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Partitioning of U and Th between low-Ca-Clinopyroxenes and Anhydrous Silicate Melts: Consequences for the Generation of U-Th-Disequilibrium During Mantle Melting

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Secular disequilibrium in the U-decay series (i.e. excess of ^{230}Th over ^{238}U and ^{226}Ra over ^{230}Th) is frequently used to decipher the nature of mantle melting processes. Disequilibrium in the U decay-series elements in mantle partial melts requires the presence of a solid phase on the mantle solidus which exhibits D_U/D_{Th} and D_{Th}/D_{Ra} both greater than 1.0 and a bulk partition coefficient for U of the same order as the threshold porosity at which melt is extracted (e.g. Elliott 1997), which is likely to be greater than 10^{-5} (McKenzie 1989). Previous experimental studies show that olivine and orthopyroxene have very low absolute D_U values of about 10^{-5}, while clinopyroxene has $D_U/D_{Th} < 1$. Therefore according to available data these phases alone cannot contribute to the generation of U decay-series disequilibria. In contrast, the pyropic garnets typical of mantle peridotite yield high D_U/D_{Th} of about ~6 and absolute D_U values similar to those of clinopyroxene (e.g. Beattie 1993). For this reason the generation of U decay-series disequilibria is almost universally attributed to the initiation of mantle melting in the garnet lherzolite stability field, rather than occurring entirely in the spinel-lherzolite stability field.

A major shortcoming of most of the available experimental partitioning data used in modelling U decay-series disequilibria is that they refer to low pressure calcic clinopyroxene (> 0.8 Ca atoms per formula unit) and do not take into account the clinopyroxene composition on the mantle solidus nor its changing behaviour with P and T. Predictions from recently developed models for trace element partitioning and preliminary experimental results (Blundy & Wood 1994, Wood et al. 1999) suggest, however, that for subcalcic (< 0.6 Ca atoms per formula unit) aluminous clinopyroxene now known to characterize the mantle solidus, D_U/D_{Th} is > 1 at pressures above 1.5 GPa. Incorporating their partitioning data in dynamic mantle melting calculations Wood et al. (1999) were able to show that $^{230}Th/^{238}U$ activity ratios of up to about 1.23 in the liquid are consistent with melting in the spinel lherzolite stability field. However, due to the limited availability of experimental partitioning data for Ca-poor clinopyroxene it is still a matter of debate whether excess of ^{230}Th over ^{238}U can be generated entirely in the spinel lherzolite stability field.

To test the predictions of Wood et al. (1999) and to extend the existing database for partitioning of U and Th and of trace elements between Ca-poor clinopyroxene and anhydrous silicate melt, we performed partitioning experiments in the NCMAS simplified lherzolite model system of Walter & Presnall (1994) over a wide P and T range. Clinopyroxene and melt compositions of experimental run products are in good agreement with the compositions given by Walter & Presnall (1994). The Ca-contents of the experimentally produced clinopyroxenes vary from 13.79 to 17.65 weight-% CaO, corresponding to a variation in x_{Ca} on the M2-site from 0.521 to 0.657 cations per formula unit, respectively. Using the model of Blundy & Wood this decrease in Ca-content leads to a decrease in r_0 -values for the M2-site from 0.0992 nm to 0.0984 nm. Measured D_U/D_{Th}-ratios are found to be strongly correlated to x_{Ca} and vary from 1.08 ±0.06 at x_{Ca} = 0.657 to values as high as 1.34 ±0.12 at x_{Ca} = 0.521 and are in good agreement with the predicted values from Wood et al. (1999). These results confirm the model predictions of Wood et al. (1999) that extensive ^{230}Th-excess can be generated at much shallower depths than previously thought well within the spinel lherzolite stability field and that residual garnet is not necessarily required to generate ^{230}Th-excess in mantle melts.

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