Isotope release properties and modelling

The isotope release is known to be closely related to the materials micro and nano structure. In the case of diffusion-bound processes and if the target material is prepared in smaller particles the release of the same isotopes should consequently happen in a shorter time. Within ActILab this dependency was closely analysed experimentally with a variety of uranium carbide targets of different grain size and microstructure.

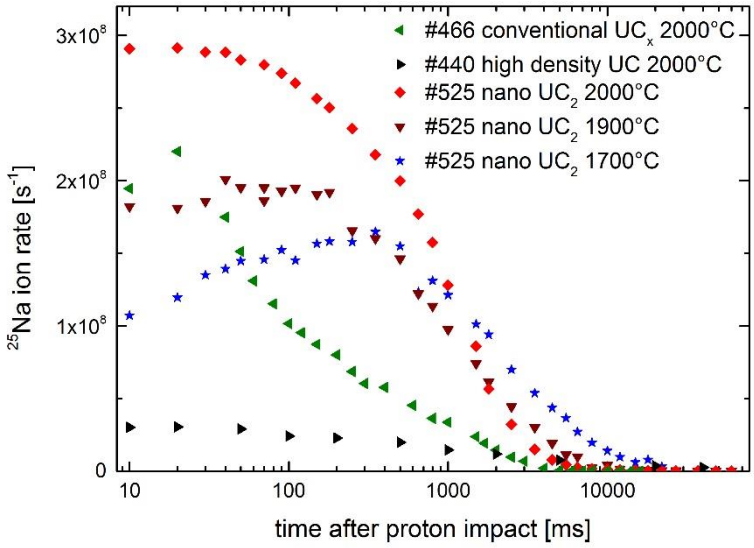

Figure 1: The release structure of ^{25}Na ions after a very short proton pulse at t=0 for different uranium carbide target materials and different material temperatures. Except for the clearly different overall yield (area under the release curve) the different release components are clearly visible that occur as a function of time.

Fig. 1 shows the release curves (isotope intensity after a proton pulse at t=0) for various target materials and temperatures. Obvious is the significantly increased yield from the nano-material based target #525, especially at temperatures above 2000°C. Within ActiLab it was shown that this kind of material withstands temperatures as high as 2100°C with very little effects of ageing, while even higher temperatures seem to be within reach and should be tested systematically in the future. The high temperature resistance has been arranged by the design of a special uranium carbide – carbon composite, where direct contact points between the nanometric uranium carbide particles are avoided within a refractory multi-walled carbon nanotube fiber matrix, which prevents sintering greatly.

The release time structure of nano UC_2 shows the same or even a higher fast release component but exhibits an additional component in the time domain between 100 ms and 100 s that is absent, strongly suppressed or distributed over much greater time scales in the case of conventional UC_x. Exploiting this additional release fraction of the produced radio-isotopes, total release efficiencies close to 1 were reached for isotopes of Na, K, Rb and Fr with half-lives of 1s or longer.

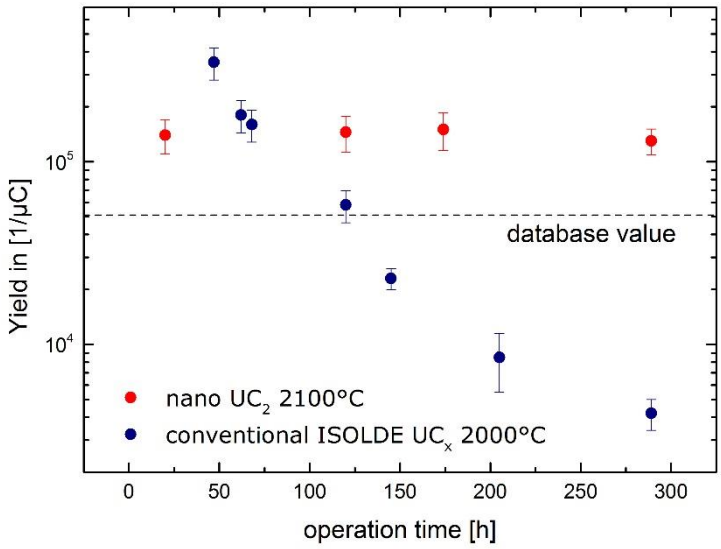

Figure 2: Evolution of exotic ^{30}Na yield while operation (irradiation). For conventional UC_x the yield drops by two orders of magnitude while in the case of nano UC_x it stays constant over an irradiation period of more than 10 days.

For the case of many exotic radioisotopes the beam intensity reduces over time more or less drastically, making the planning of an experimental campaign challenging. Fig. 2 shows this ageing effect as observed on conventional UC_x target materials (blue points) for short-lived ($T_{1/2}$ = 48 ms) in comparison to the novel ActILab nano UC_2 (red points). Over the time of a typical ISOLDE target life cycle (5-15 days) a drastic drop of isotope rates of two orders of magnitude has been observed for ^{30}Na from several conventional UC_x target units at ISOLDE. Assuming that losses are caused entirely by the growth of uranium carbide particles (sintering) and consequent decay of ^{30}Na during diffusion, a simple diffusion model (R. Kirchner, NIM B, **B70**, 186-199 (1992)) can be employed to relate every observed yield to a certain particle size. Since the temperature dependant diffusion constants *D(T)* of Na in UC and UC_2 is not known an assumption has to be made that can describe the initial crystallite size of both, the non-irradiated UC_x (see MS87) and the irradiated UC_x (see MS89) observed in synchrotron-based micro X-ray diffraction studies.

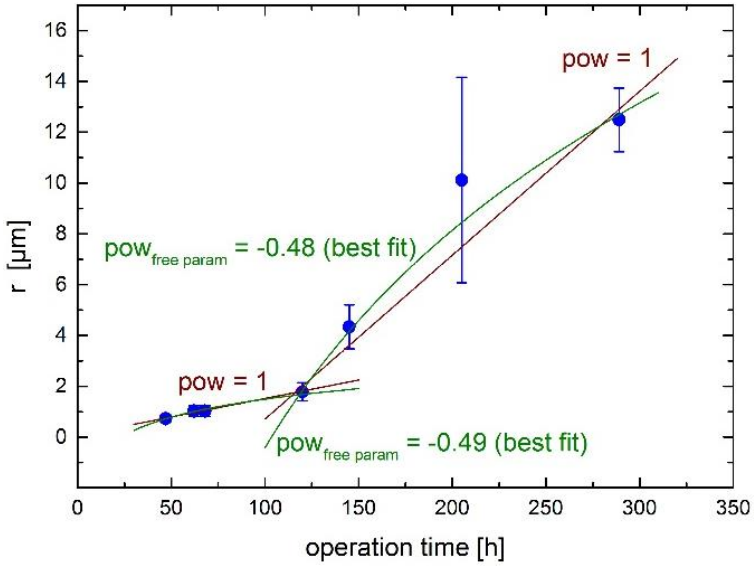

Figure 3: Conventional UC_x particle size evolution assuming that losses of ^{30}Na occur uniquely due to nuclear decay during diffusion and assuming a diffusion constant $D=6\cdot10^{-11}$ $cm^{-2}s$.
In the beginning sintering is kinematically hindered and accelerates once carbon diffusion causes a microscopically homogeneous carbon content.

The resulting particle size evolution is shown in Fig. 3. Two domains of particle growth rate can be found in this way, indicating two competing mechanisms of sintering. Within the ActILab project the origin was found to be the coexistence of UC and UC_2 phases that causes a kinematic limitation of sintering as long as microscopic gradients in the carbon content are present and loses its effect as carbon diffusion overtakes (see MS87).
This information has been taken into account when the new uranium carbide nano material was developed (see MS86) where sintering has been significantly reduced causing the yield of ^{30}Na to be stable over long periods of time, see red points in Fig. 3.