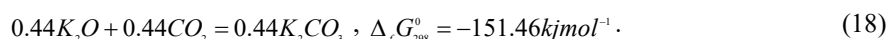
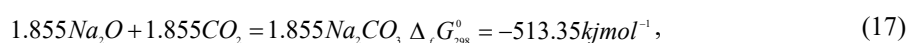
$$\Delta_f G^0_T = 961406 + 171.885 T \ln T - 33.851510^{-3} T^2 - 32.05510^5 T^{-1} - 3577.004 T , \quad (16)$$

$$\Delta_f G^0_{370.29} = 0 ; T = 370.29 K = 97.15^\circ C ; P = 293 mmHg , Torr .$$

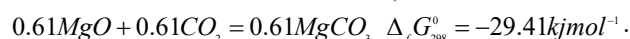
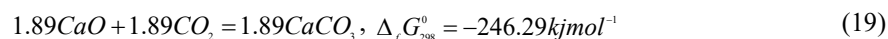
Possibility of chemical sorption of CO₂ in the dispersion system "TZCPT – CO₂".

Studies on CCUS processes have confirmed the sorption of CO₂ molecules on the surface of TZCPT, as well as in the structural and anhydrous pores of its framework (Skhvitaridze et al., 2024). TZCPT contains alkali (Na⁺, K⁺) and alkaline earth (Ca²⁺, Mg²⁺) metals, which are likely to undergo carbonization through chemical sorption forming carbonates:

Alkaline oxides:



Alkaline earth oxides



According to the report, 4.785 mol CO₂ can be chemically sorbed from the 15.01 mol CO₂ physically sorbed in TZCPT. The corresponding activities are as follows: Na⁺ > Ca²⁺ > K⁺ > Mg²⁺.

Chemical sorption of CO₂ in the TZCPT can occur according to the following conditional equation:



Using formula (2), we calculated the chemisorbed (modified) ZTCPT- CO₂CHEMSOR :

$$\text{Enthalpy} - \Delta_f H^0_{298 TZCPTdeh} + 4.785 CO_2 = \Delta_f H^0_{298 TZCPTchem.mod} = -33682.9625 kJmol^{-1} \quad (21)$$

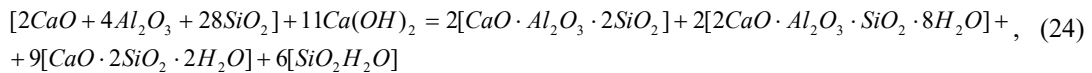
$$\text{Gibbs free energy} - \Delta_f G^0_{298 TZCPTdeh} + 4.785 CO_2 = \Delta_f G^0_{298 TZCPTchem.mod} = -34723.853 kJmol^{-1} \quad (22)$$

$$\Delta_f G_r^0: \Delta_f G_r^0 = 837684 - 343.4 \cdot T \ln T - 78.3 \cdot 10^{-3} T^{-1} - 22.7 \cdot 10^5 T^{-1} - 168.4 \cdot T, \quad (23)$$

$$\Delta_f G_{372.5}^0 = +720 \text{ kJ mol}^{-1}; \Delta_f G^0 372.8 = 0; 99.65^\circ\text{C}; K = 0.929 \text{ atm} = 706 \text{ mm Hg, Torr}.$$

THD forecast of possibilities for reducing CO₂ emissions into the environment in cement production using TZCPT is highly relevant. Therefore, it is important to study the THD abilities of calcium hydrosilicates/aluminosilicates by rehydrosintering in the reaction of TZCPT with CH during hydrate hardening of TZCPT-containing cement using approximate methods.

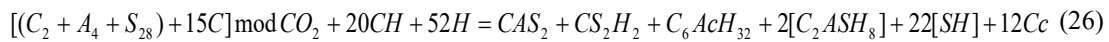
To simplify the calculations of the THD interactions with TZCPT and CH, we identified, as a first approximation, the primary compounds reactive with CH in the composition of TZCPT, as a result of which the values given in formulas (1) and (2) were modified as follows: $TZCPT_{\text{mod}} = 2CaO + 4Al_2O_3 + 28SiO_2 \cdot$



with abbreviated designations of compounds obtained in cement chemistry



$$\Delta_f G_{298}^0 = -8717.00 \text{ kJ / mol}.$$



$$\Delta_f G_{298}^0 = -870.00 \text{ kJ / mol}.$$

Results

Determination of the thermodynamic function parameters of natural and dehydrated TZCPT, as well as the ability to carbonize with CO₂ and pozzolanize with rehydrogenation with CH, allows planning and implementing a reduction in the footprint of 890 kg of CO₂ emitted into the atmosphere by producing 1 ton of cement clinker.

According to TZCPT_{nat}, CEM II/B-Q,32.5 cement (grade 32.5) can be added as an additive in an amount of ≤ 35 mass %, and the mechanical strength when tested according to standard procedures, the resulting mechanical strength should be (≤ 32.5 MPa). As a result of pozzolanic, rehydrogenation CO₂ – footprint decreases from 890 kg to 578.5 kg, i.e. by 311.5 kg = 35mass %;

The calculations indicate that in TZCPT, during the (ad) sorptive capture of CO₂ from flue gases, the CO₂ capture – regeneration cycle is repeatable. In this process, 15.01 mol of CO₂ is physically (ad)sorbed-captured in the void of 366.35 cm³ created by the evaporation of H₂O. The process begins at $t = 97.15^\circ\text{C}$ and $P = 293$ mm Hg Torr. 4.785 mol of the captured 15.01 mol of CO₂ are chemisorbed in TZCPT with alkali/alkaline earth metals at $t = 99.65^\circ\text{C}$ and $P = 706$ mm Hg Torr with the formation of carbonates.

By mixing TZCPT_{modCO₂} into CEM II/B-Q,32.5 type cement while maintaining strength (≤ 32.5 Mpa), 1 mol TZCPT ($M = 2819.08$ g/mol) in the CCUS process (ad)sorptionally captures 15.01 mol (660.59 g) CO₂ and is modified into TZCPT_{modCO₂}.

The study confirms that by mixing TZCPT_{modCO₂} into CEM II/B-Q,32.5 type cement at a rate of 35 mass % instead of 45 mass % and maintaining mechanical strength, out of 890 kg of technological CO₂ emitted into the atmosphere, "bound – captured" will be: $(890 \times 0.45) \text{ kg} + 105.5 \text{ kg} = 506 \text{ kg CO}_2$, which reaches 57 mass % of the emitted CO₂ and is an innovative achievement.

Conclusion

The dehydration of TZCPT, its carbonation and the rehydrocarbonation of cement for pozzolanic and carbonate modification, the formation of crystalline hydrates during hydration with cement minerals, demonstrate the capabilities of TZCPT. These include: transition from a hydrated to a dehydrated state; capture of CO₂ generated during production through physicochemical sorption; rehydrogenate, pozzolanic, and carbonate modification of cement; and reduction of the carbon footprint, thereby contributing to the greening of cement technology.

The calculated THD function value for TZCPT, $\Delta_f H_{0298} = -40740.45 \text{ kJ mol}^{-1}$, is close to the previously reported values, $\Delta_f H_{0298} = \{ [-40232 - (-40452)] - [-41290 - (-41527)] \} \text{ kJ mol}^{-1}$, whereas with CPT-TF, studied by Skhvitaridze et al., (2024): $\Delta_f H_{0298} = -39825.84$, the difference is $626.61 \text{ kJ mol}^{-1} = 1.57\%$.

In the TZCPT – CO₂ dispersion system in CCUS processes, the onset of TZCPT dehydration is expected at 94.7°C and a pressure of 459 mmHg Torr, but the temperature is lower than required for CPT (94.85°C), and the pressure is higher (745 mmHg Torr).

In the TZCPT – CO₂ dispersion system during CCUS processes, physical sorption of CO₂ from flue gases in TZCPT will occur at 97.15°C and a pressure of $P = 293 \text{ mmHg Torr}$ (higher than for CPT at 96.35°C, and a pressure lower than for CPT at 636 mmHg Torr (Skhvitaridze et al., 2024)). Chemisorption occurs at 99.65°C and a pressure of $P = 706 \text{ mmHg Torr}$ (higher than for CPT at 95°C), and a pressure of $P = 706 \text{ mmHg Torr}$, which is higher than the CPT pressure of 534 mmHg Torr). These results confirm that TZCPT exhibits excellent thermodynamic and sorption properties for CO₂ capture and pozzolanic activity, supporting its potential use in environmentally optimized cement production.

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თემძის კლინოფთილოლითის თერმოდინამიკური ფუნქციები, დეჰიდრატაციისა და CO₂ სორბციით კარბონიზირების, ცემენტის რეჰიდრო-გაკირებითი პუცოლანიზაციის პარამეტრები ჯერ კიდევ არ არის შესწავლილი, რაც აფერხებს მის გამოყენებას ისეთ პროექტებში, რომელთა მიზანია ცემენტის დამზადებისას ატმოსფეროში ემისირებული CO₂-ის რაოდენობისა და კვალის შემცირება. კვლევა ითვალისწინებდა თემძის ნედლი და დეჰიდრატირებული კლინოფთილოლითის თერმოდინამიკურ ფუნქცია-პარამეტრთა და ცემენტის გამყარებისას კალციუმის ჰიდროალუმოსილიკატთა წარმოქმნის შესაძლებლობათა პროგნოზირებას ანგარიშის მიახლოებითი მეთოდებით. კვლევების შედეგად თემძის კლინოფთილოლითის უნარებია: წყლიანი მდგომარეობიდან უწყლოში გადასვლა; ფიზიკურ-ქიმიური სორბციით ტექნოლოგიურ პროცესში წარმოქმნილი CO₂-ის შთანთქმა; ცემენტის რეჰიდრო-გაკირებითი პუცოლანიზაცია და კარბონიზაცია; CO₂-ის კვალის შემცირება; ცემენტის ტექნოლოგიის ეკოლოგიზაციით გარემოს დაცვითი პრობლემების ხელშეწყობა.

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