

RADIATION EFFECTS IN MESOPOROUS MATERIALS – APPLICATION TO NUCLEAR WASTE PACKAGING

Considering their large interfacial surface, nanoporous materials offer interesting perspectives for the study of the evolution of damage induced by irradiation¹. In order to study this phenomenon, thin films (IPC, ICC) (figure 1) and pellets of mesoporous silica (SBA15, MCM41) produced by sol-gel process were irradiated at JANNUS, SIRIUS and GANIL facilities. The irradiations conditions are as following (i) Xe-0.55 to 0.71MeV/A, C-0.98MeV/A, Ca-9.17MeV/A, Ar-0.98MeV/A, Kr-0.86MeV/A and 2 MeV electrons, corresponding to electronic damage processes (ii) Au-0.0025/0.06 MeV/A corresponding to ballistic processes. The porous structure was analyzed by X-Rays Reflectivity or gaseous adsorption-desorption iso-therms and the silica wall network by various spectroscopy IR and NMR. All of the experimental results demonstrated that the ordered porous structure was damaged and a significant collapse of the mesoporosity inducing a compaction was observed [2-4]. The radius of the tracks representative of the damage to the structure is shown in figure 2 as a function of the total stopping power of the ions. As could be expected, higher is the stopping power larger are the track radius. For a same total stopping power, non-porous silicas have a greater damage radius than mesoporous one, that is mean that porous silicas present a better radiation tolerance than non-porous material. This conclusion is supported by the results of IR spectroscopy which demonstrate that the IR changes caused by irradiation on silica network in the porous material are less than that observed in non-porous silica at the same dose. However, as previously indicated, when the irradiation dose becomes sufficiently high, the mesoporous structure of the silica collapses.

From a technological point of view, mesoporous silica can be used to develop a new strategy for the treatment of radioactive effluents based on the use of

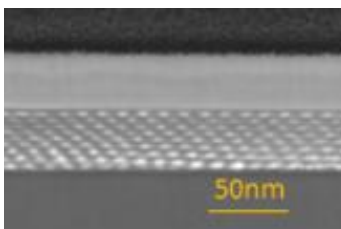


Figure 1. SEM image of SiO₂ mesoporous ordered thin film (IPC).

a porous functionalized support. After functionalization, this support would allow both the separation of the radionuclide, and their encapsulation after the collapse of the porosity by irradiation effect. This new concept would result in obtaining a primary wasteform matrix. Mesoporous silicas will be used as support materials, because the nanometric size of their pores allows easy closure. This new so-called separation / conditioning strategy would constitute a significant simplification of the number of step, compared to “traditional” processes for the treatment of radioactive effluents. The ANR Automact project aims to study the potential collapse of a mesoporous silica doped with a radioactive element (238Pu). Another field of application concerns the evolution during aging of the mesoporosity formed in the alteration gel of nuclear glasses. This mesoporosity is a factor aggravating the dissolution of the wasteform matrix. The effects of irradiation could lead to a closure of this mesoporosity, inducing self-healing of the gels. A thesis began in 2022 on this subject in the laboratory.

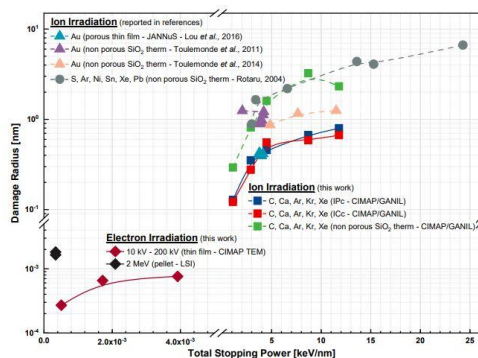


Figure 2. Summary of damage radius of the track induced by radiation damage as a function of total stopping power of the projectile (ions or electrons).

¹ P. Makowski, X. Deschanel, A. Grandjean, D. Meyer, G. Toquer and F. Goettmann - **Mesoporous materials in the field of nuclear industry: applications and perspectives** - New J. Chem., 36 (2012) 531.

² Y. Lou, S. Dourdain, C. Rey, Y. Serruys, D. Siméone, N. Mollard, X. Deschanel - **Structure evolution of mesoporous silica under heavy ion irradiations of intermediate energies** - Micropor. Mesopor. Mater., 251 (2017) 146.

³ J. Lin, G. Toquer, C. Grygiel, S. Dourdain, Y. Guari, C. Rey, J. Causse, X. Deschanel, - **Behavior of mesoporous silica under 2 MeV electron beam irradiation** - Microporous Mesoporous Mater. 328 (2021) 111454.

⁴ Y. Lou, B. Siboulet, S. Dourdain, M.R. Rafiuddin, X. Deschanel, J-M. Delaye - **Molecular dynamics simulation of ballistic effects in mesoporous silica** - J. Non-Cryst. Solids (2020) 549, 120346.