Altitudinal scaling of cosmogenic ^{3}He and ^{21}Ne in artificial quartz targets

PIETER VERMEESCH[1]*, HEINRICH BAUR[1], VERONIKA S.HEBER[1], FLORIAN KOBER[1], PETER OBERHOLZER[1], JOERG M. SCHAEFER[1], CHRISTIAN SCHLÜCHTER[2], STEFAN STRASKY[1] AND RAINER WIELER[1]

[1]IGMR, ETH-Zurich, Switzerland (*p.vermeesch@ucl.ac.uk)
[2]University of Bern, Switzerland

We measured cosmogenic ^{3}He and ^{21}Ne in artificial quartz targets after one year of exposure at mountain altitudes in the Swiss Alps. The targets were inconel steel tubes containing one kg of artificial quartz sand (250-500μm), degassed for one week at 700° C in vacuum prior to exposure. From August 2006 until August 2007, ten of these targets were exposed at five locations in Switzerland: Zürich (556m), Davos (1560m), Säntis (2502m), Jungfraujoch (3571m), and Monte Rosa (4554m). Additionally, a sixth set of two blank targets was kept in storage and effectively shielded from all cosmic ray exposure. Cosmogenic noble gases were measured at room temperature and at 700° C. Up to 9% of the cosmogenic ^{3}He was measured in the cold step, indicating that helium diffuses out of quartz at room temperature. The remaining ^{3}He was released at 700° C, as shown by a repeat measurement at 800° C for the Monte Rosa target, which yielded almost no additional cosmogenic neon. As expected, the Monte Rosa target contained the highest cosmogenic nuclide content, with $1.56 \pm 0.07 \times 10^{6}$ atoms of excess ^{3}He and $4.4 \pm 1.1 \times 10^{5}$ atoms of excess ^{21}Ne. After correction for non-atmospheric blanks, shielding (roof + container wall), tritiogenic helium and solar modulation (normalised to the average neutron flux over the past five solar cycles), our measurements yielded weighted mean production rates at sea level and high latitude of 111.4 ± 5.9 a/g/yr for ^{3}He and 15.8 ± 2.0 a/g/yr for ^{21}Ne (all errors are 2σ). These estimates are in good agreement with production rates derived from long-term exposure experiments at natural calibration sites. The main goal of the artificial target experiment was to determine the production rate attenuation length. Our best estimates for the ^{3}He and ^{21}Ne attenuation lengths are 134.6 ± 5.9 g/cm^{2} and 140 ± 28 g/cm^{2}, respectively, again agreeing very well with currently used scaling models. We conclude that TCN production rates are indeed proportional to neutron monitor count rates, and that ^{3}He and ^{21}Ne production rates follow the same altitudinal scaling relationships as the cosmogenic radionuclides.

Coupled chemistry-transport modeling of glass alteration using archaeological fractured samples

A. VERNEY-CARRON[1,2]*, S. GIN[1], P. FRUGIER[1] AND G. LIBOUREL[2]

[1]CEA DTCD/SECM/LCLT, Bagnols-sur-Cèze, France (*correspondence: verney@crpg.cnrs-nancy.fr)
[2]CRPG-CNRS, Vandoeuvre lès Nancy, France

To improve the confidence in glass alteration models (in the nuclear and in the natural fields), their long-term predictive capacity has to be validated. For this purpose, we studied archaeological glass blocks weathered for 1800 years in seawater, a known and stable environment. They also presented a good morphological analogy (fracturing) with vitrified nuclear waste.

We developed a specific geochemical model of alteration that we coupled with a transport model. The long-term predictive capacity was tested by comparing extended simulation results with the alteration products and kinetics.

Three steps describing the glass alteration process were implemented in the chemical model: (1) the formation of a hydrated glass by interdiffusion, whose kinetics are controlled by a pH- and temperature-dependent diffusion coefficient; (2) the dissolution of the hydrated glass, whose kinetics is based on an affinity law; (3) the precipitation of secondary phases when thermodynamic equilibrium is reached. The parameters of the model were measured by performing various laboratory experiments in different conditions (solution composition, pH, temperature, S/V ratio).

Then the model was coupled with diffusive transport in solution in order to simulate the alteration in cracks. Simulation results over 1800 years are consistent with archaeological glass block observations concerning (1) the nature of alteration products (hydrated glass, smectites, and carbonates) and (2) crack thicknesses (apparent alteration rates). These results not only improve the understanding of chemistry-transport coupling in fractures but also validate the confidence in glass alteration predictive models.