Impact of α radiolysis on Bituminized Waste Products (BWP)

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In France, bitumen is used since four decades as a container material for low/intermediate activity and long lifetime radionuclides. Industrial Bituminized Waste Products (BWP) are produced by continuous extrusion of 60 %w. of bitumen and 40%w. of salts (volumic ratio ~3:1), in which a very small fraction is lastly radioactive. The organic matrix subjected to irradiation (α, β, γ) produced gases (mainly H_2), which can give rise to swelling of BWP in its container; the extent depends on the competition between the gas production rate and the evacuation rate by diffusion and migration of the gas bubbles. The determination of hydrogen source term requires the knowledge of the dose rate and the radiolytic yield of production G. For the γ irradiation and the β particles, a value of $G = 3{,}3\ 10^{-8}$ $mol.J^{-1}$ is commonly admitted and statistically consolidated by many studies [1, 2]. On the other hand, concerning the interaction of the α particles with bitumen, the results of some self-irradiation tests carried out on synthetic and inactive BWP show a rather broad dispersion certainly due to some screening phenomena.

Currently, the radiolytic yield (G) for α particles is assumed to be six times higher the γ radiolytic yield [3]. However, recent measurements on active BWP show that this value is over-estimated [4]. Moreover, it is supposed to be constant in time since one does not take into account "the consumption" of bitumen surrounding the α emitter.

In order to reduce these conservatisms, a better understanding of phenomena and physical mechanisms is necessary. The proposed approach is close to that adopted for the α radiolysis of polymers. It consists, in a first stage, to measure gas production yields using irradiations of simulation with swift heavy ions (GANIL, Caen, France) having the same stopping power as α particles emitted by the radioelements. The variation of the yield with the dose and according to the chemical composition (rate of aromaticity for example) will be quantified. The acquisition of these experimental data will be done on model samples. Bitumen is composed of organic compounds that belong to 4 families: saturated oils, aromatic oils, resins and asphaltenes. Before the irradiations, the bitumen will be fractionated in these various generic families in order to study them separately. Then after swift heavy ion irradiations, the degradation of these compounds will be characterized by IRTF-ATR and NMR, the radicals formed under irradiation will be analyzed by RPE.

The aim of this first part of work is to complete the study carried out by P. Bernat [1] which investigates the degradation mechanisms of model BWP under γ irradiation. In particular, we will examine how these mechanisms are modified in the case of irradiation with of high Linear Energy Transfer (LET) particles.

In the second time, the screening phenomena due to salts constituting the BWP will be evaluated by the use of the dosimetry model developed with the CIMAP (Caen, France). This model is able to estimate the fraction of energy deposited in the organic matrix whatever the geometry of the system emitter-matrix.

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[2] S. Legand, V. Dauvois, S. Esnouf, NT CEA DPC/SECR 07-032 (2007)

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