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Low-Temperature Aluminosilicate Nanostructured Binder. Characteristics and Applicability

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Abstract. Current trends in the field of construction material is focused on enhancement of sustainability of building materials and constructions urging on development of new types of inorganic binders and composites in order to meet the modern requirements of service performance and special properties. This research studied and demonstrated the opportunity to develop zero-cement heat-resisting granite-based nanostructured binder (GNB) using «green» technology production. XRD and DTA analyses demonstrated that the thermal exposure of GNB to wide range of temperatures of 20–1000 °C leads to such phase transformations in the binder as α -quartz to β -quartz transformation; amorphous alkali-aluminosilicate (gel) to crystal phase of Ca-albite. The calculation of cell volumes characteristics for low-temperature (before thermal exposure) and high-temperature (after thermal exposure) phases was performed using following equation: $V_{specific} = \sum_i C_i \cdot V_i^{un.cell}$ where C_i is concentration (by wt. %) of mineral phases; V_i is unit cell volume of mineral phases, Å. The calculated ratios of unit cell volumes were close to 1 which ensures a structural stability of the GNB under thermal exposure and confirms its heat-resistant performance.

Introduction

Development of low-temperature technologies for synthesis zero-cement silicate and aluminosilicate binders is one of the promising research areas in the field of construction material science [1–5]. In this paper low-temperature technology introduced as the technologies, that can be realized without high-temperature treatment of raw materials required for the binder synthesis.

There is a hypothesis that binders synthesized using low-temperature technology have a non-hydration mechanism of structure formation. For instance, zero-cement silicate and aluminosilicate nanostructured binders (NB) hardens by geopolymerization mechanism where the key role of structure formation is accounted for the nano-sized component of the binder system.

The production technology of NB is mainly consists of mechanochemical synthesis in water medium, that ensures its sustainability and environmental safety. In addition, the production technology of NB allows application wide range of SiO₂-bearing raw materials therefore, makes it possible to be adapted in different regions.

Earlier studies [6–9] reported on the possibility of NB synthesis using different SiO₂-bearing materials such as quartz sand, sandstone, quartzite, perlite, zeolitized tuff etc.

The objectives of this research was to study geopolymerization mechanism of NB as well as heat-resistant performance typical for this group of binders.

Materials and Methods

Granite from Poltava deposit (Ukraine) was used as SiO₂-bearing raw component. Chemical composition of granite shown in Table 1.

Table 1. Chemical composition of granite

Oxide composition [%]									
SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	SO ₃	K ₂ O	Na ₂ O	LOI	Σ
69.2	15.7	3.35	3.14	0.89	0.09	2.06	4.29	0.75	99.47

Mineral composition of the GNB was analyzed with XRD using Database PDF-2 provided by the software Crystallographica SearchMatch v 2,0,2,0 (Oxford Cryosystems). XRD patterns for the experimental samples were prepared with diffractometer STOE STADI (STOE & Cie GmbH, Germany) using CuK α .

Quantitative XRD-analysis focused on determination of mineral crystal phases (excluding X-ray amorphous constituent) was accomplished with Full Profile XRD using DDM v.1.95d in algorithm of Derivative Difference Minimization [10].

XRD results confirmed the presence of rock-forming minerals such as α -quartz, Ca-albite and biotite in GNB. In addition, a considerable portion of corundum was detected in the system of GNB in the result of mechano-activation. Data of quantitative XRD-analysis for crystal phases in GNB are presented in Figure 1.

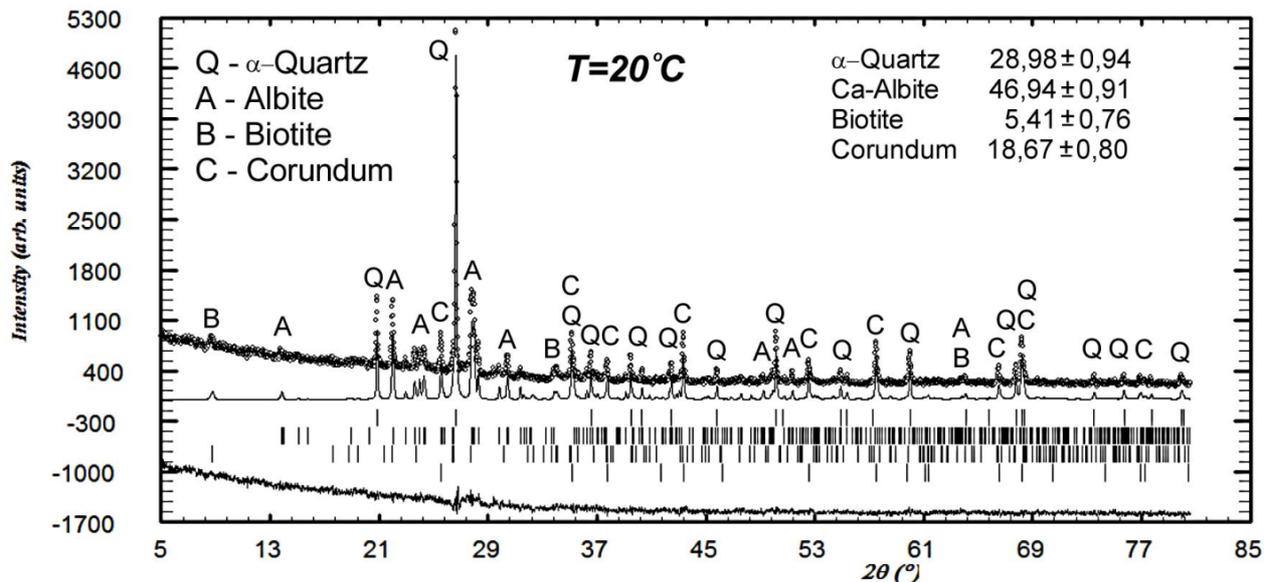


Figure 1. The XRD pattern of NB at 20 °C, wt.%

Experimental Part

The evidence of geopolymerization mechanism of structure formation in GNB based on the hypothesis of two-phase composition of the binder that is crystal and amorphous. Amorphous phase is an alkali-aluminosilicate gel which forms when dissolution of both crystal granite constituents quartz and feldspar under mechano-chemical impact. It is assumed that alkali-aluminosilicate gel has zeolite-like structure, which upon crystallization will form zeolite minerals. The aforementioned process would serve as a confirmation of geopolymerization mechanism of structure formation in GNB.

In order to indicate the amorphous-to-crystal transformation thermal DTA-analysis of GNB under the temperature range of 20–1000 °C (Fig. 2) using STA 449 F1 Jupiter® machine (NETZSCH, Germany) was performed.

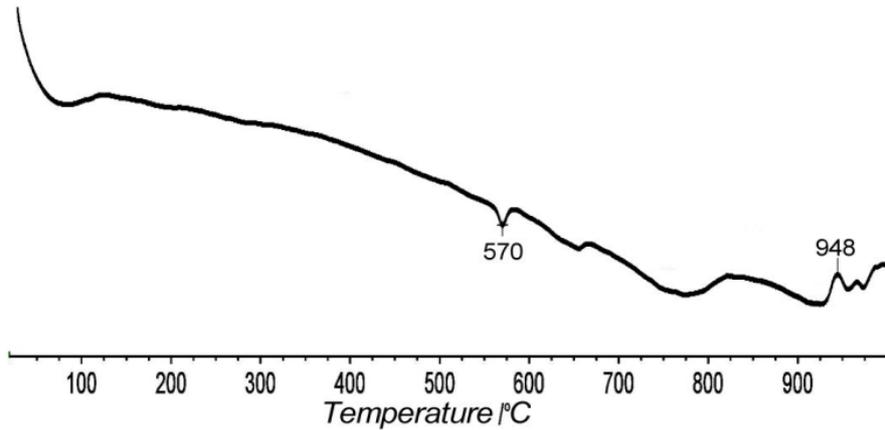


Figure 2. DTA-thermogram of GNB

Thermogram (Fig. 2) demonstrated endo-effect at 570 °C that associated with α - β polymorph phase transformation as well as exo-effect at 948 °C. To study the origin of the detected exo-effect in GNB the high-temperature XRD-analysis at 600, 900 and 1000°C with 1-hours curing at each temperature was performed. High-temperature XRD-analysis was accomplished using high-temperature device (Stoe) that allows XRD investigation within the temperature range of 20–1500 °C.

The obtained high-temperature XRD-patterns for GNB (Fig. 3) demonstrated spurious reflections from Pt–Rh heating element which were located next to the GNB pattern during the test. Even though these reflections were not part of the test object, in order to provide a higher quality of XRD-analysis, this data was included in the calculation of crystal components concentrations (by wt. %) in GNB.

Mineral composition of GNB at 600°C and 900°C (Fig. 3, a, b) appeared to be the same as at 20 °C (Fig. 1) with exception of quartz transformation at higher temperatures. However, the X-ray spectrum of GNB obtained at 1000 °C detected the presence of a new phase of Leucite ($K[AlSi_2O_6]$) that is analogue of Analcime ($Na[AlSi_2O_6] \cdot H_2O$), one of the types of zeolite. Thus, the appearance of Leucite explains the exo-effect at 948 °C (Fig. 2) and confirms the geopolymerization mechanism of GNB structure formation due to crystallization of zeolite-like phase (Leucite) from alkali-aluminosilicate amorphous gel.

Generally, thermal resistance is the resistance of structure to fail under a high-temperature exposure. One of the key parameter of thermal resistivity is a constancy of unit cells volume of crystal under different thermal conditions. In this study, the unit cells volumes for all crystal phases of the GNB presented in Table 3 were calculated using equation (1):

$$V_{Specific} = \sum_i C_i \cdot V_i^{Un.cell}$$

where C_i is concentration (by wt. %) of mineral phases;

V_i is unit cell volume of mineral phases, \AA^3 .

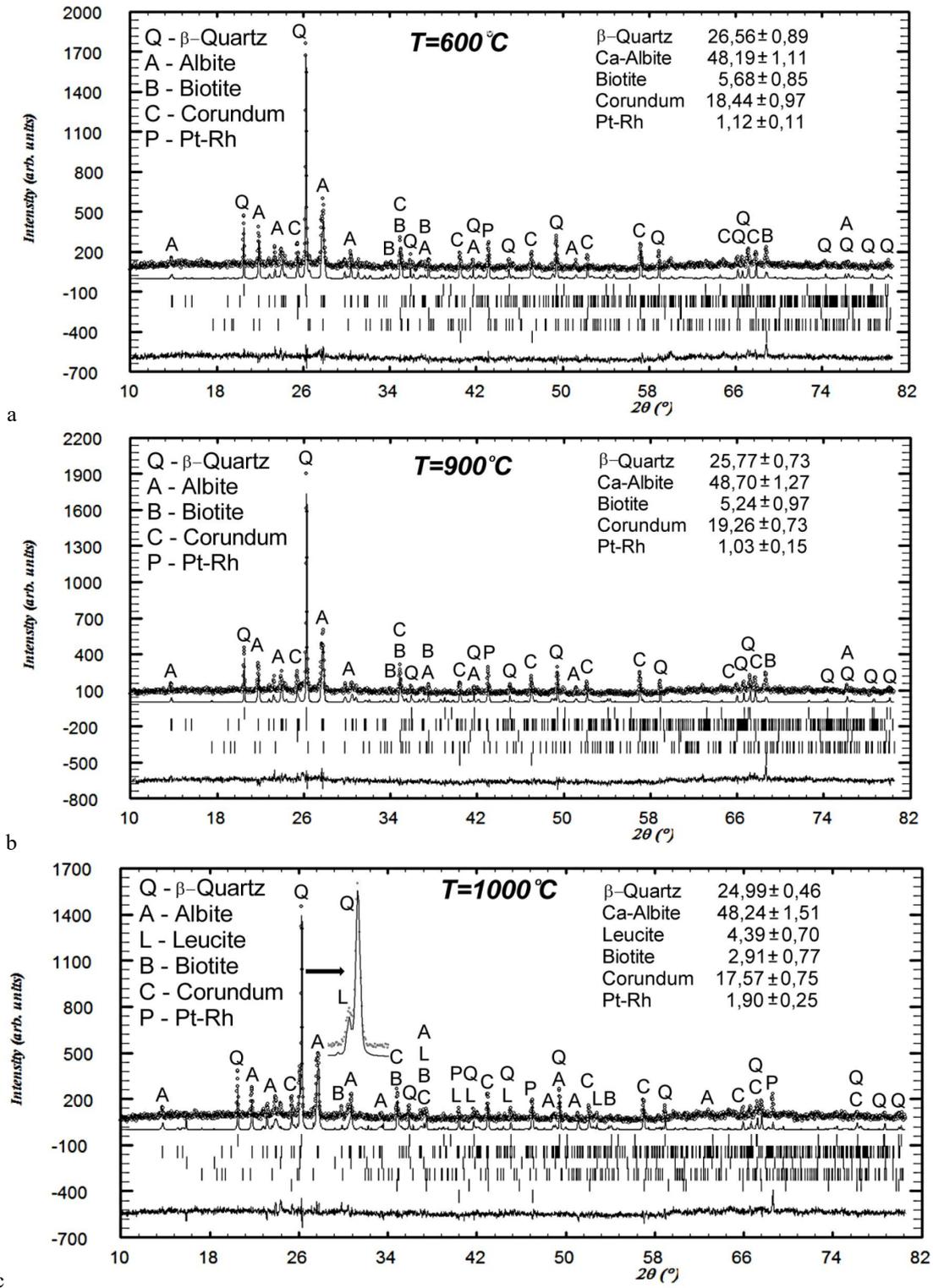


Figure 3. XRD-patterns of GNB after thermal treatment, (wt. %) a) 600 °C; b) 900 °C; c) 1000 °C

Table 2. Mineral composition of GNB, wt.%

T°C	Quartz	Albite	Biotite	Corundum	Leucite
20	28.98 ± 0.94	46.94 ± 0.91	5.41 ± 0.94	18.67 ± 0.8	–
600	26.86 ± 0.89	48.74 ± 1.11	5.74 ± 0.85	18.65 ± 0.97	–
900	26.04 ± 0.73	49.21 ± 1.27	5.29 ± 0.97	19.46 ± 0.73	–
1000	25.47 ± 0.46	49.17 ± 1.51	2.97 ± 0.77	17.91 ± 0.75	4.39 ± 0.70

The calculated data of the unit cells volumes for all crystal phases of GNB at 20, 600, 900 and 1000 °C are presented in Table 3.

Table 3. Volumes of unit cells $V_i^{Unit.cell}$, Å³

T°C	Quartz	Albite	Biotite	Corundum	Leucite	$V_{specific}$
20	112.759	665.41	516.2	254.52		420.466
600	117.923	674.13	515.6	258.09		437.967
900	117.835	680.5	521.8	260.06		443.770
1000	117.684	682.4	534.5	260.72	2535.9	539.421

It is worth to remark that an anomaly reduction of value was observed for quartz when temperature goes up. This phenomenon was manifested in earlier studies [11]. For the other phases temperature increase leads to monotonous increase of value up to 900 °C. However, at 1000 °C a value of total volume boosts dramatically due to formation of a new zeolite-like phase – Leucite. The ratio of values $V_{Specific}$ at high temperatures (600, 900, 1000 °C) and at 20 °C can be considered as a quantitative indicator of variation in the following equation:

$$\frac{V_{Specific}^{600^{\circ}C}}{V_{Specific}^{20^{\circ}C}} = 1.042; \quad \frac{V_{Specific}^{900^{\circ}C}}{V_{Specific}^{20^{\circ}C}} = 1.055 \quad \text{and} \quad \frac{V_{Specific}^{1000^{\circ}C}}{V_{Specific}^{20^{\circ}C}} = 1.283$$

Summary

The calculated ratios of unit cell volumes for mineral phases of GNB before and after thermal treatment up to 900 °C are close to 1 (with the deviation in the range of 5 %) which ensures a structural stability of the GNB under thermal exposure and confirms its heat-resistant performance. The more detailed study on heat resistance performance of GNB will be performed and presented in the future research work.

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