Cement and Concrete Research 39 (2009) 1052–1059

Contents lists available at ScienceDirect

Cement and Concrete Research

journal homepage: http://ees.elsevier.com/CEMCON/default.asp

Experimental investigation of the effect of temperature on the first desorption isotherm of concrete

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ARTICLE INFO

Article history:
Received 18 December 2007
Accepted 18 June 2009

Keywords:
Temperature
Surface area
Adsorption
Durability
Concrete
Radioactive waste
Waste management

ABSTRACT

In the framework of the radioactive waste management in France, interim storage concrete structures should be submitted to temperatures up to 80 °C and subsequent desiccation. The impact of temperature on the sorption properties of concretes has been poorly studied and results are scarce. An experimental campaign was thus carried out to characterize the first desorption isotherms of a modern concrete at 30 °C and 80 °C. The results show a significant influence of the temperature increase that will have to be accounted for the durability assessment of the long-term interim storage concrete structures. Investigating the causes of these modifications, it appeared that desorption induced by temperature might be the principal mechanism rather than microstructure alteration and water properties evolution.

1. Introduction

In the framework of radioactive waste management, the concrete structures for radioactive waste management are expected to undergo significant heating due to the waste thermal power. In the French concept of subsurface structures, the cooling is likely to be achieved using natural convection with air taken from outside. Doing so, the concrete temperature is expected to reach (but not to exceed) 80 °C in normal conditions and the concrete structure is expected to undergo severe drying (due to the temperature increase of the ambient air) [1,2]. The durability assessment of these structures over 300 years thus requires the accurate knowledge of the sorption properties of concretes at such unusual temperature levels with particular interest in desorption (corresponding to the drying process).

The influence of temperature on the sorption isotherms has been poorly studied and experimental results are scarce in the scientific literature. Among these studies Daïan has experimentally determined the adsorption isotherms of a water-cured mortar at four different temperatures: 20, 35, 45 and 55 °C [3,4]. The samples have been initially dried at 80 °C and then exposed to different increasing RH for the four temperatures. The curves thus obtained are reproduced on Fig. 1 (according to the results published in [4]). The temperature influence can be summarised as follows: the higher the temperature, the lesser the amount of water adsorbed. Nevertheless, the offset between the curves obtained at 20 °C and 55 °C was found to be low.

This observation is in good agreement with the experiments of Radjy et al. [5] who characterised the first adsorption branch of two mature (about 2 years old) steam-cured at early-age (at nearly 100 °C) hardened cement pastes with water-to-cement ratio (W/C ratio) equal to 0.35 and 0.45 respectively for temperatures between 0 °C and 60 °C. The results have shown a very limited dependence of the isotherms to temperature in this range. In the same way, the first desorption isotherm of a mature (kept 1 year under water) hardened cement paste cured at ambient temperature with a W/C ratio equal to 0.50 was characterised for temperatures between 0 °C and 40 °C. As mentioned before, the resulting desorption isotherms showed hardly any dependence on the temperature used.

In a recent attempt to develop a model capable of predicting the behaviour of concrete in any arbitrary environment Ishida et al. [6] have characterised the sorption isotherms (the first adsorption and the first desorption branches) of an 80-day water-cured hardened cement paste (W/C ratio equal to 0.50) for 20 °C, 40 °C and 60 °C. The results obtained for the first adsorption have shown little differences between the curves for the three temperatures. However, the results of the first desorption appeared to be much different: the first desorption isotherm was greatly modified by the temperature increase. The decrease of the amount of water adsorbed at higher temperatures was more pronounced than that obtained for adsorption. Moreover the shape of the isotherms was also modified by the temperature increase.

These facts have already been pointed out by Hundt and Kantelberg [7] who characterised the first desorption isotherm of different cementitious materials for temperatures ranging from 20 °C to 70 °C.

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doi:10.1016/j.cemconres.2009.06.019

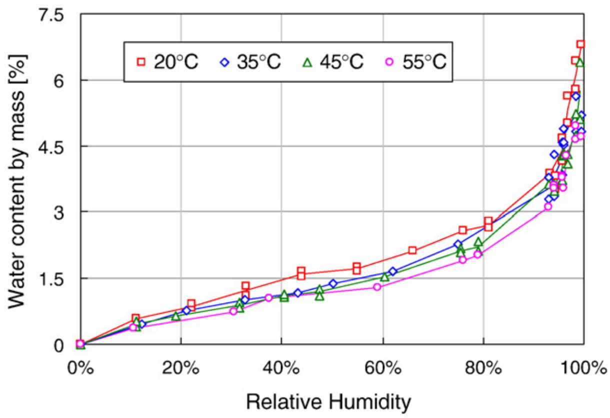

Fig. 1. Adsorption curves for a mortar between 20 °C and 55 °C after Daïan [4].

The results obtained with a 3.6 year old mortar (W/C ratio equal to 0.50) are reproduced on Fig. 2 (according to the data published in [7]). As mentioned before the higher the temperature, the lesser the amount of water adsorbed at equilibrium. One can see on Fig. 2 that the 20 °C desorption isotherm was nearly a straight line. This pattern was greatly modified when working at higher temperatures: see for instance the linear part between 20% and 50% with a decreasing slope related to the temperature increase and the pronounced upwards deviation for high RH. In addition, one can note the significant decrease of the amount of water adsorbed at saturation as a function of temperature: the higher the temperature, the lesser the amount of water at saturation. Hardened cement pastes with varying W/C (between 0.40 and 0.55) and concretes with various preliminary treatments were also tested by Hundt and Kantelberg [7], all the results obtained in this study (various cementitious materials with various curing methods) confirm all these observations.

An experimental campaign was then undertaken to highlight the influence of temperature and gather experimental data for further numerical approaches and durability assessment of interim storage concrete structures. The first desorption isotherm of a modern high-performance concrete was characterised at 30 °C and 80 °C. The temperature dependence of cementitious materials sorption properties is generally related to the coarsening of the pore structure (ettringite dissolution and C–S–H alteration) together with the evolution of water properties [8–11]. In this study, the examination of the results obtained indicates that another phenomenon may be at work and suggests that the microstructure alteration and the water properties evolution may have negligible effects.

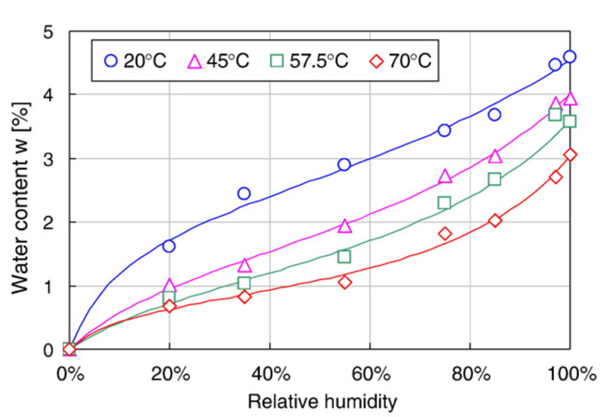

Fig. 2. First desorption isotherms for a mature mortar for four temperatures ranging from 20 °C to 70 °C according to Hundt and Kantelberg [7].

Table 1
Concrete formulation.

Component	Nature	Origin	Qty	Unit
Cement	CEM I 52.5	Lafarge Val d'Azergues, France	400	kg/m^3
Sand	Calcareous [0–5]	Boulonnais quarry, France	858	kg/m^3
Gravel	Calcareous [5–12.5]	Boulonnais quarry, France	945	kg/m^3
Water	–	–	172	L/m^3
Superplasticizer	Glenium 27	BASF	10	kg/m^3

2. Experimental programme

2.1. Material tested

The material used for the experiments was a modern high-performance concrete, the formulation is reported on Table 1. It is based on CEM I cement. The water-to-cement ratio (by weight) is equal to 0.43. This formulation was used because it is one of those which are studied in the framework of radioactive waste management by the French agency for radioactive waste management (Andra).

The main properties of the concrete have been determined experimentally on samples taken out of the same batch and cured more than 1 year under water at 20 °C. The values obtained are reported on Table 2. All the characterization tests were performed on saturated samples at 20 °C (except for the gas permeability measurements and water porosity which implied complete drying at 60 °C) and after thermal treatment at 80 °C (until constant mass was achieved, residual properties after cooling). The intrinsic permeability to gas (nitrogen) was estimated using the Klinkenberg approach [12].

The porosity to water at 30 °C and 80 °C was estimated using the mass loss obtained using silica gel (RH ~ 3%). It was also estimated using two other usual experimental protocols: drying at 60 °C with silica gel and drying at 105 °C. All the results are presented in Table 3. The concrete saturated density was calculated using hydrostatic weighing, the mean value was found to be equal to 2.48.

2.2. Technique used

The concrete first desorption isotherm was determined at 30 °C and 80 °C. The desiccator method using saturated salt solutions was chosen for its simplicity and its reliability. This is a standalone system which minimises human interventions and is recommended by the European Committee for Standardization [13]. The different RHs

Table 2
Main concrete properties, all given data are at least the mean of three values.

Concrete properties	20 °C	80 °C	Unit
Tensile splitting strength (Brazilian test)	5.80	4.95	MPa
Compressive strength	89.7	79.5	MPa
Elastic modulus	49.2	41.7	GPa
Poisson's ratio	0.18	0.17	–
Water porosity	11.3	12.0	%
Intrinsic permeability (nitrogen)	2.0×10^{-17}	4.2×10^{-17}	m²
Klinkenberg coefficient	0.13	0.06	MPa
Heat conductivity	2.49	2.09	W/mK

Table 3
Porosity to water according to the experimental protocol.

Protocol	30 °C (silica gel)	60 °C (silica gel)	80 °C (silica gel)	105 °C (no silica gel)
Porosity to water	10.7%	11.3%	12.0%	12.3%