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Multi-scale investigation of the effect of γ irradiations on the mechanical properties of cementitious materials

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HIGHLIGHTS

- Mechanical properties of gamma irradiated mortar and cement paste were studied.
- Gamma irradiation induced physical-chemical and mechanical modification of the uncarbonated cement materials.
- Water content influences the effect of gamma radiation on mechanical properties.
- Micro-indentation results agree with macroscopic observations.

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ABSTRACT

We studied the effects of low doses of gamma radiations on physical and hydro-thermo-mechanical behavior of concrete in the context of radioactive wastes storage. Mortar samples were irradiated with a Cs-137 gamma source to perform three-points bending, compression and physical analysis. Dry, humid and carbonated mortars and dry and carbonated cement pastes were used. The structural and mineralogical modifications at microscopic scale due to irradiations are determined. The results show an important drop of mechanical strengths except in the case of carbonated samples. Thus, the hydrated phases of the mortar/cement are likely involved in the degradation of the mechanical properties.

1. Introduction

Concrete is a multi-phases material often used as structural material for the storage and disposal of radioactive wastes [1,2]. As the structures are exposed to different types of radiation (mainly neutrons and gamma), it is essential to understand their effects on concrete and determine the evolution of its mechanical properties after irradiation over long-term period.

Concrete behavior under radiations has long been studied mainly under neutron radiations in the case of nuclear power plants. For this radiation type, Hilsdorf [3] showed that the mechanical properties of concrete and particularly the strength characteristics may deteriorate because of nuclear radiations if the cumulated dose exceeds limiting values (1.10^{19} n/cm^2). Some authors agreed that RIVE phenomenon (Radiation Induced Volumetric Expansion) is the predominant phenomenon which occurs in concrete after irradiation [4]. Kontani [5] suggests neutron or high dose rate gamma radiation effects are mainly concentrated at the level of hydrated and crystalline phases even though gamma radiations have a lesser impact on crystalline phases. Indeed, their effects mainly concern aggregates, suggesting that the variation of the concrete strength would be mainly created by cement paste intrinsic damages due to the volume variations of aggregates. The variation of the mechanical properties of concrete depends therefore on aggregates composition under neutronic radiation. Reviewing literature, Field [6] shows that whatever is the chemical composition of the aggregate type, a variation of the material strength appears only beyond specific threshold. This variation is also observed by Maruyama [7] under gamma radiations but for high doses (2.10^8 Gy). Giorla [8] describes this strength variation as a result of three steps, an initial damage located in the aggregates/paste interface (ITZ), an increase of these damages as a result

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of the various applied constraints and a propagation of the damages through the paste. However, it is important to note that the variation of mechanical strength is highly dependent on the type of aggregates, studies having mainly been made on siliceous ones. Rosseel [9] describes these variations as a result of the thermal expansion of the aggregates and of the radiolysis drying of the paste. With their numerical model, Le Pape [10] and Maruyama go farther by explaining that the radiation effect occurs in Si-O bonds situated in the CSH phases generating a modification of the porosity of the material. Hence, pre-existing damage before irradiations in the cement phases are aggravated by CSH degradation (modification of the organization of the CSH chains) and volumetric expansion of aggregates after irradiations.

However, it is important to note that these conclusions were drawn in the case of high-dose irradiation of cementitious materials, mainly in the case of neutronic radiation. Although some effects of gamma irradiations on cementitious materials have been described in the literature [7,11,12], the degradation mechanisms and their mechanical impact are not well understood yet especially under low gamma irradiations where it is difficult to separate radiation effects and environmental effects. Most of the studies focused on physico-chemical evolutions, mainly because of radiolysis effects. Indeed, concrete is a material including an important part of water that could be affected by the radiations. Water radiolysis is a phenomenon that occurs under ionizing radiation (gamma radiation) and produces many species which can react with each other or with the environment to produce gases especially H_2 [13]. Hydrogen production occurs mainly with the radiolysis of the free water present in macro porosity which can be composed by connected or unconnected pores [14]. Hence, H_2 gas produced by water radiolysis can be evacuated in the case of connected pores but is trapped in the material in the case of unconnected pores. Bouniol [15,16] indicates that in a closed system saturated in gas, a gas/solution equilibrium occurs. Therefore, the H_2 gas stays in solution and reacts with other radicals according to the Allen cycle, in particular in the smaller pores [17]. Indeed, if pore diameters are smaller than the radicals distribution area, then the radicals reactions with each other are increased. Hydrogen gas production in closed porosity can create constraints inside the material and generates cracks weakening the concrete. However, porosity can be directly impacted by atmospheric carbonation. Indeed, Groves and Kobayashi [18,19] showed carbonation deteriorate C-S-H and portlandite phases which are the main phases constituting cement

Table 1
Mortar compositions.

Cement (kg/m^3)	Calcareous Sand 0/4 (kg/m^3)	Water (kg/m^3)	W/C	Paste volume (%)	Air (L/m^3)
566	1344	270	0.43	45	20

Table 2
Mortar samples description.

Series	Name	Storage in sealed conditions	Drying	Carbonation	Irradiation	Storage in air-conditioned room
Series 1	HM-257kGy-P1	X				X
	HM-257kGy-P2	X				X
	HM-257kGy-P3	X				X
	HM-257kGy-I1	X			X	
	HM-257kGy-I2	X			X	
	HM-257kGy-I3	X			X	
Series 2	DM-257kGy-P1		X			X
	DM-257kGy-P2		X			X
	DM-257kGy-P3		X			X
	DM-257kGy-I1		X		X	
	DM-257kGy-I2		X		X	
	DM-257kGy-I3		X		X	
Series 3	CM-257kGy-P1		X	X		X
	CM-257kGy-P2		X	X		X
	CM-257kGy-P3		X	X		X
	CM-257kGy-I1		X	X	X	
	CM-257kGy-I2		X	X	X	
	CM-257kGy-I3		X	X	X	

Table 3
Cement paste samples description.

Series	Name	Storage in sealed conditions	Drying	Carbonation	Irradiation	Storage in air-conditioned room
Series 1	DCP-85.7kGy-P1		X			X
	DCP-85.7kGy-P2		X			X
	DCP-85.7kGy-P3		X			X
	DCP-85.7kGy-P4		X			X
	DCP-85.7kGy-P5		X			X
	DCP-85.7kGy-I1		X		X	
	DCP-85.7kGy-I2		X		X	
	DCP-85.7kGy-I3		X		X	
Series 2	CCP-85.7kGy-P1		X	X		X
	CCP-85.7kGy-P2		X	X		X
	CCP-85.7kGy-P3		X	X		X
	CCP-85.7kGy-I1		X	X	X	
	CCP-85.7kGy-I2		X	X	X	
	CCP-85.7kGy-I3		X	X	X	