Experimental study of the breakdown of dolomite: Effects of grain size and aggregation on rate

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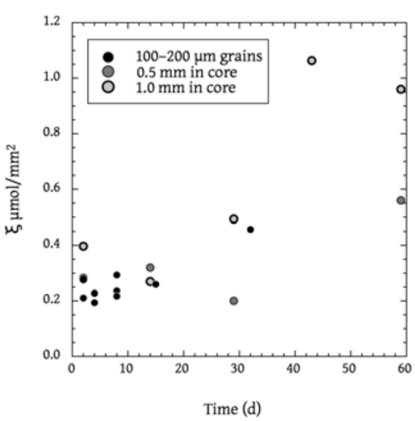
We compared the laboratory-determined reaction rate of mineral grains with that of rock samples in the dolomite– H_2O system. The rate of the breakdown reaction dolomite = calcite + periclase + CO_2 was measured experimentally at 700°C, 100MPa, with samples of dolomite ranging from 100–200 µm powders to rock cores having grains ~ 1 mm in diameter. Our experiments were conducted in cold-seal hydrothermal vessels over periods ranging from 2 to 60 d with the dolomite bathed in H_2O. Extents of reaction after experimentation were measured with electron probe x-ray micrographs of core sections and grain mounts. We found that the core samples reacted more slowly than the powder, as anticipated from the requirement of transport of CO_2 and H_2O along grain boundaries. Our studies showed that the extent of reaction depended on the square root of time, suggesting a diffusion-controlled process. However, the grain experiments also showed a rate depending on the square-root of time, and when the extents of reaction were normalized to the surface area, the rates in the grain and rock-core experiments were the same (see Figure). This suggests that there is little discernible difference in rates whether the dolomite is as individual grains or a rock. It appears that transport of CO_2 and H_2O is fast along grain boundaries in the rock core and that the reaction rate is controlled by the interdiffusion of CO_2 and H_2O through the mantle of reaction products.



Direct search for primordial ^{244}Pu

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Long-lived radionuclides provide important knowledge on the time scales of processes during the formation of the solar system. Because of its complete suppression of molecular background accelerator mass spectrometry is the most sensitive and unambiguous method to determine traces of these nuclides directly.

At the tandem accelerator of the Maier-Leibnitz-Laboratory in Garching a time-of-flight sytem has been set up for the detection of actinides. This setup was used for a highly sensitive search for ^{244}Pu ($T_{1/2}$ = 81.2 Ma), a nuclide which is naturally produced exclusively during the r-process in explosive stellar nucleosynthesis. By measuring the current abundance of ^{244}Pu and comparing it to the longer lived ^{238}U one may deduce the portion of r-process material from the last nucleosynthesis process that has contributed to the protosolar cloud before the formation of the Earth. But because of the short halflife compared to the solar age its decay has reduced its abundance probably almost completely. Indirect evidence for the existence of ^{244}Pu on Earth has been deduced from heavy Xenon isotopes generated by its fission in the past. Hoffman *et al*. [1] succeeded in detecting small amounts of natural ^{244}Pu in the rare-earth rich mineral Bastnaesite directly. But their results were never confirmed.

Our approach to measure Plutonium in the same mineral was completely free of background events and can exclude an abundance in the order of 2500 atoms ^{244}Pu per gram Bastnaesite as observed by D. Hoffman and her collaborators.

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[1] Hoffman *et al*. (1971) *Nature* **234**, 132-134.