Gas diffusion effect on H_2 release caused by water radiolysis in geopolymer

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The hydrogen production of geopolymers under gamma irradiation was evaluated and confronted to a model considering hydrogen production, its diffusion and potential recombination reactions. We were able to highlight the importance of hydrogen diffusion and recombination to predict the hydrogen explosion risk accurately [1]. An experiment is planned in FY2019 for its demonstration in collaboration with French CEA.
Keywords: geopolymer, radiolysis, hydrogen, diffusion, waste treatment

1. Introduction

Hydrogen gas production of a wasteform is an important factor to assess the risk caused by the treatment of nuclear waste. Although geopolymers gained interest in the past years for nuclear waste treatment [2], present an important amount of water, and may release hydrogen by radiolysis. When radiolysis occurs, the phenomena usually considered are the energy absorption by the wasteform, and a complex system of chemical reactions. In usual conditions, the hydrogen is transferred to gas phase faster than it is produced (released by the sample, creation of bubbles, cracks etc…). When released in gas phase, the hydrogen can hardly react, it is thus linearly produced and a G value can be measured or estimated, independent of the sample size, provided the total amount of gas is taken into account. However, when produced in the a porous solid the hydrogen must diffuse to gas phase and could only form a bubble inside the material if the local hydrostatic pressure was sufficient to overcome the capillary pressure. Being able to accumulate in liquid phase the radiolytic yield becomes dependent of the sample size and diffusive properties.

2. Model and Experimental Results – Irradiation of geopolymers

Under ^{60}Co irradiation, the hydrogen production yield was measured for water saturated geopolymer of different size to increase the diffusion length for the gas (Figure 1). From powder to 40 cm high cylinders a strong decrease of the hydrogen yield was measured, even after the hydrogen trapped inside the sample was allowed to diffuse out of the matrix. With f a function representing the hydrogen concentration, it was found that approximation of the solution of equation 1 allows to predict the hydrogen of all samples with a reaction constant of $6.10^{-4}\ s^{-1}$, all other parameters (P and D) fixed on literature values. However, if these equation allows to predict accurately the hydrogen production after experimental irradiation the recombination should be indubitably observable during the irradiation using *in situ* gas measurements.

$$\frac{\partial f}{\partial t} = P - kf + D\frac{\partial^2 f}{\partial x^2} \quad (1)$$

3. Conclusions

When the sample is large, the H_2 accumulates in the sample because of its low diffusion constant and is available for recombination reactions, making the global yield a lot lower than powdered samples. In this presentation, these results will be presented alongside with other literature result that could be explained by similar mechanism. Finally, the plan to investigate further the recombination with *in situ* measurements throughout a collaboration with French CEA will be developed

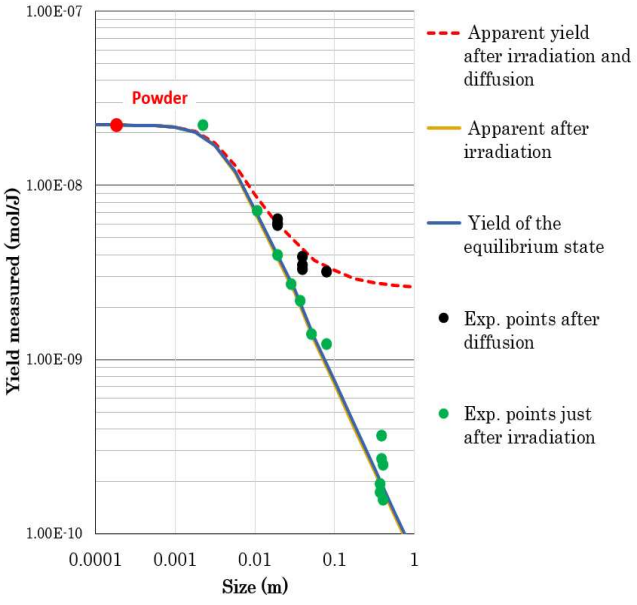

Figure 1 : Hydrogen production of geopolymers samples of different sizes and model predictions

References

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