Occurrence of Fe in sulfides, silicates and metals in enstatite and ordinary chondrites: Wide ranges in oxidation-reduction due to variations in H$_2$O gas?

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Introduction: Iron is one of the most abundant elements in solid materials of the solar system, and occurs in chondrites in 3 main mineral groups: silicates, metals, and sulfides [1]. Speciation of Fe between these mineral groups is an indicator of the conditions where chondrites formed in the solar nebula. Fe-speciation of chondrites between silicates and metal indicates wide variations in oxidation state [1]. Were there similar variations in the extent of sulfidation? In this project, we use Fe speciation among silicates, metal, and sulfides in enstatite, ordinary, and Rumuruti-like chondrites to address variations in oxidation, reduction and sulfidation in the part of the solar nebula where these chondrite groups formed.

Methods: We used elemental and BSE maps of polished thin sections (pts) to determine modes of minerals (enstatite, olivine, Fe,Ni-metal, troilite,⋯) in: Bensour (LL6), Mt. Tazerzait (L5), Tamdakht (H5), LEW 88180 (EH5), NWA 974 (E6), NWA 953 (R3). Elemental maps were collected using a JEOL JXA-8900 electron probe micro-analyzer (EPMA) at Waseda University (WU). Modes were determined manually using grids overlain on the pts maps in a graphics program. The compositions of major minerals were analyzed using the WU EPMA. We calculated moles of Fe in each mineral using the mineral compositions, published molar volumes and the modes.

We also used wet chemical analyses of chondrite whole rocks from two data sets: one collected and compiled by the Smithsonian Museum, US [2], and the other by the National Institute of Polar Research (NIPR), Japan [3]. In these analyses, FeO, Fe-metal and FeS were determined directly.

A reaction space approach [4,5] was used to identify the main reactions possible between minerals and O- and S-rich gas in E, O and R chondrites. The reacting system consists of the following phases and solid solution vectors: NaAlSi$_3$O$_8$, CaMgSi$_2$O$_6$, MgSiO$_3$, AlMg$_{1-x}$Si$_x$, SiO$_2$, O-rich gas, S-rich gas, FeMg$_{1-x}$Fe-metal, FeS, CaAlNa$_{1-x}$Si$_x$ and Mg$_2$SiO$_4$. These phases can be described by the components: Na, Ca, Mg, Al, Si, O$_2$, S$_2$ and Fe. In this system, all transfers of mass between the silicate, sulfide and metal subsystems can be described as progress along two reactions: (R$_m$) Mg$_2$SiO$_4$ + 2 FeMg$_{1-x}$ = 2 Fe + SiO$_2$ + O$_2$ and (R$_s$) Mg$_2$SiO$_4$ + 2 FeMg$_{1-x}$ + S$_2$ = 2 FeS + SiO$_2$ + O$_2$. Increasing reduction is indicated by progress on R$_m$, increasing sulfidation by progress on R$_s$, and oxidizing conditions are indicated by minor progress on both reactions. Progress on R$_m$ is designated by $X_m$ and ranges from 0 to 1; likewise, $X_s$ shows progress on R$_s$.

Results: Fe-speciation determined from all three data sets indicate wide variations in FeO/Fe-metal ($X_m$ from 0 to 1) and limited variations in FeO/FeS ($X_s$ mostly from 0.1 to 0.3). The results from most oxidized to most reduced are: R-, LL-, L-, H-, E-chondrites, in agreement with [1].

Considering a model for flow of ice and other materials in the solar nebula [6], the extent of oxidation was high at an evaporation front. In this model, evaporation of H$_2$O-ice caused the local gas to become enriched in H$_2$O, increasing the oxygen fugacity of the gas. With more oxygen present in the gas, R$_m$ could proceed to the left, transferring Fe from metal to silicates. The R chondrites could have formed just inside the evaporation front, where the gas was enriched in H$_2$O-vapor. Ordinary and enstatite chondrites might have formed farther inward from the evaporation front.


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