

# Evaluation Of Hydrogen Generation From Radiolysis Of Water

*N.K.Abbasova*

Baku State University

Azerbaijan, Baku, Z. Halilova st.-23, AZ1148, (+994 12)539 05 35

Fax (+9942 12)598 33 76; [info@bsu.az](mailto:info@bsu.az)

E-mail: ramazanova.abbasova.nermin@gmail.com

Hydrogen production by  $\gamma$ -radiolysis of the mixture of beryllium oxide and water was studied in order to provide basic points of view for the influences of beryllium oxide and of rise in temperature on the hydrogen production by the radiolysis of water. As the influence of the beryllium oxide, an additional production of hydrogen besides the hydrogen production by the radiolysis of water was observed. This hydrogen production is strongly modified at oxide surfaces. Here we show that beryllium metal immersed in water uptakes considerable amounts of hydrogen when exposed to  $\gamma$ -radiation. Additionally we show that the amount of hydrogen absorbed by beryllium depends on the total dose of radiation.

**Key words:** Hydrogen production, BeO/water system,  $\gamma$ -radiation

## INTRODUCTION

Nuclear power is often argued to be a fossil-free alternative in the global spectrum of electricity generation [1,2]. The safety of operating nuclear power plants is usually of main concern in discussions comparing different energy production techniques. However, at present, one of the most difficult issues to tackle is the long-term safety of repositories for radioactive waste originating from nuclear power plants, in particular the spent nuclear fuel [3]. The consequences of the exposure of many homogeneous systems to ionizing radiation are well-known on the basis of both experimental and theoretical studies performed over a period close to a century [4]. However, most systems of practical relevance are not homogeneous. In fact, one of the most crucial and thereby also interesting components of a system from a performance perspective is the interface between two phases[5,6]. However, no convincing mechanistic explanation that accounts for these observations has been given. In addition to the somewhat puzzling observations regarding the radiation chemical yield of  $H_2$ , a number of studies on the interactions between other aqueous radiolysis products and oxide surfaces have been presented [7,8]. Additionally, under certain conditions, the decomposition of  $H_2O_2$  at oxide surfaces can also lead to the formation of  $H_2$  [9]. Multiple previous

studies have examined product formation by water radiolysis at the interface between beryllium oxide and water. Our study provides a general quantitative model for calculating radiolytic production rates as a function of distance from the beryllium-water interface. In our example, we focus on radiation dose profiles and radiolytic  $\text{H}_2\text{O}_2$  and  $\text{H}_2$  production profiles around spent fuel before and after barrier failure.

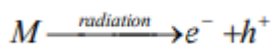
### **EXPERIMENTAL PART**

Each type of radiation,  $\alpha$ ,  $\beta$  and  $\gamma$ , has different radiolytic product yields. In addition, each follows a different attenuation law because  $\alpha$  and  $\beta$  radiation behave normalized dose rates for both  $\alpha$  and  $\beta$  radiation, and a somewhat different method for  $\gamma$  radiation. We first present our equations for calculating the dose rate of charged particles and then present the equations and additional geometric conditions needed to account for  $\gamma$  radiation.

How much radiation reaches the water depends on the particle's path to the solidwater interface. We assume the path is linear over the projected range along the initial travel direction. The molecular products -  $\text{H}_2$ ,  $\text{O}_2$  and  $\text{H}_2\text{O}_2$ , received at radiation and heterogeneous process have been determined by a gas chromatograph. Therefore, more exact information on kinetic regularity of process of radiation and heterogeneous transformation of water has been received on the basis of amount of molecular hydrogen.

### **RESULTS AND DISCUSSION**

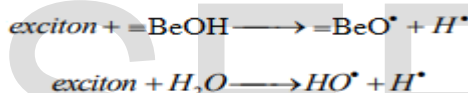
The radioactivity of spent fuel is highest during this time and  $\alpha$ ,  $\beta$  and  $\gamma$  radiation are emitted. Hazards associated with spent fuel storage include release of radionuclides into the water or atmosphere and, in some cases, buildup of dangerous levels of  $\text{H}_2$  gas. However, even in this case the contribution of  $\gamma$  radiation to the total dose, within the range of  $\alpha$  particles, is almost an order of magnitude greater than  $\beta$  radiation. As stated in our methods, for this study, we assumed homogenous distribution of radionuclides throughout the solid. Our model can be adapted to include different zones of activity within the solid, which will produce different radiolytic production profiles. In an irradiated heterogeneous system, when the two phases each constitute a significant fraction of the total mass, the ionizing energy is absorbed significantly by both phases. After the absorption of a high-energy photon, a high energy Compton electron is ejected. As charge carriers may cross from one phase to the other one, phenomena are thus different from those observed in a homogeneous phase. The interaction of radiation with an oxide causes electronic excitations, which promote an electron from the valence band to the conduction band, leaving a hole in the valence band.



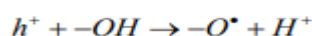
When an excited electron and a hole in the valence band remain bound together by Coulomb interactions, they are referred to as an exciton. The energy of the exciton is a little lower than the band gap energy. In semiconductors, the energy transfers by the coherent excitons. Among all the oxides of interest, the most extensive work has been performed on beryllium, due to the development of high-purity beryllium for optical fiber applications. Calculations evidence that there is no barrier for self-trapping .



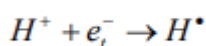
Calculations have also proven that the exciton self-trapping is accompanied by a weakening of one of the two Be-O bonds with an oxygen atom and by a small displacement of the oxygen ion towards an Be•≡interstitial position. We point out that the direct excitation of hydroxyl groups, which are present as impurities on the silica surface, is negligible. The division of bulk beryllium into nanometric particles emphasizes the role played by the surface. This dramatic surface effect is then gradually suppressed when the size of the beryllium particles is increased.



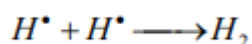
Nevertheless, the hydroxylation of the beryllium surface has an effect on the nature of the mechanism and a strongly dehydrated surface can tune the excitonic chemistry to a more ionic one as shown by the reactivity of the electron at the surface (dissociative electron attachment to -OH groups). It is suggested that positive holes rapidly react with surface -OH groups to give:



The protons will then react with trapped electrons at the surface to generate H atom.



Whether ionic or excitonic chemistry, the resulting H atoms dimerize forming H<sub>2</sub>



## CONCLUSION

We present a general model for quantifying water radiolysis by  $\gamma$  radiation near solid-water interfaces. Our model includes explicit consideration of the radiation's energy attenuation. By incorporating the activity, irradiance, and attenuation of radiation, our model separately calculates radiolysis due to  $\gamma$  radiation as a function of distance from the solid surface. As an example, we

calculate total dose rates and radiolytic production rates for spent fuel to illustrate the importance of including the contribution from all types of radiation in a general model of water radiolysis.

## REFERENCES

1. Chu, S. & Majumdar, A. Opportunities and challenges for a sustainable energy future. *Nature* 488, 294–303 (2012).
2. Lichter, S. R. & Rothman, S. Scientists' attitudes towards nuclear energy. *Nature* 305, 91–94 (1983).
3. Ewing, R. C. Long-term storage of spent nuclear fuel. *Nat Mater* 14, 252–257 (2015).
4. Friedlander, G. & Herrmann, G. in *Handbook of Nuclear Chemistry* (eds Attila Vértes et al. ) Ch. 1, 1–37, 10.1007/978-1-4419-0720-2; (Springer, US, 2011).
5. Féron, D. in *Nuclear Corrosion Science and Engineering* (ed Féron, Damien ) 31–56, 10.1533/9780857095343.1.31 (Woodhead Publishing, 2012).
6. Wolski, K. in *Nuclear Corrosion Science and Engineering* (ed Féron, Damien) 104–130, 10.1533/9780857095343.2.104 (Woodhead Publishing, 2012).
7. Haber, F. & Weiss, J. The Catalytic Decomposition of Hydrogen Peroxide by Iron Salts. *The Royal Society* 147, 10.1098/rspa.1934.0221 (1934).
8. Lousada, C. M., Johansson, A. J., Brinck, T. & Jonsson, M. Mechanism of H<sub>2</sub>O<sub>2</sub> Decomposition on Transition Metal Oxide Surfaces. *The Journal of Physical Chemistry C* 116, 9533–9543 (2012).
9. Lousada, C. M., LaVerne, J. A. & Jonsson, M. Enhanced hydrogen formation during the catalytic decomposition of H<sub>2</sub>O<sub>2</sub> on metal oxide surfaces in the presence of HO radical scavengers. *Physical Chemistry Chemical Physics* 15, 12674–12679 (2013).