

AN EXPERIMENTAL STUDY OF IRON SULFIDE FORMATION KINETICS IN H₂-H₂S GAS MIXTURES AND APPLICATION TO IRON SULFIDE CONDENSATION IN THE SOLAR NEBULA, D. Lauretta¹ and B. Fegley, Jr.^{1,2} (1) Dept. of Earth & Planetary Sciences, and (2) McDonnell Center for the Space Sciences, Washington University, St. Louis, MO 63130-4899 USA.

Introduction. Troilite (FeS) is the most common sulfur-bearing mineral in meteorites. Furthermore, FeS, which is the dominant sulfur-bearing condensate in solar gas, was presumably an important sulfur-bearing phase accreted by the terrestrial planets. It is generally believed that troilite is the only iron sulfide to form in a solar composition gas [1,2]. Recently it was suggested that pyrrhotite (Fe_{0.875}S) would condense before troilite [3]. Thermochemical equilibrium calculations were done for stoichiometric FeS and pyrrhotites ranging from Fe_{0.98}S to Fe_{0.875}S (see Figure 1). These show that FeS forms at the pressure independent temperature of 719 K in a solar composition gas via the net thermochemical reaction H₂S(g) + Fe(metal) = FeS(solid) + H₂(g). The calculations show that FeS forms 56° before Fe_{0.875}S. This is in agreement with [1] and [2] but contrary to [3]. Using thermodynamic data from [4] and [5] the uncertainty in the condensation temperatures is ±20K. Fegley [6-8] presented theoretical models of troilite formation kinetics which predict that FeS formation is rapid relative to the estimated nebular lifetime of 10¹³ seconds. However, prior to this study no experimental data were available on the kinetics of troilite formation in the solar nebula. Here we present experimental results which show that the reaction rate is indeed very rapid.

Experimental Methods. The above reaction was studied by suspending iron foil (Johnson Mathey Puratronic grade, 99.9975%), of known weight and surface area in a Deltech vertical tube furnace within an H₂S /H₂ gas mixture (868 ppm H₂S ±2%) for varying time periods and at temperatures of 500-650 °C with an uncertainty in the temperature of ±5K. These conditions fall within the troilite stability field (see Fig. 1). Standard techniques were used to control gas flow rates and temperature. The reaction rate was determined by measuring the weight change of the reacted samples after removal from the furnace.

Results. Figure 1 shows condensation temperatures of FeS and pyrrhotites (Fe_{0.98}S, Fe_{0.90}S, Fe_{0.89}S, Fe_{0.875}S) at different H₂/H₂S ratios calculated from thermochemical data presented in [4]. The diagram predicts that either troilite or Fe_{0.98}S is the first iron sulfide to condense from a solar composition gas. However, X-ray diffraction of several experimental samples shows only troilite and no detectable pyrrhotite. Once troilite forms in a solar gas, pyrrhotites are unstable and do not condense. The solid circles in Figure 1 represent the location of the runs that reacted to produce troilite. The open circle represents run 1, which did not react over 43 hours, indicating that it lies in the stability field for iron metal. The condensation temperatures for the iron sulfides at the solar H₂/H₂S ratio (H₂S/H₂ ~ 3.69 × 10⁻⁵) are listed in Table 1. The reaction rates for each run are presented in Table 2. The samples that were exposed to the gas for relatively longer periods of time show the lowest reaction rates, even though these samples were kept at higher temperatures. This is possibly due to diffusion control and to the rate dependence on T and f_{S₂} which varies as exp[2 log₁₀(f_{H₂S}/f_{H₂})-1.740-3388.6/T].

Applications to the Solar Nebula. The sulfurization of Fe-alloy grains is widely acknowledged to have been responsible for the incorporation of sulfur into solid material in the solar nebula. This assumes that the rate of FeS formation is rapid relative to the lifetime of the solar nebula. The kinetics of this reaction are important for understanding FeS accretion by the terrestrial planets, the origin of troilite in meteorites, and gas-grain chemistry in protostellar accretion disks. However, the only available kinetic data on iron sulfurization reactions to date were obtained by materials scientists working at pressure, temperature, and compositional conditions which do not apply to the solar nebula. This study is the beginning of a larger research project designed to increase the amount of information available on gas-solid reaction kinetics under solar nebula conditions. This study shows that the retention of sulfur by troilite formation is a relatively rapid process at the H₂/H₂S ratio under consideration. Future experiments are being planned to measure these rates under conditions much closer to those estimated to occur in the solar nebula (log₁₀(P_{H₂}/P_{H₂S}) ~ 4.4).

Summary. Thermodynamic calculations show that troilite is the most likely candidate for the iron sulfide phase responsible for the retention of sulfur in the solar nebula. This requires that the reaction rate be fast enough for the reaction to occur within the lifetime of the solar nebula. The thermodynamic and kinetic results presented in this abstract strongly suggest that troilite is the only stable iron sulfide in the solar nebula and that troilite formation occurred within the lifetime of the solar nebula.

Acknowledgments. This work was supported by grants from the NASA Origins of Solar Systems and Planetary Atmospheres Program to Washington University (B. Fegley, Jr., P.I.). We acknowledge advice from K. Lodders and technical assistance from D. Kremser and R. Poli.

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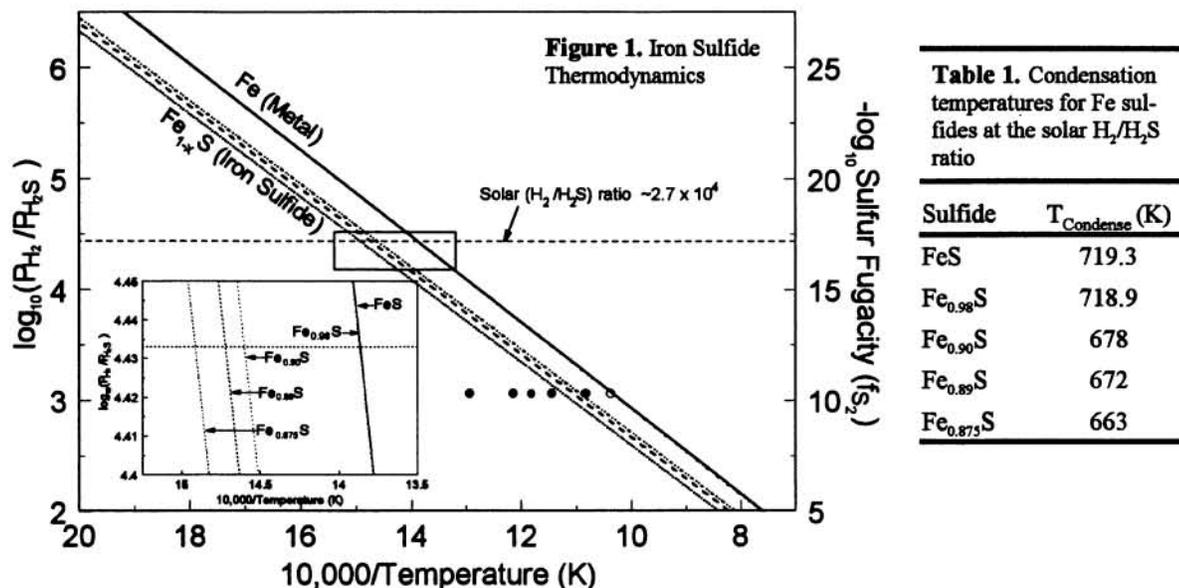


Figure 2. Photograph of a cross-section of reacted sample R2. Note the iron sulfide layer surrounding the metallic iron core. The scale bar equals 0.17mm. The iron foil had an initial thickness of 0.25 mm.

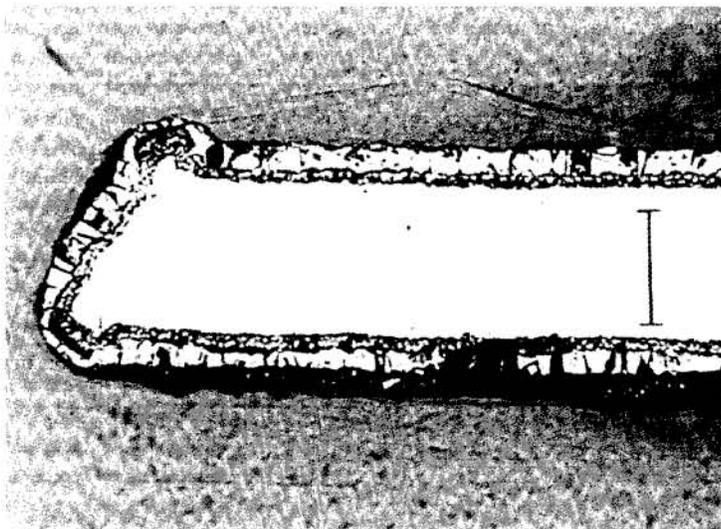


Table 2. Rate data

Run Number	Temperature (K)	Duration (min)	Rate x 10 ¹⁶ (atoms S /cm ² min)
1	963	2,575	0
2	923	2,810	5.02
3	823	2,963	4.69
4	873	872	8.53
5	776	731	6.72
6	848	456	12.86