Helium solubility in carbonate liquids: Potential for generating high ^{3}He/U mantle

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Carbonatitic melts are known to exist in the upper mantle, by direct observation of carbonate phases in residual xenoliths or diamonds, and analyses of ephemeral metasomatism. Experimental work has shown that a range of carbonatite compositions are stable at upper, and possibly lower, mantle conditions.

Geochemists have argued that metasomatic/carbonatitic agents decouple noble gases from lithophile element isotope systematics in 'plumes' (e.g. Loihi; Kerguelen; Samoa; Easter Island; Cameroon); in continental rifts; in subduction related massifs (Horroman), and generation of MORBs. Despite known carbonatitic melts in the upper mantle (and possibly lower mantle), and their alleged involvement in noble gas systematics, there is no experimental data demonstrating that noble gases preferentially partition into a carbonate phase.

We have measured the solubility and diffusivity of He in $CaK_2(CO_3)_2$ and K_2CO_3 liquids at 1 bar; He is less soluble in carbonate than in basaltic liquids: $S^{He}_{carbonate}$ =(1-2) x10^{-8} mol g^{-1} (increasing with Ca content); $S^{He}_{tholeiite}$ = 2.5 x10^{-8} mol g^{-1} [1]. He diffusion through carbonates is rapid (D_o = 8.5 cm^2 s^{-1} and E_a = 50 KJ mol^{-1}). Further measurements of He+Ar solubilities are planned in a range of carbonate compositions.

Due to high U solubilities in carbonate liquids [2], mantle metasomatism by carbonatatic melts could increase ^{3}He/U of mantle remaining after metasomatism, thus, over time, creating high $^3He/^4He$ ratio mantle, as sampled by many oceanic islands.

References

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Evolution of helium isotopes in the Earth's mantle

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The presence of primordial ^{3}He in ocean island basalts (OIB) has been considered the primary evidence for the contribution of a primitive, undegassed reservoir in the lower mantle to upwelling plumes. A new global data compilation relating helium isotopes of OIB and mid-ocean ridge basalts (MORB) to Sr-Nd-Pb isotopes and trace element abundances yields new insights into mantle dynamics. The compilation is based on data from the online GEOROC and PetDB databases. Oceanic islands are divided into groups based on the highest $^3He/^4He$ ratios measured in mineral separates: (1) "low $^3He/^4He$" ($^3He/^4He$ <7 R_A); (2) "MORB-like $^3He/^4He$" (8±1 R_A); (3) "moderately high $^3He/^4He$" (9-15 R_A); (4) "high $^3He/^4He$" (>15 R_A). These designations enable us to follow the geochemical characteristics of mantle plume sources based on their $^3He/^4He$ ratios despite the global paucity of combined He-Sr-Nd-Pb isotope data on individual samples. The compositions of the four groups of OIB are not distributed randomly within the global range but show the following systematics: (i) lower $^{143}Nd/^{144}Nd$ ratios are associated with lower $^3He/^4He$, (ii) for a constant $^{143}Nd/^{144}Nd$ ratio, higher $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, $^{208}Pb/^{204}Pb$ ratios are associated with lower $^3He/^4He$, and (iii) low $^3He/^4He$ ratios are accompanied by high Th, U abundances independent of partial melting effects.

The new compilation shows there is a global relationship between the $^3He/^4He$ groups and Th+U contents of OIB, (with only Samoa as an exception). Thus, $^3He/^4He$ in OIB appear to reflect the production rates of ^{4}He from recycled oceanic crust plus sediment variably enriched in (Th+U) in plume sources. The range of ^{3}He contents in plume sources must be small compared to that of (Th+U) in order for the correlation to persist. OIB displaying the strongest primordial $^3He/^4He$ signal are chemically and isotopically (i.e. Sr, Nd) most like mid-ocean ridge basalts, indicating a common history over geological time. We show through modeling that helium isotopes in the mantle can be explained through continuous, incomplete degassing by continent and ocean crust formation. It appears that the high $^3He/^4He$ component in plume sources does not represent unmelted primitive mantle but rather "old" depleted mantle, isolated from convection and upper mantle degassing for 1-2 billion years.