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Experimental study of Pb diffusion in monazite by Pb^{2+} + $Th^{4+} \Leftrightarrow$ 2 Nd^{3+} exchange

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We report a new set of data on Pb diffusion in synthetic $NdPO_4$ monazite performed with the aim of constraining the Pb loss rate from this mineral during H.T. metamorphism events following crystallization. H.T. diffusion, such as dissolution-precipitation, is one of the possible processes able to change the isotopic contents of the monazite and consequently its radiogenic age. The present work follows those of Smith and Giletti (1997) and of Cherniak et al. (2004) that provide somewhat discrepant data on Pb diffusion, with respectively 180 $\pm$ 48 kJ.mol^{-1} and 592 $\pm$ 39 kJ.mol^{-1} for the activation energy E of the diffusion coefficient D = D_0exp(-E/RT). We synthetized $NdPO_4$ single crystals by the flux-growth method, with (-101) large faces ($\sim$1 mm^2). These surfaces were covered by a thin film ($\sim$100 nm thickness) of a solid solution S = ($NdPO_4$ 65% + $Pb_{0.5}Th_{0.5}PO_4$ 35%) by RF-sputtering. At the begining of diffusion anneal, the layer crystallizes with homogeneous thickness forming an epitaxial contact with the underlying crystal. This diffusant reservoir has the advantage of having chemical composition and structure very similar to the $NdPO_4$ crystals. The covered samples were enclosed in sealed Pt capsules filled with sintered powder pellets of S acting as buffer, avoiding direct contact with the specimen. Diffusion anneals were performed at 1300-1500˚C for durations ranging from several hours to several days. Resulting Pb and Th diffusion profiles were then analysed using both TEM and RBS, showing 50-500 nm characteristic lengths of diffusion. A TEM section perpendicular to the diffusion surface was prepared with the Focused Ion Beam (FIB) technique at the GFZ-Potsdam. It resulted that Pb and Th diffuse with the same rate from the diffusant layer toward the interior of the monazite crystal. This observa-

tion goes in favor of a coupled diffusion of Pb and Th exchanging by interdiffusion with Nd. RBS spectra obtained with the Van de Graaff accelerator at the GPS of the University of Paris VII were analyzed accounting for the fact that Pb and Th diffuse at the same rate. Our provisional set of data yields E = 404 $\pm$ 40 kJ.mol^{-1} and log D_0 = -6.63 $\pm$ 1.25 (D_0 in m^2.s^{-1}) for both Pb and Th. Our values are intermediate between the previously published data. For instance, when extrapolating at 700˚C and for a 100 Ma duration, the characteristic length of diffusion $(4Dt)^{1/2}$ is $\sim$1μm from data of the present study while it is $\sim$100 μm for Smith and Giletti's and $\sim$0.01 μm for Cherniak et al.'s ones.

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