



Effect of metallic Si addition on polymerization degree of *in situ* foamed alkali-aluminosilicates

Valentina Medri^{a,*}, Elettra Papa^a, Jiri Dedecek^b, Hana Jirglova^b, Patricia Benito^c, Angelo Vaccari^c, Elena Landi^a

^aNational Research Council of Italy, Institute of Science and Technology for Ceramics (CNR-ISTEC), via Granarolo 64, 48018 Faenza, RA, Italy

^bJ. Heyrovsky Institute of Physical Chemistry ASCR, Dolejskova 2155/3, 18223 Prague 8, Czech Republic

^cDipartimento di Chimica Industriale "Toso Montanari", ALMA MATER STUDIORUM, Università di Bologna, Viale Risorgimento 4, 40136 Bologna, Italy

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Abstract

Geopolymerization is an aqueous based process to produce synthetic alkali-aluminosilicates with porosity that can be tailored from the nano- to the ultra-macrometric range. In order to fulfill the requirements for many different applications and porous 3D networks (namely alkali-aluminosilicate foams) were prepared by inducing interconnected ultra-macro-porosity (up to the millimeter range) in the alkali-bonded matrices, exploiting the ability of silicon powder to generate H₂ in the aqueous medium. Being H₂ the product of a water-consuming process which competes with geopolymerization, the process parameters and the characteristics of the resulting foamed materials are strongly influenced by the amount of added silicon. Polymerization degree, accessibility of the geopolymer inner volume and the micro-ultra-macro-structure of the foams were determined and related to the process parameters of the contemporary geopolymerization and foaming.

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1. Introduction

Porous ceramics have been extensively used because of their high specific surface area, permeability, sound absorption, thermal insulating and other properties [1]. Industrial uses include structural lightened parts, insulator panels, filters and membranes, radiant burners, gas or chemical sensors, support materials for catalysis or adsorbents.

For all the mentioned applications, it is absolutely necessary to control porosity, in terms of the following: pore size dimension and distribution; amount and pore structure (shape, morphology, orientation, and surface properties). For example, the required pore dimension for molecular sieves ranges from 1 Å to 1 nm; for sorbents from 1 Å to a few tenths of nanometers; for catalysts and supports from 1 nm to few tenth of microns; for filtering and purification systems from few nanometers to a few millimeters;

and for dust collecting from few micrometers to few millimeters [1,2]. A porosity amount over 50% and up to 95% is required in view of a lightweight device [3] and thermal insulation [2]; porous ceramics with three-dimensionally interconnected and distributed open pores (3D structures) are useful as catalysts, catalyst supports, filters, and scaffolds, adsorbents, because of the high accessibility of pores [1,2,4].

Specific porous materials, such as zeolites, meso-porous silica, gels, ceramic membranes, ceramic foams, etc., are used to fulfill specific porosity requirements in view of specific applications [1,2].

Synthetic alkali-aluminosilicates, namely alkali-bonded ceramics or geopolymers [5,6], were produced by reacting an aluminosilicate powder with an aqueous alkali hydroxide and/or alkali silicate solution. They could be used to develop porous ceramic materials and components which cover the pore size range from a few tenths of nanometers to few millimeters and total porosity amount from 30% up to 90%. They are amorphous to semi-crystalline materials with three-dimensional aluminosilicate network, formed by condensation polymerization. The final microstructure of a fully

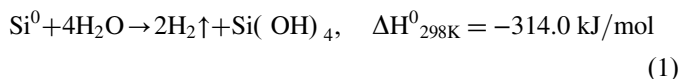
*Corresponding author. Tel.: +39 0546699751; fax: +39 054646381.

E-mail address: valentina.medri@istec.cnr.it (V. Medri).

reacted geopolymer matrix consists of nano-particulates ranging from 5 to 15 nm, separated by pores in the nanometric range [7,8]. On the atomic scale, geopolymer amorphous network is formed by SiO_4 and AlO_4 tetrahedra shearing oxygen corners. Recent results indicate that these tetrahedrons form in the network rings of various sizes analogous to those in zeolites. The formation of geopolymer network from Si–O–Al–O rings results, in analogy with zeolites, in ion exchange properties of geopolymer matrix [9,10]. Thus, geopolymers can be regarded as the amorphous counterpart or precursor of crystalline zeolites.

Chemical consolidation by geopolymerization and contemporary *in situ* inorganic foaming are suitable to produce ceramic foams with 3D porous architectures without using high temperature treatments (such as burnout of organics and sintering) that are indeed necessary for the production of ultramacroporous ceramics using conventional techniques [11].

In detail, *in situ* inorganic foaming can be obtained causing gas evolution in the geopolymer mixture [12–16]. The Pourbaix diagram of silicon indicates that in alkaline conditions hydrogen evolution is always favoured [17], following the reaction:



In a previous study [8] optimal geopolymerization conditions were determined to develop porous 3D networks by inducing interconnected ultra-macro-porosity (up to the millimeter range) in selected alkali-aluminosilicate material, using low amount (≤ 0.03 wt%) of metallic silicon powder as a foaming agent. However, the solutions for specific application requirements must be tailored by a correct mixture and process design to optimize properties and possibly reduce costs. This work reports on several aspects of the intrinsic structures and properties of inorganic polymers, as well as on some of the extrinsic properties that may be conferred by an appropriate raw material selection, mixture and process design.

In particular, aim of this study is to check the effect of increasing Si^0 additions (≥ 0.03 wt%) on *in situ* foamed alkali-aluminosilicates to correlate the effect of heat release,

occurring during the redox reaction (1), to the degree of polymerization (namely the conversion of aluminosilicate raw powder into geopolymer) and thus to the final micro- and macrostructure of the obtained foams. For this purpose, the developed alkali-aluminosilicate foams were fully characterized in term of microstructure, intrinsic and induced porosity size distribution, specific surface area, polymerization degree and ion exchange capacity.

2. Experimental procedure

2.1. Preparation of geopolymer foams

Metakaolin grade M1200S was purchased from AGS Minéraux, Clérac, France. For more details on powder characteristics see Refs. [10,18]. Potassium silicate solutions with molar ratios of $\text{SiO}_2/\text{K}_2\text{O}=2$ and $\text{H}_2\text{O}/\text{K}_2\text{O}=13.5$ and 23.0 were prepared by dissolving KOH pellets (purity > 98%, Merck, Darmstadt, Germany) into distilled water and adding fumed silica powder (99.8%, Sigma-Aldrich, Steinheim, Germany) under magnetic stirring. Metallic silicon powder (grade AX10, H.C. Stark, purity 99.995%, $D_{50}=4.50 \mu\text{m}$) was used as a foaming agent.

Geopolymer slurries with Si/Al and K/Si atomic ratios, respectively, equal to 2.00 and 2.33 were prepared by mechanically mixing the metakaolin with the $\text{KOH}/\text{K}_2\text{SiO}_3$ aqueous solutions for 19 min at 100 r.p.m. for clarity, and correct if necessary. \rightarrow . Foams were prepared by adding from 0.03 to 2.60 wt% of Si powder into the geopolymer slurries. The Si/Al and K/Si nominal atomic ratios of the foam formulations were consequently higher than those of the starting geopolymeric slurries; values ranging from 2.01 to 2.36 for Si/Al and from 2.33 to 2.75 for K/Si were experimented.

Then mechanical mixing was performed for another minute after metallic silicon additions.

The slurries (20 ml) were cast in plastic cylindrical open molds with a diameter of 40 mm and matured at room temperature (RT), 40 °C, 60 °C or 80 °C for 24 h. The foam expansion mainly occurred in the axial direction of the cylindrical mold. After curing,

Table 1
Si/Al and Si/K atomic ratios in alkali-aluminosilicate foams compositions, polymerization degree obtained from ^{27}Al MAS NMR and NH_4^+ exchange capacity, namely accessibility of the geopolymer inner volume.

Sample	Si/Al atomic ratio	Si/K atomic ratio	Polymerization degree (%) ^a	Accessibility (%) ^b
F13-0.38%Si-RT	2.05	2.39	56	25
F13-0.38%Si-80°C	2.05	2.39	48	23
F13-1.15%Si-RT	2.14	2.49	56	18
F13-1.15%Si-40°C	2.14	2.49	53	23
F13-1.15%Si-60°C	2.14	2.49	39	16
F13-1.15%Si-80°C	2.14	2.49	51	15
F13-1.90%Si-RT	2.23	2.60	52	16
F13-1.90%Si-40°C	2.23	2.60	42	11
F13-1.90%Si-60°C	2.23	2.60	30	17
F13-1.90%Si-80°C	2.23	2.60	50	16
F13-0.04%Si-RT	2.01	2.33	97	27
F23-0.03%Si-RT	2.01	2.33	97	26

^aThe data errors for polymerization degree is $\pm 5\%$ and $\pm 1\%$ for polymerization degrees below and above 70%, respectively.

^bThe data error for accessibility of geopolymer network is $\pm 5\%$.

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