

LETTER

Why natural monazite never becomes amorphous: Experimental evidence for alpha self-healing

**ANNE-MAGALI SEYDOUX-GUILLAUME^{1,2,*,*†}, XAVIER DESCHANELS³, CÉDRIC BAUMIER⁴,
STEFAN NEUMEIER⁵, WILLIAM JOHN WEBER^{6,7}, AND SYLVAIN PEUGET⁸**

¹CNRS, Université Clermont Auvergne, IRD, UJM, Laboratoire Magmas et Volcans, F-63000 Clermont-Ferrand, France

²University Lyon, UJM-Saint-Etienne, UCA, CNRS, IRD, LMV UMR 6524, F-42023 SAINT-ETIENNE, France

³ICSM, CEA, CNRS, ENSCM, University Montpellier, Marcoule, France

⁴CSNSM, JANNuS-Orsay/SCALP, University Paris-Sud, CNRS/IN2P3, Orsay, France

⁵Forschungszentrum Jülich GmbH, Institute of Energy and Climate Research, Nuclear Waste Management and Reactor Safety (IEK-6), D-52425 Jülich, Germany

⁶Department of Materials Science and Engineering, University of Tennessee, Knoxville, Tennessee 37996, U.S.A.

⁷Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, U.S.A.

⁸CEA, DEN, DE2D, SEVT, LMPA BP 17171, 30207 Bagnols-sur-Ceze Cedex, France

ABSTRACT

Monazite, a common accessory rare-earth orthophosphate mineral in the continental crust widely used in U-Pb geochronology, holds promise for (U-Th)/He thermochronology and for the immobilization of Pu and minor actinides (MA) coming from spent nuclear fuel reprocessing. Previous results obtained on natural and plutonium-doped monazite have demonstrated the ability of this structure to maintain a crystalline state despite high radiation damage levels. However, the low critical temperature (180 °C), above which amorphization cannot be achieved in natural monazite under ion irradiation, does not explain this old and unsolved paradox: why do natural monazites, independent of their geological history, remain crystalline even when they did not experience any thermal event that could heal the defects? This is what the present study aims to address. Synthetic polycrystals of LaPO₄-monazite were irradiated sequentially and simultaneously with α particles (He) and gold (Au) ions. Our results demonstrate experimentally for the first time in monazite, the existence of the defect recovery mechanism, called α -healing, acting in this structure due to electronic energy loss of α particles, which explains the absence of amorphization in natural monazite samples. This mechanism is critically important for monazite geo- and thermochronology and to design and predictively model the long-term behavior of ceramic matrices for nuclear waste conditioning.

Keywords: Monazite, α self-healing, in situ TEM irradiation, dual-ion beam irradiation, helium, geochronology, thermochronology, nuclear waste