THE PROBLEM OF THE ORIGIN OF ^{16}O ANOMALIES IN CARBONACEOUS CHONDRITES. D.A. Wark[1] and R.D. Brown[2]. (1) Lunar and Planetary Lab., University of Arizona, Tucson, AZ 85721 and (2) Dept. of Chemistry, Monash University, Clayton, Vic., Australia.

The anomalous excess of ^{16}O discovered by Clayton and co-workers [1] in refractory inclusions of carbonaceous chondrites remains a major problem in meteoritics. Explanations in terms of nucleosynthesis, either by the introduction of supernova ejecta containing refractory ^{16}O-rich grains [2] or by proton irradiations within the solar nebula [3], suffer from the fact that other nuclides expected to be produced (or destroyed) along with the ^{16}O are either not anomalous [4] or are at best imperfectly correlated with ^{16}O anomalies [5].

In fact, the most clear-cut correlation shown by excess ^{16}O abundances is with the refractoriness of the material e.g. in Allende, the ^{16}O enrichments (rel. to SMOW) of refractory Ca-Al-rich inclusions (CAI's) are ~ 4%, while those of less refractory, fine-grained and amoeboid olivine aggregates are 0.5-1.5%, those of olivine chondrules are 0.5%, and the volatile-rich matrix is actually depleted in ^{16}O by 0.2% [1].

Molini-Velsko *et al.* [6] made the extremely important observation that "Oxygen isotopes(of Ca-Al-rich materials being evaporated at high temperatures in solar furnaces) seem to have exchanged with oxygen in the ambient atmosphere of the solar furnace". This implies that, for CAI's that are evaporative residues, the O isotopic composition of the parental solids was relatively unimportant and the enrichment of ^{16}O in the CAI was controlled largely by the local abundance of ^{16}O in the nebular gas of the CAI formation region. This would of course also be true for CAI condensates.

Thiemens and Heidenreich [7] have shown that non-mass-dependent isotopic fractionations of oxygen can be produced in gases by photo-chemical mechanisms. It has therefore been suggested that ^{16}O enrichments might have been produced in the nebula by UV from T Tauri solar flares. Another chemical kinetic mechanism for producing ^{16}O enrichments had previously been discovered by Arrhenius and McCrumb [8] and it is this which we investigate qualitatively below. The mechanism utilizes the fortuitous pre-dissociation of the $^{12}C^{16}O$ molecule (but not $^{13}C^{16}O$ or $^{12}C^{17,18}O$) by hydrogen Lyman α UV photons. Our preliminary calculations suggest that the mechanism would not produce significant isotopic fractionation in low density gas because the short (~100 nanosec.) mean lifetime of the excited $^{12}C^{16}O^*$ molecules would allow most of them to relax before they could collide with another molecule and dissociate. However, at particle densities of more than $\sim 10^{15}$ cm^{-3} (which are equivalent to plausible solar nebula pressures of $\sim 10^{-4}$ - 10^{-5} atm.) the dissociation rate could be significant and the mechanism may therefore have operated in the solar nebula. Excited CO^* would have been most likely to collide in the nebula with H_2 and He. Collisions with H_2 would probably have resulted in the following exothermic reaction, assuming that the (presently unknown) activation energy is not excessive:-

$$^{12}C^{16}O^* + H_2 \rightarrow {}^{12}CH + {}^{16}OH$$

Both of the product radicals CH and OH are very reactive, and the reaction $^{16}OH + H_2 \rightarrow H_2{}^{16}O + H$ would have created a reservoir of H_2O molecules showing ^{16}O enrichment, provided that the rate of this reaction exceeded the rate of oxygen isotopic re-equilibration between CO and H_2O.

If the temperature were sufficiently high (say, 1300K) for there to have been significant amounts of Ca, Al, Ti, Mg, etc. in the gas, then reactions of the type:-

$M + H_2{}^{16}O \rightarrow M^{16}O + H_2$ (where M = metal atom)

would have transfered a ^{16}O enrichment from the H_2O reservoir into stable refractory oxides, provided that the rate of this reaction exceeded the rate of reaction of metal with de-enriched CO. Both of the above provisos seem plausible in view of the high stability and relative inertness of the CO molecule. In this way, therefore, it appears likely that in high temperature regions of the protosolar nebula exposed to a sufficient flux of Lyman α, ^{16}O-enriched H_2O molecules and ^{16}O-rich grains of Ca, Al, Ti, etc. oxides may have formed along with a de-enrichment of CO in ^{16}O.

Reactions of the ^{12}CH radical might also provide an explanation for the greater enrichments of ^{12}C that are observed in refractory carbonaceous polymers and hydrocarbons than in amino acids and inorganic carbonates in the carbonaceous chondrites [9].

Two phenomena in the solar nebula associated with the strong UV fluxes and high temperatures required by the Arrhenius-McCrumb and Thiemens-Heidenreich mechanisms were: (1) solar flares during the T Tauri phase of solar evolution, and (2) H recombination in the accretionary post-shock regions of the protosolar nebula. Because the chondrites appear to have condensed from, and accreted gently in the presence of, a gas, it is generally presumed that the chondrites formed in the 10^5 y before the start of the sun's T Tauri stage (because T Tauri stars have optically-thin envelopes containing little gas and dust). However, T Tauri flares cannot be completely excluded because (1) T Tauri flares may have taken a long time to blow out the gas and dust, and (2) a lot of gas and dust might have remained in an ecliptic disc.

To sum up, the imperfect correlation of ^{16}O excesses with nucleogenic isotopic anomalies, and the evidence from solar furnace experiments that oxygen isotopic enrichment occurred at the time of CAI formation, strongly suggest that the ^{16}O effects in Allende were due to high-temperature, non-nuclear processes in the solar nebula. If one or other (or both) of the processes suggested by Arrhenius and McCrumb or Thiemens and Heidenreich were responsible, then the most plausible nebular location was probably near the accretion shock regions.

References: [1]Clayton R. et al. (1973) Science 182, 485; Clayton R. et al. (1977) EPSL 34, 209. [2]Dearborn and Boynton (1981) Meteoritics 16, 306; Clayton D. (1982) Qtly. J.R. Astr. Soc. 23, 174. [3]Lee (1978) Ap.J. 224, 217. [4]Phinney et al. (1979) Proc. LPSC 10th, 885. [5]Niederer et al. (1980) Ap.J. 240, L37; Niemeyer and Lugmair (1980) Meteoritics 15, 341; Wasserburg and Papanastassiou (1982) in "Essays in Nucl. Astrophys.", eds. Barnes, Clayton and Schramm, Cambridge U.P. [6]Molini-Velsko et al. (1983) abstract, Met. Soc. Mtg., Mainz. [7]Thiemens and Heidenreich (1983) Science 219, 1073. [8]Arrhenius et al. (1980) LPS XI, 34. [9]Yuen et al. (1983) LPS XIV, 875; Clayton R. (1963) Science 140, 192.