U-Pb dating of zircon by excimer laser ablation MC-ICPMS

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Generalised adoption of U-Pb dating by laser ablation(LA)-ICPMS has been hampered by elemental and isotopic fractionation which lead to relatively poor reproducibility and accuracy. In order to understand the fractionation phenomena a systematic study of the main analytical parameters affecting elemental and isotopic ratios of interest was carried out with an excimer laser (193 nm) coupled to a IsoProbe instrument. Several hundred single-spot analyses of fragments of a macroscopic zircon crystal selected for their low magnetic susceptibility, limpidity and lack of imperfections, reveal that the ratios of interest (including $^{238}U/^{235}U$) are afftected to different degrees by laser fluence and beam diameter, repetition rate, focus position of the laser on the sample surface, carrier gas flow rate and surface roughness. In addition, the influence of these parameters depends on whether the ablation medium is Ar or He. A comparison of the results for zircon with those for NIST613 glass indicates that the glass is not an appropriate standard for zircon analysis, and further reveals that some of the effects are material independent and must result from the ablation-transport-ionisation processes.The main conclusions, briefly stated, are that the parameter that most affects fractionation is carrier gas flow rate, that ablation under He yields significantly better precision, that $^{207}Pb/^{206}Pb$ and Pb/U generally display opposite behaviour and that $^{208}Pb/^{232}Th$ is highly fractionated for accurate values of Pb/U. Therefore, analytical conditions yielding accurate Pb/U and Pb/Th ages are incompatible. Based on these data, a procedure using an external zircon standard is proposed that yields the following concordia ages for samples previously dated by ID-TIMS (in brackets): 1138±7 Ma (1143±1); 1779±2 Ma (1771±2); 1885±5 Ma (1884±2); 2789±3 Ma (2780+3/-2) and 725+7/-10 Ma (727±1). Between 6 and 14 grains were analysed with a beam diameter of 80 micra and at a fluence of 10J/cm^2. All ages are accurate to better than 0.5%. Overall, 50% of the analyses are concordant within error and an additional 30% are less than 1% discordant but lie mostly above concordia, indicating that a degree of Pb enhancement over U is not accounted for in the procedure. Analyses of the same macroscopic zircon crystal by rastering a 35 _m beam, are more precise but show higher Pb/U variability (greater overall discordance) when compared to single-spot analyses.

The influence of temperature on ion adsorption phenomena

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The adsorption of ionic species by mineral surfaces influences many important geochemical processes including colloidal transport and reactivity, mineral dissolution and precipitation reactions and corrosion processes. However, until recently experimental studies over a wide temperature range that includes the hydrothermal regime (above 100ºC) were virtually nonexistent. For the past ten years we have been employing stirred hydrogen electrode concentration cells (SHECC's) to measure ion adsorption by various metal oxides to 290ºC, with the relatively inert oxide rutile (TiO_2) receiving the most attention to date, and Fe_3O_4 and ZrO_2 receiving more limited attention. Ionic species studied have included Na, Rb, TMA, Ca, Sr, Zn, Co, Nd, Cl, and oxalate.

Surface complexation models are being used to rationalize these results. In particular, we have extended the MUltiSIte Complexation (MUSIC) Model of Hiemstra, van Riemsdijk, and coworkers to 300ºC. A particular advantage of this approach is that spectroscopically identifiable adsorption complexes can be explicitly included in the modeling efforts. This presentation will summarize the results of our extended temperature range ion adsorption studies and modeling efforts, as well as the potential implications of these results to the transport of pollutants in environments where wide temperature ranges are expected to occur such as in the vicinity of high-level nuclear waste repositories.