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Effect of γ-irradiation on strength of concrete for nuclear-safety structures

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Abstract

Concrete applied for construction of nuclear power plant (NPP) Temelín (Czech Republic) has been exposed to γ-irradiation up to dose 6×10^5 Gy. Depending on the level of irradiation, changes in strength, porous structure and phase composition of the concrete have been studied. It is found that irradiation lowers both the strength of concrete (about 10%) and volume (resp. surface) of porous space. On the other hand, γ-irradiation increases the ratio of calcite, $CaCO_3$, in the concrete. Observed effects are discussed with respect to safety of NPPs.

Keywords: Concrete; γ-irradiation; Strength; Porosity

1. Introduction

In the present paper, we deal with the following open problems:

(A) Can the interaction of γ-irradiation with concrete influence negatively its mechanical properties?
(B) How role is played by the "radiation brittleness" in the safety of nuclear power plants (NPP)?
(C) What is the real mechanism leading to the (eventual) deterioration of mechanical properties of the concrete?

To answer the first question, we have studied the changes of mechanical properties of irradiated concrete in terms of changes of appropriate strengths (compressive, resp. transversal) of material; also the bend-tests of samples were performed.

The second problem (B) is closely connected with the analysis of radioactive conditions within hermetic part of real NPP. Thus, we have chosen the situation in NPP Temelín, where the single components constructed from concrete are distant no less than 3.739 m from the axis of reactor [1]. For such a distance, the incident dose rate does not overcome the value 1.04 Gy h^{-1} [2] under standard operational activity. Since the service life of NPPs corresponds maximally to 40 years (including one upgrading) it seems to be acceptable to suppose that constructional concrete absorbs during this period approximately 3.5×10^5 Gy of absorbed radiation dose.

Bouniol and Aspart [3] explain interaction of hardened cement paste (HCP) with γ-irradiation by the following scheme:

(1) Because of hydrolysis of molecular water, the peroxide H_2O_2 is systematically formed during this process.
(2) H_2O_2 reacts with cement calcium and the peroxide octahydrate $CaO_2.8H_2O$ is produced.
(3) Due to its metastability, this substance is then decomposed by the subsequent carbonation reactions onto calcium peroxide and water, respectively.
(4) CaO_2 then reacts with H_2O to produce portlandite $Ca(OH)_2$ and oxygen.
(5) Finally, portlandite reacts with CO_2 (involved within pores, air, etc.) and calcite $CaCO_3$, resp. water are formed.

Consequently, to solve problem (C) we assume that interaction of concrete with γ-irradiation leads to increase of

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Table 1
Composition of 1 m^3 of concrete

Component	Mass
Cement	499 kg [42,5R]
Water	215 kg
Plasticizer (Ligoplast SF)	2.8 kg
Aggregates: siliceous gravel	
0–4 mm	705 kg
8–16 mm	450 kg
16–32 mm	527 kg

calcite content in material. As an irradiation source, ^{60}Co has been used with initial activity 1.73×10^{16} and of 1.68×10^{16}Bq at the end of experiment. Energy range has not been measured. On the other hand, from the analysis of carbonization of HCP it follows that crystallites of calcite grow into pores (and closing them) and, simultaneously, they destroy tobermorite gel by crystallization pressure. Hence, we adopt additional hypothesis.

(6) Interaction of γ-irradiation with concrete leads to lowering not only its strength, but also its porosity (and other characteristics of pore space).

2. Experimental

In our experiments, we use concrete of the same composition as in the case of NPP Temelín (see Tables 1 and 2). From the initial mixture, 16 samples (of size 0.4×0.1×0.1 m) were prepared. Then, after 90 days of aging, 12 samples have been equipped with dosimeters (calibrated Si diodes) and irradiated. Maximal dosage during 83 days of irradiation does not exceed the value of 10^6 Gy. After 180 days from beginning of concrete mixing, we started to measure (by standard methods) strength in bend both on unradiated standards and also on irradiated samples. Furthermore, the strengths (compressive and transversal) were measured on fragments of samples remaining from previous bend-tests. (Duration of the strength-measurements does not exceed 1 day.) Fragments of samples then have been analyzed from the structural point of view during next 5 days: mercury porosimetry, resp. BET method and also diffraction analysis have been applied.

Table 2
Composition of cement CEM I 42,5R Mokrá

Sample	CEM I 42,5R Mokrá	
Type	Quantitative composition in % by volume	Quantitative composition in % by mass
Phase clinker composition		
C_3S	70.0	68.5
C_2S	11.4	11.6
C_3A	7.9	7.4
C_4AF	9.7	11.5
C_{free}	1.0	1.0
Total	100.0	100.0
Fraction of components in cement		
Clinker	93.3	95.0
Gypsum	4.6	3.5
Fly ash	1.8	1.3
Slag	0.3	0.2
Total	100.0	100.0

3. Results and discussion

In Figs. 1–3, the results of strength-tests are presented. It is clear that with increasing dosing of irradiation all of three types of strengths markedly decrease. Consequently, the answer to the first question (A) reads: interaction of concrete with γ-irradiation negatively influences its mechanical properties (already for dosages ranging from 5×10^5 Gy). For example, in the case of compressive strength this decreasing represents (for dose 5×10^5 Gy) approximately 10% of the value of nonirradiated sample. This result is really surprising, since according to recent opinions (see, e.g., Ref. [5]) it is assumed that the dosage limit for deterioration of mechanical properties of the concrete is larger than 10^8 Gy.

From analysis of real situation in NPP Temelín, it follows that the level of dosage 5×10^5 Gy corresponds to 57 years of the "normal" operation time of NPP. This

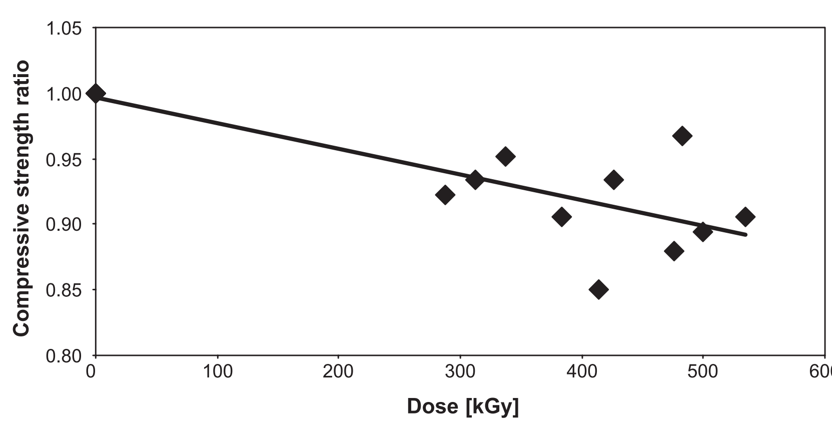

Fig. 1. Dependence of compressive strength ratio on dose of gamma radiation. Value of standard (dose zero) is 69.4 MPa.

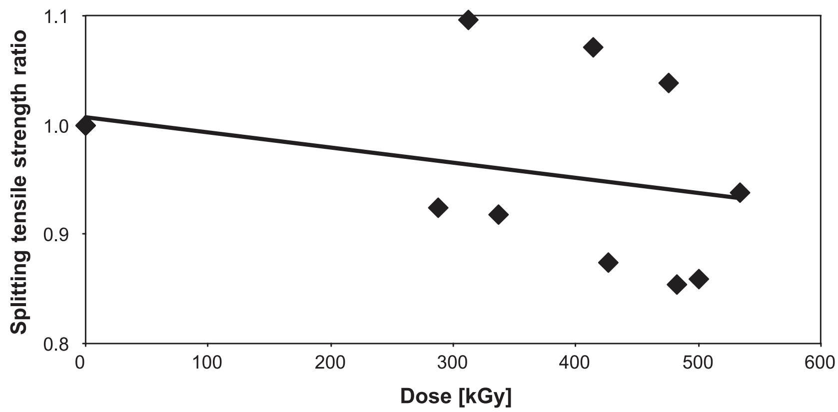

Fig. 2. Dependence of splitting tensile strength ratio on dose of gamma radiation. Value of standard (dose zero) is 7.36 MPa.

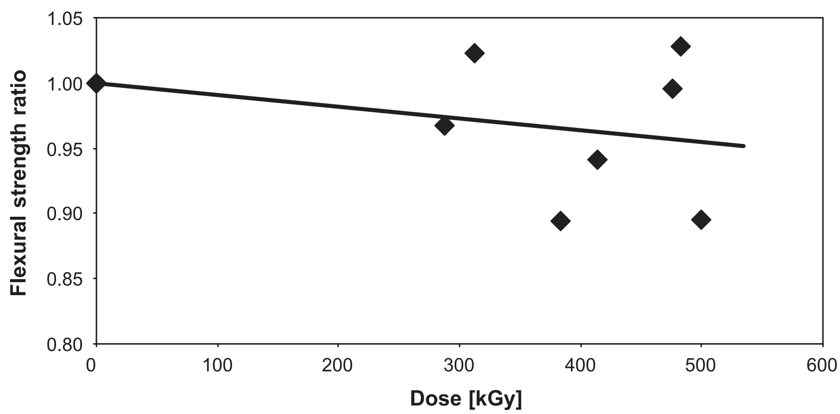

Fig. 3. Dependence of flexural strength ratio on dose of gamma radiation. Value of standard (dose zero) is 6.75 MPa.

period, of course, substantially exceeds its service lifetime. Hence, with respect to our problem (B) it can be claimed that "radiation brittleness" of the concrete produced by its interaction with γ-irradiation does not lower the safety of NPP under standard conditions (i.e., without any heavy accidents).

Average values of characteristics of pore space of nonirradiated samples are collected in Table 3.

In Figs. 4–7, the changes of the above characteristics produced by dosage are shown. It may be readily seen that with increasing dose of irradiation the values of these characteristics decrease. For example, porosity decreases to the value less than one half of that of nonirradiated standard sample. Fig. 8 represents the results of diffraction analysis of calcite content in concrete for different values of dosage. (All curves are normalized to intensity of Si diffraction line, which exhibits the same value for all samples under consideration.)

Table 3

Porosity [%]	Specific volume of micropores ($mm^3\ g^{-1}$)	Specific surface of pores ($m^2\ g^{-1}$)	Specific surface of mesopores ($m^2\ g^{-1}$)
12	1.1	4.4	2.5

It is clear that content of calcite increases with increasing dosage. Because the diffractograms have been obtained within the time interval of 48 h, "natural" carbonization of concrete may be excluded. Thus, interaction of concrete with γ-irradiation may lead to the phenomenon called "radiation" carbonization. Consequently, answer to our problem (C) can be formulated as follows.

Interaction of concrete with γ-irradiation generates the succession of chemical reactions in material, starting with radiolysis of water and terminating in formation of calcite, crystals of which decrease both the size of pore space and also the strength of material [4].

The last statement could be understood to be rather working hypothesis which has to be confirmed by more detailed and sophisticated measurements (e.g., by electron microscopy).

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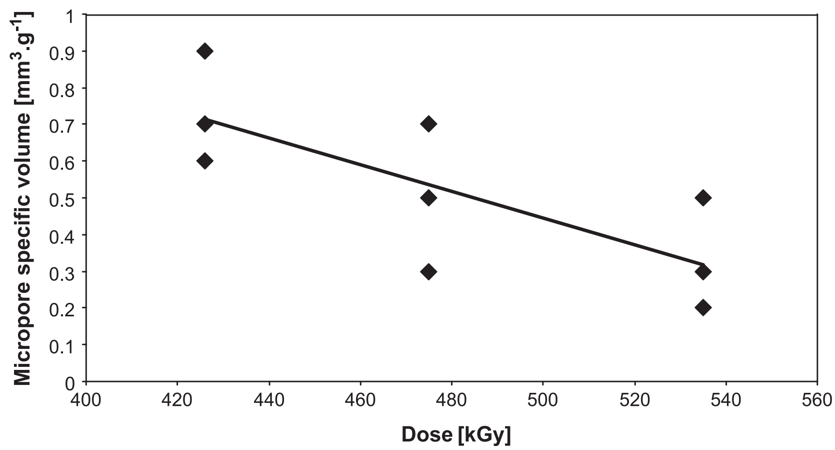

Fig. 4. Dependence of porosity on dose of gamma radiation. Value of standard (dose zero) is 12%.

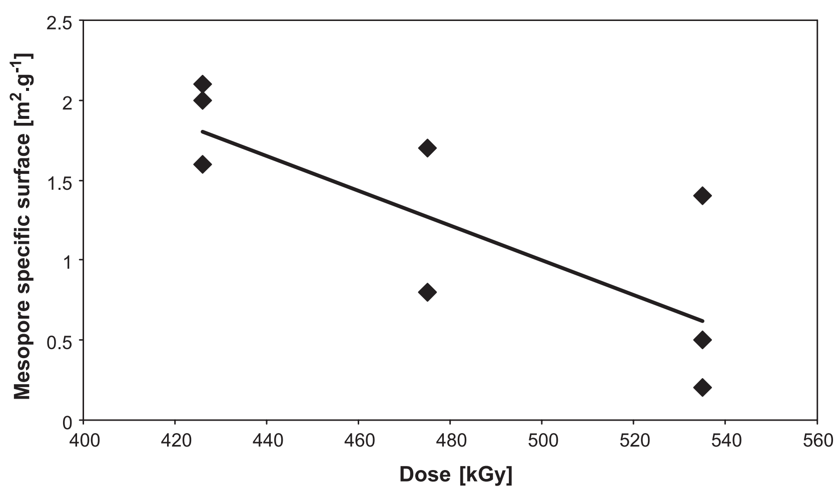

Fig. 5. Dependence of specific volume of micropores on dose of gamma radiation. Value of standard (dose zero) is 1.1 mm^3 g^{-1}.

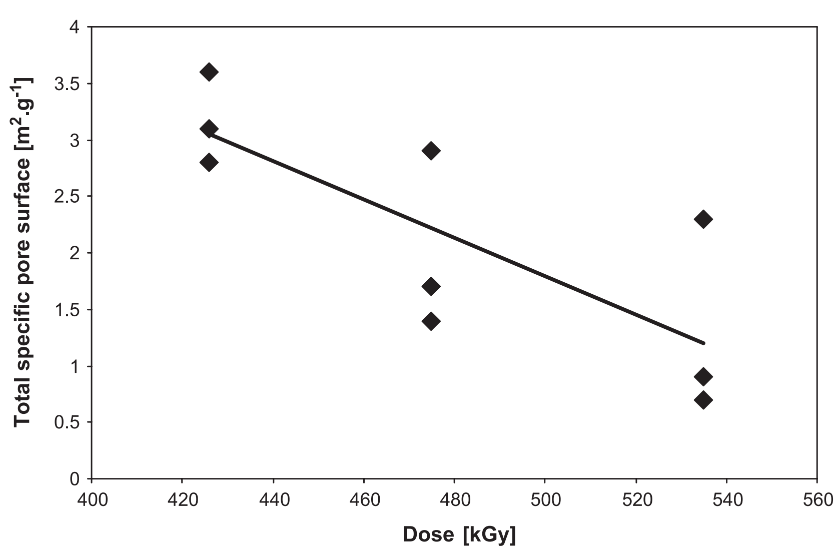

Fig. 6. Dependence of specific surface of pores on dose of gamma radiation. Value of standard (dose zero) is 4.4 m^2 g^{-1}.

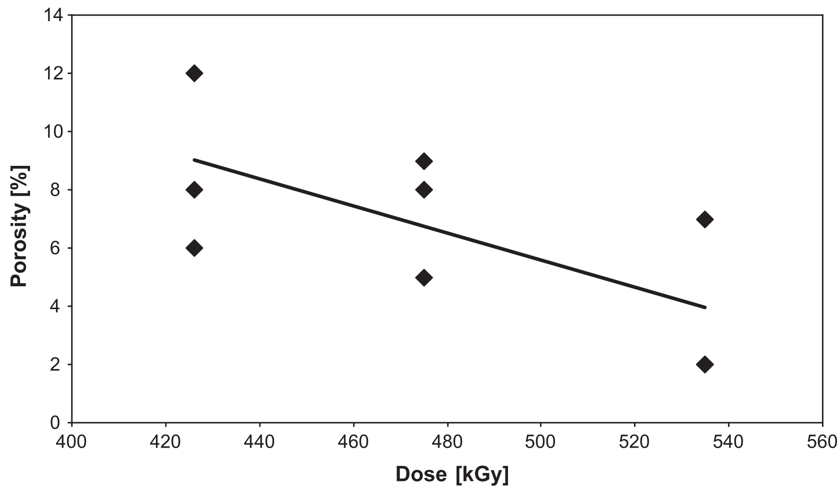

Fig. 7. Dependence of specific surface of mesopores on dose of gamma radiation. Value of standard (dose zero) is 2.5 m^2 g^{-1}.

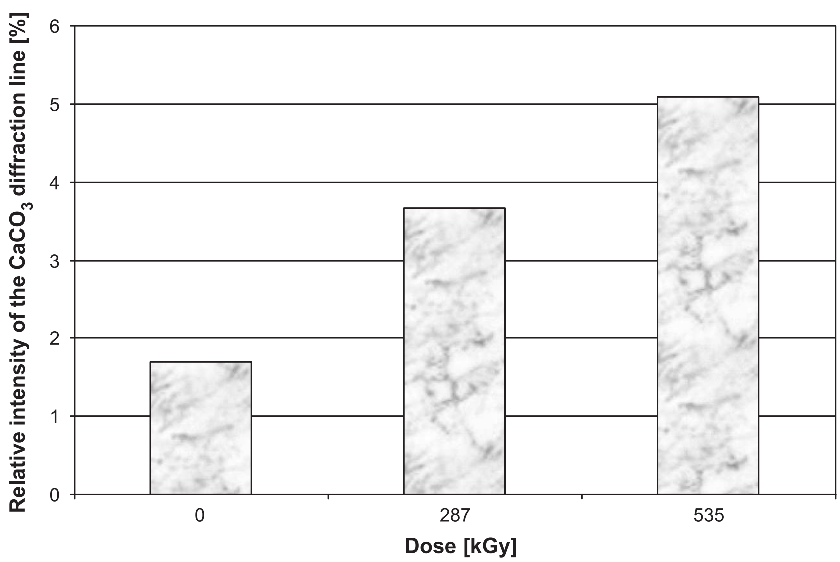

Fig. 8. Dependence of intensity of the $CaCO_3$ diffraction line on dose of gamma radiation.

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