The degree of metamictization in zircon: 
a Raman spectroscopic study

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Abstract: A series of natural zircon samples representing various degrees of metamictization were investigated by Raman microprobe (RMP) analysis. We found systematic changes in wavenumbers and half-widths of the Raman bands, caused by increasing irregularities of bond-lengths and bond-angles and a general breaking-up of the structure as a result of metamictization. Therefore, Raman spectroscopy can be used to measure the crystallinity of zircons. The half-width of the antisymmetric stretching vibration band \((6_{1g})\) of the SiO\(_4\) tetrahedra, which has a frequency of about 1007 cm\(^{-1}\) in well-crystallized and 1000 to 955 cm\(^{-1}\) in metamict zircons, is most suitable for estimating the degree of metamictization: Its value increases from about 5 cm\(^{-1}\) in well crystallized samples up to 30–55 cm\(^{-1}\) in highly metamict, X-ray-amorphous zircons, and is strongly dependent on the degree of lattice destruction by metamictization. In contrast, the Raman parameters seem to be almost uninfluenced by chemical variations. The potential value and advantages of such RMP measurements, especially in radiometric age determinations, are discussed in the light of other methods.

Key-words: zircon, metamictization, Raman microprobe analysis.

Introduction

Zircon is an important mineral for radiometric age determination, as it occurs widely as an accessory mineral, has a high melting temperature, and shows considerable stability against chemical and physical influences. Zircons incorporate trace amounts of a number of elements during crystallization. For age determination the most important is uranium, which can substitute for Zr\(^{4+}\) and decays to lead over geological time.

With time, the zircon structure can break down to the metamict state. Metamictized zircons are characterized by poor crystallinity of the lattice, that is, loss of medium range ordering. Several papers related to the hypotheses and models for the metamict state of minerals have appeared (e.g., Lipova, 1966; Graham & Thornber, 1974; Ewing, 1975; Ringwood, 1985; Caruba et al., 1985; Aines & Rossman, 1986; Chakoumakos et al., 1987; Lumpkin & Ewing, 1988; Hawthorne et al., 1991). Metamictization results from radiation damage to the lattice caused by \(\alpha\) particles originating from the decay of uranium and thorium (Murakami et al., 1986). In general, the degree of destruction of the zircon lattice can be interpreted as a function of the intensity of the \(\alpha\)-radiation and the duration of its influence (Kulp et al., 1952; Deliens et al., 1977).

In addition to physical and structural changes, metamict zircons show a reduced chemical stability (Sommerauer, 1976), which often results in considerable loss of radiogenic lead (Gentry et al., 1982; Bogomolov & Morozova, 1988). The loss of Pb in metamict zircon was investigated experimentally by Pidgeon et al. (1966) and Tole (1985).

DOI:10.1127/ejm/7/3/0471
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0935-1221/95/0007-0471 $ 2.00