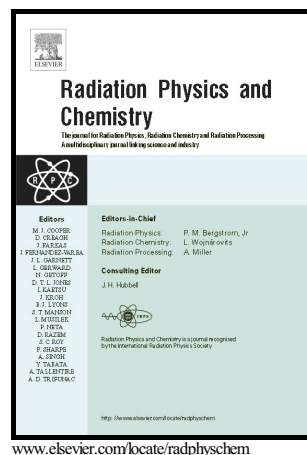


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Original behavior of pore water radiolysis in cement-based materials containing sulfide: coupling between experiments and simulations

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ABSTRACT

Blended cements with high content of blast furnace slag (CEM III/C) can be used for nuclear waste conditioning because of their low hydration heat as compared to ordinary Portland cements (CEM I). They however contain some sulfide, an impurity whose role needs to be investigated. Indeed, they can have an effect on the radiolytic H₂ production under irradiation. To study the impact of sulfide species on H₂ production, gamma irradiation, at a dose rate of 356 Gy h⁻¹, was performed during 6 months in a closed system without O₂ on a cement paste made with CEM III/C. At short time, the radiolytic H₂ production rate is higher than that measured using CEM I. On the basis of reaction data collected in the literature on sulfur species, radiolysis simulations performed for both systems confirm this behavior. Moreover, they suggest that the sulfide concentration, initially imposed in pore solution by the slag is of the order of 180 mM, and is responsible for this H₂ production. For the first two irradiation months, the following phenomena are then evidenced in CEM III/C: 1) conversion of sulfide into polysulfide anions; 2) pH increase; 3) production of H₂ due to the H^{•+} + H₂S reaction having a very high rate constant. Nevertheless, in the medium term, the residual sulfide

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