H_2CO_3(s): A new candidate for CO_2 capture and sequestration

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To reduce the magnitude of anthropogenic global climate change it will be necessary to remove CO_2(g) from the effluent streams of power plants (and possibly directly from the atmosphere as well) and to then sequester it as either a condensed phase or in solutions and/or compounds produced by chemical reaction. A major difficulty in achieving this goal arises from the very weak acidity of CO_2(g), causing it to react only incompletely with weak bases, forming complexes which readily decompose at high temperatures. Reaction with strong bases yields more stable complexes but strong bases are more costly and massive amounts of chemical products would still need to be sequestered. When gas-phase CO_2 reacts with the weak base water, or when bicarbonate reacts with acid, an unstable product "H_2CO_3" can initially be formed. Although the monomeric form of this compound is highly unstable, its oligomers are much more stable due to H-bonding, with free energies (per H_2CO_3 unit) for the larger oligomers approaching that for the gas-phase combination of CO_2 and H_2O. Although it is still thermodynamically slightly unstable, a oligomeric or solid form of H_2CO_3 may well be kinetically stable, particularly in the absence of H_2O. This indicates that it may be possible to capture gas-phase CO_2 directly, using only cheap and abundant H_2O, and to store it in the form of a solid or an oligomer. These conclusions are based on quantum computations which faithfully reproduce the structures and stabilities of H_2CO_3 oligomers and HCO_3^- dimers. Methods for producing and characterizing H_2CO_3 oligomers to establish their stability are discussed.

Hafnium-Tungsten chronometry of the eucrite parent body

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There is a debate about the timescales of accretion and differentiation of the eucrite parent body (EPB). Based on Al-Mg systematics, some have argued that the EPB accreted within less than ~1.5 Ma [1], whereas others, based on Sr isotope evidence, stated that accretion must have been later than ~3 Ma [2]. To better constrain the chronology of the EPB, we applied the extinct ^{182}Hf-^{182}W system combined with precise Hf, U, and W concentration measurements to a comprehensive set of eucrites, including both basaltic and, for the first time, cumulate lithologies.

Our new data reveal that in contrast to an earlier report [3], the Hf-W data for eucrites do not define an isochron but, at least in part, reflect contamination of some samples with W having a terrestrial isotopic composition. If the samples that are clearly contaminated are excluded from the regression, no statistically meaningful isochron is obtained for the basaltic eucrites. These have ε^{182}W values between ~20 and ~30 and Hf/W ratio ranging from 20 to 35. The measured Hf/W of the basaltic eucrites are similar to their source Hf/W, which are calculated based on their measured U/W ratios. This is consistent with the Nd-Hf isotope systematics of eucrites that suggest derivation as large degree melts from a source with chondritic abundances of refractory lithophile elements [4]. Therefore, the Hf-W systematics of basaltic eucrites might reflect that of their sources, such that core formation ages can be estimated. The two-stage model ages for the individual samples range from ~1 Ma up to ~4 Ma after CAI formation and are consistent with the ~4566.5 Ma age obtained from the ^{53}Mn-^{53}Cr whole rock isochron (recalculated from [5] relative to angrite D'Orbigny) and with Mg isotope evidence for differentiation of the EPB at ~3 Ma after CAI formation.

Cumulate eucrites, in spite of having highly variable Hf/W ratios, are characterized by constant ε^{182}W values of ~18, indicating that their different Hf/W ratios were established later than ~60 Ma after CAI formation, probably related to the presence of ilmenite in the cumulate eucrite source. This and the >60 Ma age is consistent with results from ^{147}Sm-^{143}Nd and ^{176}Lu-^{177}Hf whole-rock isochrons corresponding to an age of ~100 Ma after CAI formation [4].

[1] Bizzarro *et al.* (2005) *ApJ* **632**, L41-L44. [2] Halliday & Porcelli (2001) *EPSL* **192**, 545-559. [3] Quitté *et al.* (2000) *EPSL* **184**, 83-94. [4] Blichert-Toft *et al.* (2002) *EPSL* **204**, 167-181. [5] Lugmair & Shukolyukov (1998) *GCA* **62**, 2863-2886.