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Thermal insulating cementitious composite containing aerogel and phosphate-based binder

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Abstract. Thermal conductivity is an important material property in the energy design process of the buildings. While cement-based materials are the most consumed materials in the construction industry, thermal properties of these materials can play a significant role in energy efficiency of the buildings. Cementitious materials with low thermal conductivity can be desirable for using as a part of heat insulation or for thermal bridge calculations. They may be used in different forms such as cast concrete, on-site 3D printing or surface finishes. In this study, aerogel granules was used as aggregate and combination of phosphate based cement and fly ash was considered as binder in order to achieve a fast setting material with low thermal conductivity. Furthermore, the calcination process was eliminated for the phosphate cement, which led to a material with lower environmental impact. Thermal conductivity as low as 0.04 W/mK was measured using the proposed material composition.

1. Introduction

A building material which can be used in construction of the buildings with properties such as energy efficiency, sound insulation, fire resistance, light weight, versatility and quick production and installation has always been the dreaming material for architects and builders. Different types of concretes such as light weight aggregate concrete or aerated autoclaved concrete have been under development in the last years to obtain a more efficient material for this purpose.

Aerogel granules are used as aggregate in concrete to improve the material properties [1, 2]. Having a hydrophobed pore system, with extremely small pores in the range of 20 to 60 nanometers, aerogel particles have excellent insulation properties. The maximum size of these granules is about four millimetres. Although aerogel is the lightest solid material known at present, these particles are very strong towards uniform pressure and are even elastic. They can be squeezed together and will spring back after the pressure is released. Using these particles as granules in cementitious composites, applies sheer forces to them during the steering process. The particles easily break into smaller pieces under sheer forces. However, the smallest particles that can be generated by the “grinding process” are in the range of a couple of thousands of nanometers. Therefore, the smaller particles created after steering, will still have the same hydrophobic nano-pores with excellent insulating properties.

Chemically bonded phosphate ceramics [3] can be good alternatives to portland cement due to their special properties. They can be made without firing which makes them easier, cheaper and more environmental friendly to produce. This is also the reason than the term “cement” is also used for this ceramics, since mixing them with water results to a hardened paste without the need of firing. Furthermore, comparison between the energy costs for producing such types of binders compared to portland cement showed that production of these binders usually demands much less energy compared to production of portland cement [4]. Magnesium potassium phosphate ceramic (ceramicrete) is an example of such type of ceramics. Dissolving monopotassium phosphate (KH_2PO_4) in water results an acidic solution in which magnesium oxide (MgO) can be dissolved and an acid-base reaction initiates between the two dissolved components. Usually a small amount of boric acid is also used as retardant to slow the setting. Moreover, since the acid-base reaction is highly exothermic, the reaction rate of the dissolved components needs to be slowed down by using additional materials or methods to avoid the rapid setting of the slurry and thus ending up with an incomplete product with no workability, especially for production in large size. Thus, MgO is calcined at 1300 °C prior to mixing in order to reduce its solubility for practical applications [4]. However, the exothermic heat of the reaction may be needed to be reduced further and thus, monopotassium phosphate with low solubility of for example about 15 g/l may be used for this purpose [5].

Since the main focus of this study is on thermal conductivity, the attempt was made to make a composite with even lower thermal conductivity. It is noteworthy that although replacing chemically bonded phosphate ceramics by portland cement can result in a composite with lower thermal conductivity as well as eliminating moisture problems, the thermal conduction of the binder is still much higher than the aerogel granules [0.27 W/mK versus 0.012 W/mK). Thus, a simple method is proposed to reduce thermal conductivity of the matrix which leads to a more environmental friendly product as well.

2. Materials and Methods

In order to reduce thermal conductivity, the porosity of the matrix is increased by highly increasing the water-cement ratio without leading to a paste with high consistency which causes separation problem. The amount of water which is normally used in the ceramicrete mixes is also about 50 % of the binder weight (weight of MgO and KH_2PO_4). However the amount of water is increased up to higher than 300 % of the binder weight (MgO and KH_2PO_4) in this study without leading to a paste with high consistency.

In order to increase the demand for water in the mix, the calcined MgO is replaced by uncalcined MgO which has finer particles compared to after calcination. This also leads to a more environmental friendly product due to elimination of calcinations process and thus less energy consumption. Using uncalcined MgO is not practical in the normal ceramicrete mixes and the attempt for making the ceramic may fail due to a very high exothermic heat production. However, using hydrophobic aerogel granules in the mix as aggregates reduces the total exothermic heat production in the volume compared to the plain magnesium potassium phosphate ceramic mixes. Furthermore, the demand for water will also increase by using aerogel granules. On the other hand, using more water in the mix helps cooling the mixture during the mixing process. In fact, using uncalcined MgO with finer particles and aerogel granules helps to increase the water content of the mix, and increasing the water content together with using aerogel granules as aggregate make it practical to use uncalcined MgO with fine particles instead of calcined MgO.

Three mixes of with about 60.1 vol%, 45.3 vol% and 0 vol% aerogel with density od 0.12-0.18 g/cm^3 were made using uncalcined MgO with density of 3.58 g/cm^3 and KH_2PO_4 with density of 2.34 g/cm^3 and high solubility of 222 g/l at 20°C as the cement (See Table 1). Furthermore, fly ash with the density of 2.10 g/cm^3 as well as boric acid and super plasticizer (SP) based on modified acrylic polymers with 30% dry matter, were used in mixes. Although the considered cement composition reacts rapidly in presence of water, materials were mixed for about 20 minutes using Hobart mixer to ensure proper dispersion. The amount of water was adjusted to the limit that the slurry will not be dry and the mix becomes a firm paste.

Table 1. Compositions of the mixes

Mix name	mass (g)							w/b**	w/c*
	Fly Ash	KH_2PO_4	MgO	Water	Aerogel granule	Boric acid	sp		
M0	252.6	189.5	63.1	341	0	1	10	0.67	1.35
M1	192.9	144.7	48.2	424.5	75	1	10	1.10	2.20
M2	110.9	83.2	27.7	358.3	105	1	10	1.62	3.23

* w/c=water to cement ratio. Total mass of MgO and KH_2PO_4 is considered as cement mass, and fly ash taken account as binder.
**w/b=water to binder ratio

The samples were then cast in moulds of 4x4x16 cm and cured at a room with 50 % RH for four month when thermal conductivity of samples was measured at air dried condition. The samples were then dry sawed and dried at 105°C for seven days to measure the dry thermal conductivity, and the last measurement was held after water submersion for three days. Moreover, compressive strength of the cubic samples with size of about 4x4x4 cm was measured after 140 days curing at the room with 50 % RH.

3. Results and Discussion

The results in Table 2, reveal the low thermal conductivity of samples containing about 60 vol% aerogel granules which is in the range of thermal conductivity of building insulation materials such as mineral wool. The thermal conductivity can still be reduced by introducing more water and aerogel granules to the mixture. Insulating property of these materials can significantly be affected in presence of moisture and using hydrophobic agents can help overcoming this drawback. Moreover, since this material has an open porosity structure (it can breathe) and the thermal conductivity is also low at air dried condition, there might be no need for internal hydrophobation of the material in some specific applications. This can be decided for instance by using building physics simulation tools. This material has also a good ductility which is favourable during transportation and installation periods. In fact, aerogel granules are stabilized in a matrix with thermal conductivity lower than about 0.1 W/mK (extracted from the upper Hashin-Shtrikman bound for this composite). This gives for example thermal conductivity lower than 0.025 W/mK for 80 vol% aerogel content in this composite based on the Hashin-Shtrikman bound (see fig. 1).

Table 2. Experimental results

Sample	Dry density	TC* after drying at 105 °C (W/mK)	TC air dried* (W/mK)	TC submerged (W/mK)	Moisture content air dried (m^3/m^3)	Moisture content submerged (m^3/m^3)	Compressive Strength (MPa)
M0	1.031	0.163	0.345	0.753	0.31	0.59	5.66
M1	0.568	0.077	0.117	0.346	0.16	0.40	0.88
M2	0.340	0.040	0.056	0.170	0.08	0.28	0.27

* Thermal conductivity
**curing at room with 50 % RH for four month

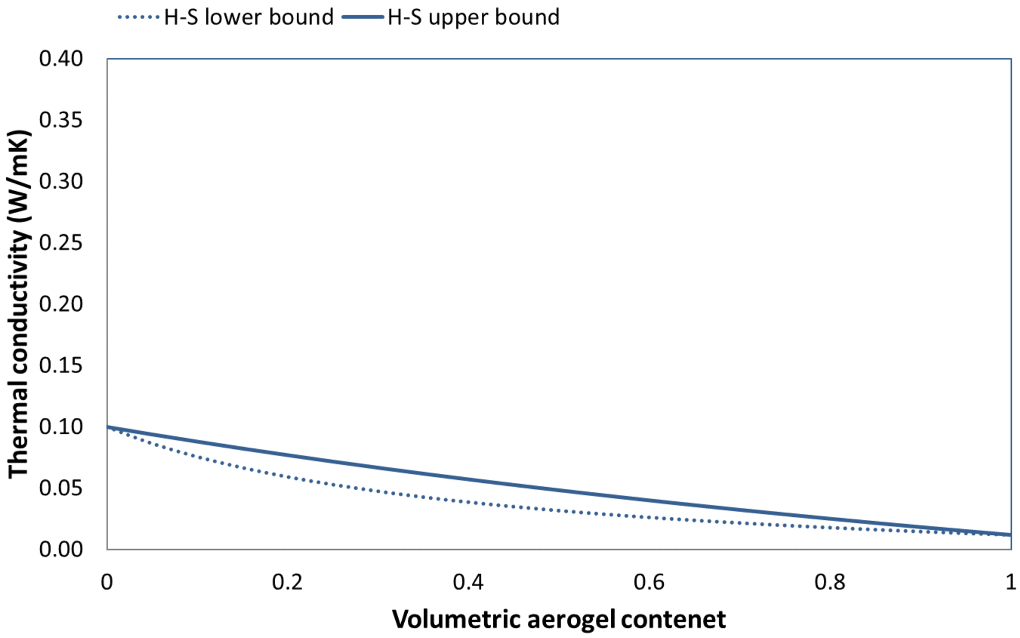

Figure 1. Hashin-Shtrikman bounds for thermal conductivity of the composite containing aerogel and cementitious binder according to the properties of particles and the matrix in M2 mix.

The compressive strength of aerogel composites are significantly lower than the reference samples (M0), this reduction was expected due to incorporation of aerogel which has a high porosity (usually more than 90%). However, the material can still be used as a self-standing insulating material. This type of composite can also be used for on-site 3D printing of building envelope such as sandwich wall elements due to the fast setting property. While the mix proposed by Wagh et al. [5] using calcined MgO results in phosphate cement with high compressive strength, the mix composition in this paper results in a composite with light weight, versatility, quick production and installation, low thermal conductivity and an environmentally friendly binder. Since there has been considerable improvement in the technology of aerogel production in recent years, it is expected that the cost and environmental impacts of composites made of aerogel will be reduced significantly and there will be more opportunities to introduce such composites to the market.

4. Conclusion

Cementitious materials have a considerable potential to be exploited as multifunctional materials including thermal insulation materials. Incorporating aerogel in cementitious composites can help reducing the thermal conductivity close to insulation materials. The thermal conductivity can also be reduced by increasing water-cement ratio. Insulating property of these materials can significantly be affected in presence of moisture and using hydrophobic agents can help overcoming this drawback. This type of composite can be used for on-site 3D printing. Material properties can be adjusted for using the composite as cementitious blocks for wall element, self-standing insulating material or as a part of sandwich panels. Elimination of calcination process resulted in an environmentally friendly binder with low thermal conductivity.

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