THE BUTTERFLIES OF PRINCIPAL COMPONENTS: A CASE OF ULTRAFINE-GRAINED POLYPHASE UNITS

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Dusts in the accretion regions of chondritic interplanetary dust particles [IDPs] consisted of three principal components, viz. (1) carbonaceous units [CUs], carbon-bearing chondritic units [GUS] and carbon-free silicate units [PUs] [1]. Among others, differences among chondritic IDP morphologies and variable bulk C/Si ratios reflect variable mixtures of principal components. The spherical shapes of the initially amorphous principal components remain visible in many chondritic porous IDPs but fusion was documented for CUs [2], GUS [3] and PUs [4]. The PUs occur as coarse- and ultrafine-grained units [1,5] that include so called GEMS [6].

MODEL. Spherical principal components preserved in an IDP as recognisable textural units have unique properties with important implications for their petrological evolution from pre-accretion processing to protoplanet alteration and dynamic pyrometamorphism. Throughout their lifetime the units behaved as closed-systems without chemical exchange with other units. This behaviour is reflected in their mineralogies while the bulk compositions of principal components define the environments wherein they were formed.

OBSERVATIONS. In IDPs L2011A9 and -K7 the PUs have a mean diameter of ~120 nm. They contain randomly distributed ultrafine silicates and Fe,Ni-metal and/or oxides in an amorphous matrix. This report is based on several hundreds of energy dispersive [EDS] spot analyses of individual grains in the PUs of several chondritic porous IDPs obtained with a probe of 10-20 nm in diameter, and on hundreds of selected area electron diffraction [SEAD] patterns obtained by scanning across a PU using a selected area aperature smaller than the unit. The analyses were performed on sections that are 80-100 nm thick. With individual grains ranging from 1-50 nm in diameter [1] each EDS spot analysis is de facto a bulk analysis of ultrafine grains and amorphous matrix. The polycrystalline SAED patterns indicate olivines and pyroxenes with variable Mg/(Mg+Fe) ratio plus an iron phase that may contain nickel. The absence of sulfur in PUs rules out Fe,Ni-sulfides. The SAED data are consistent with, but not uniquely diagnostic for Fe-oxides, and Fe,Ni metal may be present. Rare PUs have a thin (< 60 nm) rim that is free of crystalline phases. This rim has the highest Mg/(Mg+Fe) ratio within the unit. Iron oxide nanograins may decorate the external surface of zoned GUs. Even rarer PUs are characterized by concentric turbostratically-stacked 1.2 nm lattice fringes. This fringe spacing and the chemical composition indicate smectite.

The spot analyses of PUs may show either increasing or decreasing Mg/(Mg+Fe) ratios from the center to the rim in individual units. Most PUs are Al-free ferromagnesiosilica materials but individual spots in some units have 1-3 wt% Al₂O₃. The spot analyses of an individual PU show considerable scatter in a Mg-Fe-Si ternary diagram. The observed scatter is not random considering the fact that the SAED data uniquely support Mg- and/or Fe-bearing minerals which is graphically represented lines originating from the Fe and Mg corners of the diagram (Fig. 1a). These lines connect the outliers of spot analyses within a PU which is shown for the most Mg- and Fe-rich PUs in IDP L2011K7 (Fig. 1a). The bulk composition of a unit as calculated from the spot analyses closely matches the center of the butterflies. A line through the wing tips intersects the diagram close to the Si-corner. These examples are representative for ALL recognisable GUs in chondritic porous IDPs.

DISCUSSION. The butterflies that enclose EDS spot analyses within recognisable PUs reveal two remarkable properties of these units. Firstly, closed-system crystallisation of these amorphous units occurred at constant bulk Mg/(Mg+Fe) ratio but variable (Mg+Fe)/Si ratios. Growth of Mg-rich olivine was counterbalanced by Fe-richer pyroxene while precipitation of Fe,Ni-metal, or oxides, promoted the formation of Mg-enriched silicates which is entirely consistent with the SAED data. The question is the timing of crystallisation of these PUs. Based on nucleation and growth kinetics alone [7], the nanocrystals could form during atmospheric entry flash-heating, or during perihelion of these cometary particles. As diffusion rates are not
Principal Component Butterflies: Rietmeijer, F.J.M.

zero at 200–225ºK the nanocrystals conceivably formed during the 4,500 Myrs residence of PUs in protocomet nuclei. The point is that the very presence of olivine, pyroxene, and Fe,Ni metal and/or oxide, nanocrystals in PUs may not be as relevant as their closed-system behaviour to understand these objects.

Secondly, the centers of the butterflies, that can be graphically constrained, represent the original PU bulk compositions which, based on their closed-system behaviour, are those of the original materials that accreted into protoplanets. This result for PUs in IDP L2011K7 shows that (1) the original bulk compositions are closer to serpentine than to olivine with several units of pyroxene composition, and (2) PUs display a wide range of bulk Mg/(Mg+Fe) ratios (Fig. 1b). This range is comparable to these ratios in Mg-rich carbonaceous chondrite matrices [8,9] but PUs include more Mg-rich compositions than these matrices.

The principal components in chondritic IDPs contain the clues to the formation of these units, e.g. vapor phase condensation. Thus, it is intriguing that the most Mg-rich PU bulk composition is similar to the mean Mg/Si ratio in an experimentally produced modal forsterite smoke [10]. Hydration of this smoke produced saponite and kerolite instead of serpentine [10]. Likewise, smectite in very rare PUs also reflects the silica saturated nature of PUs.

Figure 1: Ternary diagrams Mg-Fe-Si (at. wt%) showing [a] the butterflies for the most Mg- and Fe-rich polyphase units with bulk compositions matching (Mg,Fe)3Si2O7 in IDP L2011K7 and [b] bulk compositions of 26 PUs in this chondritic porous IDP compared to carbonaceous chondrite matrix compositions from refs. [8] (solid triangle) and [9] (dashed line).

CONCLUSIONS. Spot analyses within PUs that are recognisable as individual units in IDPs define closed-system crystallisation of amorphous PUs at constant bulk Mg/(Mg+Fe) ratio but variable (Mg+Fe)/Si ratios. Their bulk compositions are comparable, but include more Mg-rich units, to carbonaceous chondrite matrices. This report shows the first direct link between chondritic IDPs and these matrices. It also shows that the anhydrous mineralogies in the most primitive solar system materials formed isochemically and were kinetically controlled.