ACTINIDE CHEMISTRY IN CHONDRITES, M.T. Murrell and D.S. Burnett, California Institute of Technology, Div. of Geol and Planet. Sciences, Pasadena, CA 91125.

The solar system Pu/U ratio is an important quantity for cosmochronology. Although more data are needed, it appears that the Pu/Nd value in many meteorites, even highly differentiated objects such as Angra dos Reis, is essentially constant at 1.5×10^{-4} (by weight) (1, 2). This implies that the solar system (chondritic) Pu/U can be indirectly determined by multiplying the apparently ubiquitous Pu/Nd by the carbonaceous chondrite (Nd/U) leading to Pu/U = 0.005. However, we believe that, if at all possible, the solar system Pu/U should be determined directly from data on chondritic meteorites. Total chondrite ^{244}Pu fission Xe measurements are difficult, but precise data for chondrites has been obtained on phosphate separates (3, 4, 5). Although a strong case can be made for Pu/U enrichment in whitlockite (6, 7), it is important to consider whether "fractionation corrections" (7) might be applied or, alternatively, whether all meteoritic whitlockite may have fractionated Pu and U to the same degree (5), giving chronological significance to the relative Pu/U for different chondrites. Interestingly, the whitlockite Pu/U for a variety of chondrites shows an order of magnitude variation (5). The highest ratio (0.12) was observed in Nadiabondi (H5). Through the cooperation of P. Pellas and G. J. Wasserburg, we obtained samples of this meteorite for U fission track radiography (8) to see if we could provide insights into the high Pu/U in Nadiabondi whitlockite and into chondrite actinide chemistry in general. These measurements provide a quantitative image of the U distribution with ppb sensitivity and ~20 micron resolution. The basic data are summarized in Table 1.

The phosphate abundance in Nadiabondi is variable but averages ~0.5% (weight), with ~85% whitlockite. Both apatite and whitlockite are, in most cases, observed to be associated with metal or troilite grains. Microprobe analysis of apatite indicates a high F/Cl with ~3% F and only 0.6% Cl. This is unusual as chlorapatite is most often found in chondrites. As was expected, apatite has the highest U content (2.2 ppm) of any phase in Nadiabondi; our result agrees well with the value of 2.1 ppm U obtained by Pellas and Storzer (9). The U content of Nadiabondi whitlockite is low relative to other chondrites, 49±2 ppb. This result is in excellent agreement with the value of 52 ppb obtained by isotopic dilution on a ~100% whitlockite separate from Nadiabondi (10). We found a whitlockite grain within a chondrule which had a higher U content of 95±13 ppb (not included in Table 1). This value is more like the 80 ppb U result reported by Pellas et al. (5) for Nadiabondi whitlockite. Random strips of our sections yield a typical chondritic whole rock U content of 11 ppb. The contribution from the Ca-phosphates to this whole rock U value is at most 20%, the remainder of the U lies elsewhere. Nadiabondi has distinct chondrules, the majority of which contain variable amounts of Na-Al-Ca-rich interstitial material, probably devitrified glass. In many cases this "glass" also contains numerous FeNi, troilite, and/or chromite grains. The glass is found to contain substantial amounts of U (Table 1). The U content of the glass (measured in >35 micron regions) varies by as much as a factor of two between different chondrules. The exposed surface area of glass is variable among chondrules, but many chondrules which are enriched in glass have high U contents; for example, one barred olivine chondrule had an average U content of 100 ppb. We also find U to be enriched (Table 1) in two angular fragments (~200μ) of approximately pyroxene composition (normative: $En_{65}Fs_{10}Wo_{15}$). It is possible that these objects are fragments of crypotocrystalline chondrules, as it is very unusual to find such high U in pyroxene. Crozaz (11) has reported ~300 ppb U in a phase of clinopyroxene composition from Bremervorde (H3). In Nadiabondi, the majority of the U appears to lie in "glass" in chondrules and to some extent in a "pyroxene" phase. The high Pu/U for Nadiabondi whitlockite is mainly due to the U being located in other phases, although the Pu content is also 25-50% lower than that of St. Severin. It seems likely that equilibration (by solid state diffusion?) is slower for U than Pu. It then follows that Pu/U in Ca-phosphates is not entirely governed by radioactive decay, but also depends on different rates of actinide equilibration.

The available information suggests a tentative model for ordinary chondrite actinide chemistry. The model clearly needs, but is susceptible to, many additional experimental tests. The chemistry of the actinides and lanthanides in ordinary chondrites is closely coupled to that of P. The close association of phosphates in essentially all chondrites with metal and troilite grains (including type 3, Conca and Woolum, private communication), and the presence of schreibersite in unequilibrated chondrites (e.g. 12). suggest that unmetamorphosed chondritic material contains P bound in metal or sulfide phases (schreibersite?), with phosphates forming subsequently by oxidation during metamorphism. The Nadiabondi data suggest that the "primary" actinide reservoir, as far as ordinary chondrites are concerned, is chondrule glass. Thus, the actinides and P "start out" initially in different phases. It is quite possible (but not proven (7, 8)) that U is not equilibrated in any ordinary chondrite, but, at equilibrium, would be totally concentrated in Ca-phosphates. Thus, the effective equilibration lifetime (τ_4) for U in whitlockite, is significantly longer than the ^{244}Pu radioactive decay lifetime which is comparable to the cooling time (e.g. 9). It also appears that Pu (and light lanthanides (10)) concentrate in phosphates more rapidly ($\tau_3 \ll \tau_4$), and this may be the primary reason for the high Pu/U ratios in chondritic whitlockites (apatites are not easily understood from this point of view). Figure 1 illustrates this model schematically. Pu and U concentrations in whitlockite initially increase with time (t=0 is time of phosphate formation) due to migration from glass to phosphate. The Pu concentration reaches a maximum and begins to decrease due to ^{244}Pu decay ($t_{1/2}$=82 m.y.). The important qualitative point from this figure is that the whitlockite Pu/U vs. time curve is not a simple exponential (compare the

^{244}Pu decay line in Fig. 1). The functional forms and parameters used in Figure 1 are arbitrary, but the figure does qualitatively represent the inferences discussed above. In reality the Pu/U ratio of each phosphate grain may depend in a complex manner on many effects, e.g. the proximity of chondrule glass, temperature of metamorphism, etc.. Consequently, we feel that it will be very difficult to obtain quantitative chronological information from chondritic whitlockite Pu/U ratios (from tracks or fission Xe). It should be noted that the "adjacent grain" fission track studies of Pellas and Storzer (e.g. 9) are based on Pu, not Pu/U, and just require that the whitlockite Pu concentrations vary only due to radioactive decay. This will probably be valid depending on the actual time scales (τ_3) for incorporation of Pu into phosphates. However, given the variety of other U and, possibly, Pu reservoirs in Nadiabondi, adjacent grain fission track data on this meteorite require cautious interpretation.

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Table 1. U Distribution in Nadiabondi

phase	# of grains or chondrules	U (ppb)*	range (ppb)
Apatite	5	2200 ± 110	1950-2570
Whitlockite	9	49 ± 2	35-58
"Glass"	7	155 ± 15	100-210
"Pyroxene"	2	165 ± 30	135-195
Whole rock	$1.6 mm^2$	11	-

*uncertainties are the standard deviation of the mean

Figure 1.