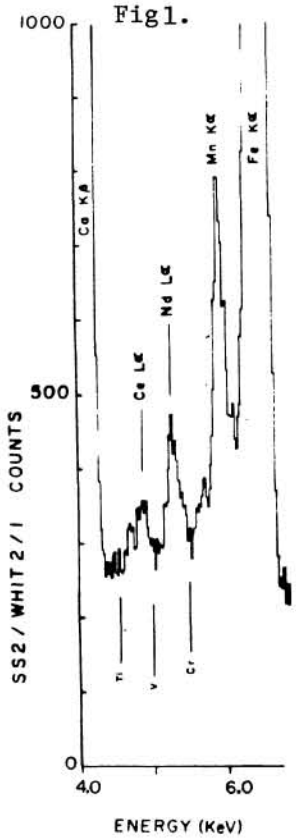
REE FRACTIONATION IN ST. SEVERIN PHOSPHATES: IMPLICATIONS FOR Pu-REE COHERENCE D.S.Woolum,Calif.State Univ.Fullerton,CA.92634; D.S.Burnett,Caltech, Pasadena,CA.91125; C.J.Maggiore and T.M.Benjamin,LASL,Los Alamos,N.M.87545.

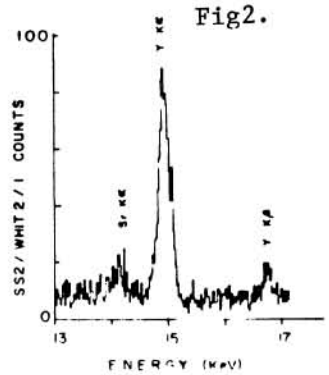
The chronological applications of Pu-244 abundances for inferring relative meteorite and lunar ages and pre-solar galactic r-process nucleosynthesis time-scales are well known. Since no other Pu isotope is available, another element must be used for normalization. The other actinides, in particular U, are no longer considered suitable(see (1) for a review). Based on most of the available natural and synthetic sample data (2 - 8) and theoretical expectations (9),the light rare earths appear to be the best candidates. However, Pu data(10) and recent REE data(11),for whitlockites and apatites in equilibrated ordinary chondrites, together suggest a major Pu-REE fractionation in the phosphates.From (10),$Pu_{apat} \sim 0.2\ Pu_{whit}$,but $Nd_{apat} \sim 1.3\ Nd_{whit}$ from (11).

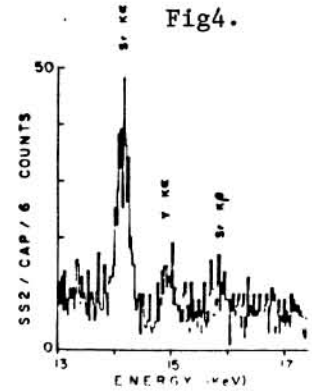
Using the LASL proton-microprobe, we have obtained (PIXE) X-ray spectra for 2 whitlockite and 2 chlorapatite grains in the St.Severin LL6 chondrite.Typical runs used a 10-25 μ beam spot and a 40 μ Al absorber to attenuate the major element X-rays, maintaining deadtimes at ~ 3%. Runs were normally of 1 hr. duration,with total integrated charges of ~ 20 μ C. All 6 whitlockite spectra are in good agreement, showing prominent Ca,,Fe,Ca escape, and (weaker)Mg and Mn peaks. P is critically-absorbed by the Al absorber and lies under the Ca K α escape peak. Typical St.Severin whitlockite has about 2% Mg & 0.4% Fe(12) and 0.02% Mn(11). Fig.1 shows the 4-6 KeV energy region from a several hundred μ whitlockite grain. Here, the Ce and Nd L α peaks are clearly evident;the weaker peaks below Ce and above Nd are probably,in part, due to La and Sm L α contributions, but their identifications are ambiguous due to interferences from Fe K α escape and Nd L β^1 counts,respectively. Fig. 2 shows that Y K α , β and Sr K α are also detected in the whitlockite. We infer 90 ∓ 40, 150 ∓ 50, 300 ∓ 50, and 15 ∓ 10 ppm whitlockite abundances for Nd, Ce, Y and Sr,respectively. The relatively large errors are primarily due to uncertainties in our present standard, and they will ultimately be reduced by a large factor; our counting statistics are only ~ 4%, at most. Other workers have reported REE abundances for St. Severin bulk phosphates: 130ppm Ce and 100ppm Nd (13), and 160ppm Ce and 119ppm Nd (11). Also, Jacobsen(pers.comm.)found ~ 140ppm Nd and 43ppm Sr. St. Severin phosphate is 90-99% whitlockite(10,11), so a comparison of these data with ours may be reasonable, and the agreement is relatively good.

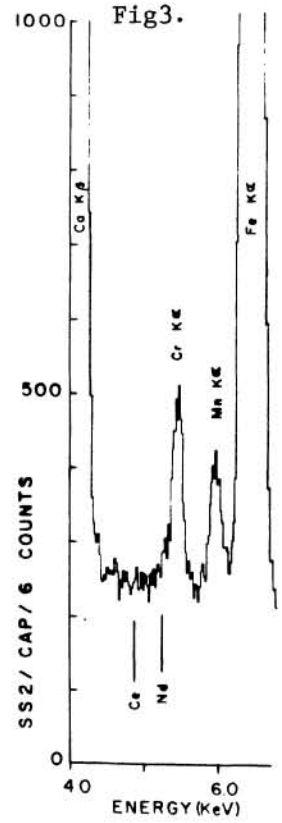
Our spectra from St. Severin chlorapatites are in distinct contrast to those from the whitlockite. As expected, Mg is virtually undetected and the Fe and Mn peaks were also less pronounced, due to these elements' lower abundances in apatite relative to whitlockite. Furthermore, Fig. 3 demonstrates that REE contributions were undetected in chlorapatites; this apatite run was nominally identical to that shown for whitlockite in Fig. 1. Only Cr and Mn K α peaks are evident in the light REE energy region. In runs using a 1 mil Kapton absorber at low beam currents,the absence of any Si or a large Fe peak indicated that there was little, if any, contributions from a phase adjacent to the apatite (primarily olivines and chromites). However, the Cr peak may be due to some weakly-induced fluorescence in an adjacent chromite; the 6 apatite runs do show about a factor of two variation in the Cr peak intensities. Based on a comparison of the Mn and Cr counts,and using the apatite Mn content from (11), either the chromite contribution is very small, or the apatite itself contains variable amounts of Cr, up to about 150ppm. The clearly reduced REE abundances in the apatite, relative to whitlockite, is mimicked in Y, as is shown by comparing Figs. 4 and 2; the opposite is true for Sr. From the whitlockite data, we conservatively estimate that 1/3 the whitlockite Nd should have been detectable in the chlorapatite, even given the presence of Cr. The ratio of Y K α counts, whitlockite/apatite, is about eight and a half.

Thus, our data do not confirm the results of (11), who reported roughly equal light-REE enrichments in whitlockite and apatite (Whit/Apat = 4/5)in Bruderheim(L6),St.Severin(LL6) and Richardton(H5). This work (11) was done on bulk phosphate separates, with relative chlorapatite/whitlockite abundances based primarily on Cl content. Their inferred apatite abundance was low (~ 3%) for St.Severin,compared to other chondrites. The discrepancy between their work and ours may reflect a problem in using Cl content to infer apatite abundance. An apparent lack of Cl material balance for St.Severin was noted by(12). Reported relative Pu abundances(whitlockite/apatite) range from 2 - 7 for ordinary chondrites(10,14). For the corresponding Nd ratio, only a lower limit of 3 is possible from our data on a model-free basis. If we assume that Y is a reliable REE analog and that no light-heavy REE fractionation was involved, our data would indicate a ratio of 8 - 9. This may provide an upper limit; with their most precise data (Bruderheim), (11) finds somewhat higher relative heavy-REE abundances whitlockite/apatite. Thus we find no evidence in St. Severin for a signficant Pu-Nd fractionation in the phosphates, but additional studies of other ordinary chondrites are necessary. Pu-Nd chronology may indeed be viable for these meteorites using bulk phosphates.









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