Picosecond pulse radiolysis at ELYSE: geminate recombinaison in homogeneous solution and in confined media

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ELYSE (meaning LYSIS by Electron) is a picosecond pulse radiolysis research centre, the only one in Europe. It offers the community access to the new generation of a laser triggered accelerator delivering 5 ps electron pulses. It provides transient observations in the range from ps to a few seconds with state-of-the art detection system. The original conception of this accelerator of new generation is based on a high synchronization between a femtosecond laser extracting the photoelectrons, and the hyperfrequence wave accelerating the electron bunches in two RF cavities, in order to adjust their final energy from 2 to 9 MeV. This synchronization opens new promising possibilities by using laser pulses for the transient measurements (pump-probe experiments), and even for exciting molecules to upper states by combining electron and laser beams. ELYSE is a very versatile facility, adapted to a wide range of detection set-ups, and therefore to a wide research community.

Interaction between ionizing radiation (energetic photons or fast charged particles) and matter gives rise to ions (positive ions and electrons) and electronically excited molecules that are concentrated along the path of the ionizing species. In pulse radiolysis experiments with e-beam, like at ELYSE, the ionization of matter is induced by the secondary electrons formed during the interaction with the e-beam. Under these conditions in solvent, the spurs occur at intervals of about 20 μm and have an initial diameter of about 2 nm. The spurs then expand and disperse the reactive species by diffusion. The reaction that take place in the spurs and in the earliest stage of the diffusion govern the final products of the radiolysis mechanism.

Among the different reactions that takes place during the expansion of the spur, geminate or ion neutralization reaction are particularly important. While these processes are extensively studied in water solution, there are a lack of information in other solvents, in particular in less polar solvent where the low dielectric constant of the medium strongly increases the Onsager radius and hence the probability of reaction between charged species. We present here results about the geminate recombination in water, alcohol and THF studied in picosecond pulse radiolysis experiments.

Another category of problems where the knowledge of the initial radiolytic yield at the picosecond time-scale are important concerns the radiation chemistry in confined media like mesoporous materials. Owing to the confinement and to the vicinity of the allumino-sillicate surface, the initial radiolytic mechanism may differ strongly from the ones observed in homogeneous phase. We present here the first picosecond pulse radiolysis experiments on MCM-41 materials, that have been permitted owing to the synthesis of special transparent monolith.