Oxygen isotopes in uranium oxides from the front end of the nuclear fuel cycle

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Oxygen isotopes have the potential to trace the origin and way of transformations of uranium ore concentrates in the framework of nuclear forensics [1,2], but this is controversial [3]. To move forward, we explored the oxygen isotope evolutions of uranium oxides along their various synthesis paths. After purification, uranium in solution is precipitated into diuranates or peroxides (T~50°C) and calcinated into U_3O_8 (T~600-800°C). The crystal structure of these uranium oxides can be complex and usually contains multiple oxygen reservoirs. Also, atmospheric moisture is readily adsorbed on uranium oxide powders. These issues not only hamper a good reproducibility of $\delta^{18}O$ measurements and satisfactory oxygen yields after extractions but also make oxygen-isotope ratios difficult to interpret. Therefore there is a need for an improved protocol for oxygen extraction and isotope analysis as well as for a better understanding of where oxygen comes from in uranium oxides.

We developed a methodology for an accurate and reproducible determination of oxygen isotopes in uranium oxides adapted from the conventional BrF_5 method [4] and the laser one [5]. Our methodology ensure extraction yield near 100% and a good reproducibility in $\delta^{18}O$ measurement, better than 0.3‰, for a wide variety of uranium oxides: U_3O_8, UO_2, peroxides and diuranates. For a large set of samples of various geographical provenance, $\delta^{18}O$ values cover a large range between -10‰ and +10‰ with systematic features.

In order to mimic the various synthesis paths used in the front end of the nuclear fuel cycle, uranium oxides were synthetized in the laboratory using controlled T-fO_2-pH conditions. Both precipitated uranium oxides and associated aqueous solutions (initial and residual) have been sampled leading to the determination of empirical fractionation factors in the system U-H_2O-O_2.

[1] Fayek *et al.*(2011) *Ore Geology Reviews* **41**, 1-21. [2] Pajo *et al.*(2001b) *Fresenius J. Anal. Chem.* **371**, 348-352. [3] Plaue (2013) Ph.D Dissertation [4] Clayton & Mayeda (1963) *GCA* **27**, 43-52. [5] Sharp (1990) *GCA* **54**, 1353-1357.