



Photoluminescence as a tool to probe nuclear waste storage matrixes

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A strong synergy between physicists, chemists and mineralogists is undergone to elaborate new matrixes to fixed and stored nuclear wastes such as fission products and minor actinides. Among those matrixes the R7T7 nuclear glass and the monazite crystalline phase are deeply studied. In such context, luminescence spectroscopy is a useful non destructive tool to probe the structure (local environment, irradiation induced defects...)

Natural monazites ($\text{Ce, La}\text{PO}_4$ (rare earth orthophosphates) are one of the rare minerals which have the original ability to incorporate great amount of actinides (Th, U) as well as to restore the radiation induced defects affecting their lattice without undergoing amorphisation (non metamict state). As a matter of fact low thermal energy is enough to cure α and nucleus recoil induced defects. Monazite is therefore considered to be one of the potential matrixes for the incorporation and storage of minor actinide wastes. Natural phases appear to be good analogues to understand the behavior of such matrix under storage and weathering conditions. Moreover it appears that all natural phases as well as synthetic ones incorporated neodymium ions as impurities when Nd^{3+} is known for its luminescent properties. As a matter of fact neodymium emission as the other trivalent rare earth element one is sensible to its atomic environment and to the crystal field. Therefore it can be use as local luminescent probe. In order to follow the healing several natural monazites with different ages, localities and Th-U contents were selected and thermally treated (500-1200°C). At each thermal

step the ${}^4F_{7/2} \rightarrow {}^4I_{9/2}$ and ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$ emissions were recorded under continuous 524nm Ar laser excitation and the FWHM of each Stark sublevels measured. It appears that the FWHM decreases continuously during thermal healing (Fig. 1). In the case of natural samples, the defects induced disorder seems to be strongly connected to the thorium content.

In order to explore the complex structure of the SON68 nuclear glass, four simplified Eu^{3+} doped matrixes relative to the four main glass forming oxides have been studied by fluorescence line narrowing. The precise calculation of their crystal field parameters and the correlation with those of the nuclear glass allowed us the determination of the nature of the trivalent europium ion environments in a simplified SON68 type glass (Fig.2). The glass presents two sites for europium, the first one is a silicate one and the second is mixed aluminate and borate one. Both sites into the different gels are similar to the glass. The first site is mainly a silicated one with the presence of phosphate groups, and the second is an aluminated one with the possible presence of boron in the surrounding of the rare-earth.