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White sintered glass-ceramic tiles with improved thermal insulation properties for building applications

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ABSTRACT

In order to provide a thermal barrier in an arid environment, highly reflective coatings were deposited on porous substrates made of natural raw materials from Saudi Arabia. Although highly reflective coatings inhibit heat absorption from the incoming sunlight, the body of conventional ceramic tiles warms up to environmental temperature through conduction, convection and radiation. A strategy to reduce the penetration of this heat into the building is to use a highly porous substrate, which reduces the thermal conductivity of the tile, coupled with a highly reflective glaze. The approach leads to the concept of "cool" tiles, aimed at improving the thermal efficiency of buildings. The present paper provides a first example, based on layered wollastonite-hardystonite glass-ceramics developed by double pressing of glass powders and additives followed by sinter-crystallization.

1. Introduction

The improvement of the thermal efficiency of buildings, especially when exposed to arid environments, represents a challenge concerning the reduction of the overall costs for cooling. The so-called 'cool roofing' involves strategies to increase the solar reflectance in the visible and near infra-red wavelengths and thermal emittance at wavelengths close to 10 μm of roofs in urban areas, thus reducing the absorbance of thermal radiation and decreasing the demand on cooling power. Cool roofs also mitigate summer urban heat islands, lowering the citywide ambient air temperature and increasing human comfort [1–3].

A valuable example of cool roofing solution is represented by functional engobes with a high albedo (i.e. exhibiting a high diffuse reflectivity or reflecting power), as developed by Ferrarin et al. [4], that can be applied to conventional ceramic tiles used for roofing. Studies performed by Synnefa et al. demonstrated that the use of reflective coatings can reduce the temperature of a white concrete tile surface under hot summer conditions by 4 °C during the day, and by 2 °C during the night [5]. The surface of the concrete tiles was warmer than the ambient air by only 2 °C during the day, and cooler than the ambient air by 5.9 °C on average during the night.

Although a coating with a high albedo is valuable, a ceramic tile may warm up to environmental temperature, by contact with the air through conduction, transferring the heat into the building. A strategy to reduce this issue is the use of a porous substrate that reduces the thermal conductivity. Thus the combination of a high reflective glaze and a porous substrate, as shown in Fig. 1, may represent a suitable strategy for obtaining "cool" tiles with improved thermal management properties [6].

The porosity of the substrate needs to be tailored in order to provide both sufficient mechanical strength and low thermal conductivity. The matching of the coefficients of thermal expansion between glaze and substrate is simpler when the chemical composition of the two layers is similar, and therefore in this work we considered glass compositions suitable for the production of both dense and porous sintered glass-ceramics. Dense tiles can be considered for paving applications whereas porous tiles, both glazed and unglazed, can be used for cladding.

Porous tiles with a density and water absorption below 2 g/cm^3 and 2 wt%, respectively, were specifically investigated thus obtaining a compromise between mechanical strength, low thermal conductivity and limited water absorption (a high water absorption would favor the formation of fungi and decrease the frost resistance). With the aim of producing white glass ceramics, natural

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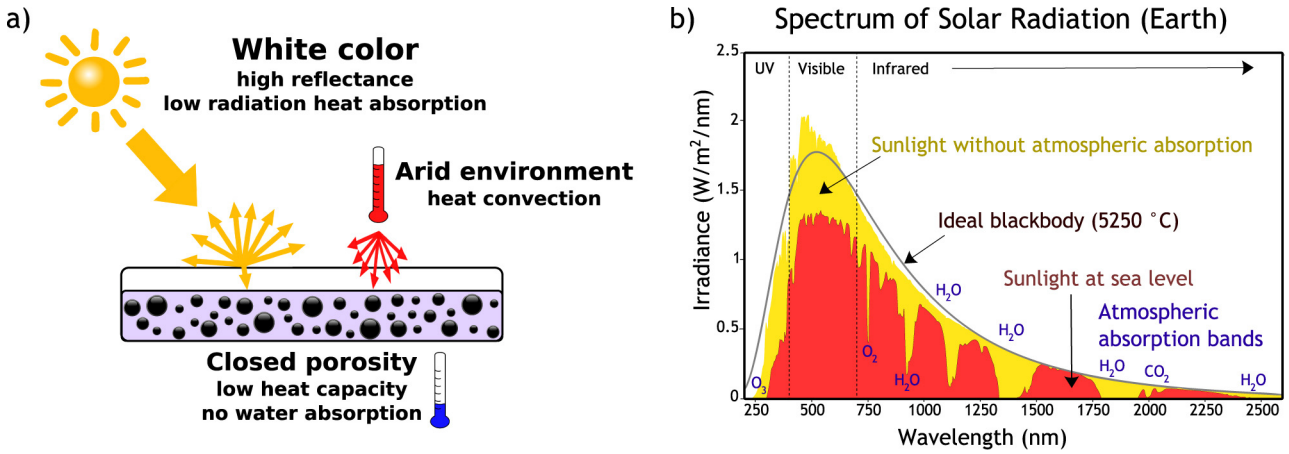

Fig. 1. (a) Schematic representation of an insulating tile engineered using a highly reflective coating deposited on a substrate possessing a low thermal conductivity. (b) Spectrum of the solar radiation on the earth.

Table 1
Chemical composition of NP glass and Neoparies™.

Oxide	NP glass		Neoparies™	
	(wt%)	(mol%)	(wt%)	(mol%)
Al_2O_3	7.7	4.8	7.0	4.4
B_2O_3	1.3	1.2	1.0	0.9
BaO	0.0	0.0	4.0	1.7
CaO	20.3	22.8	17.1	19.6
Fe_2O_3	0.4	0.1	0.0	0.0
K_2O	2.5	1.7	2.0	1.4
MgO	0.7	1.1	0.0	0.0
Na_2O	2.5	2.5	3.0	3.1
SiO_2	57.4	60.1	59.3	63.6
TiO_2	0.7	0.6	0.0	0.0
ZnO	6.5	5.0	6.5	5.2

	Clay (wt%)	Cullet (wt%)	Limestone (wt%)	Silica (wt%)	Pure chemicals (wt%)
Amount in NP glass	14.8	9.9	29.6	34.5	11.4

raw materials possessing a limited amount of impurities (i.e. transition metals) were selected. Whiteness and opacity are provided by the refractive index difference between the glassy matrix and the crystal phases.

2. Experimental procedure

Neoparies™ glass-ceramics [7] were chosen as a reference material. A glass with chemical composition resembling that of Neoparies was produced using natural raw materials from Saudi Arabia and a limited amount of pure chemicals (~11 wt%, in the form of 3 wt% ZnO, 3% borax and 5.4% K_2CO_3), as reported in Table 1.

The chemical composition of the raw materials was evaluated by means of X-ray fluorescence (Philips XRF sequential spectrometer PW 2400, Eindhoven, The Netherlands). Compared to Neoparies, BaO was substituted by increasing the mol.% of CaO, while the silica content was slightly reduced.

The raw materials were first dried and homogenized by ball milling in an agate jar for 30 min at 300 rpm and finally melted in kyanite (Al_2SiO_5) refractory crucibles at 1400 °C for 90 min in static air. The molten glass did not corrode the crucible, so that the chemical composition of the glass was not altered. After achieving complete melting of the raw materials, the melt was poured into water to produce a glass frit. The drastic quenching provided a number of fragments that were successively dried at 80 °C overnight, ball milled (30 min at 400 rpm) and sieved to obtain particles with a size below 90 μm.

Dilatometric and differential thermal analysis (DTA/TGA, STA 409; Netzsch-Gerätebau GmbH, Selb, Germany, operated at 10 °C/min in static air) were performed both on powders below 90 μm and coarser particles (above 2–3 mm), to investigate the effect of the particle size on the crystallization behavior (surface induced vs bulk nucleation).

The investigation of the evolution of the density as a function of the amount of foaming agent added was performed by adding Si_3N_4 (samples labeled SN) and mixtures of Si_3N_4 and gypsum ($CaSO_4{\cdot}2H_2O$) used as oxidizer in a molar ratio Gypsym/Silicon Nitride of 3:1 (samples labeled G_3SN_1) or 6:1 (sample labeled G_6SN_1). For the preparation of dense samples, the pure frit was used whereas for the preparation of porous samples the foaming additives were introduced in the range 0.5 to 4 wt% with respect to the amount of dry frit, and the mixtures were cold pressed mixture in a 13 mm steel mold at a pressure of 40 MPa.

The water absorption, W_{AB}, apparent, ρ_a, bulk, ρ_b, densities of the fired samples were evaluated according to the UNI EN ISO10545 norm, by means of the Archimedes method. The true density, ρ_t, was evaluated on powdered samples of size below 90 μm by means of helium gas pycnometer (Micromeritics AccuPyc 1330, Norcross, GA).

After selection of optimum compositions, larger samples were realized using 25 g of the dry frit powder uniaxially cold pressed in a steel mold of 50 × 50 mm^2 at a pressure of 40 MPa. To obtain a porous substrate coated with the dense glass-ceramic, 3 g of the dry frit powder were lightly pressed (at 10 MPa) and then 22 g of the glass frit mixed with the selected foaming agent were further deposited. The layered sample was then uniaxially pressed at 40 MPa. The produced samples were then fired in air at 950 °C for 30 min applying a 10 °C/min heating rate.

The Young's modulus of the glass-ceramic samples was determined using the resonant frequency method in the flexural mode of vibration (GrindoSonic Mk5, Leuven, Belgium). Four-point bending tests (40 mm outer span and 20 mm inner span) were performed using an Instron 1121 UTS instrument (Instron, Danvers, MA) on at least 15 specimens for each sample type, with dimensions of 4 mm × 2.5 mm × 47 mm. In order to remove surface flaws, all samples were carefully polished to a 6 μm finish before testing, using abrasive papers and diamond paste. The edges of the bars were beveled using fine abrasive papers and diamond paste. The crosshead speed was 1 mm/min until fracture. The double layer samples were tested by positioning the porous layer on the compression (upper) side and the dense layer on the tensile (lower) side.

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