

SULFIDIZATION OF IRON IN A DYNAMIC SOLAR NEBULA AND THE IMPLICATIONS FOR PLANETARY COMPOSITIONS. F. J. Ciesla¹, ¹Department of the Geophysical Sciences, University of Chicago, 5734 South Ellis Avenue, Chicago IL 60637 (fciesla@uchicago.edu).

Introduction: The reaction of condensed iron with H₂S gas is predicted in equilibrium condensation calculations [e.g. 1], and the product of this reaction, troilite (FeS), is commonly found in primitive solar system samples [e.g. 2-4]. To ensure that troilite was indeed a nebular product, the kinetics of troilite formation have been studied in the laboratory and shown to allow complete reaction between the H₂S gas and Fe solids on timescales that are less than the lifetime of the solar nebula [2,5].

However, evaluating whether particular chemical reactions can proceed within the solar nebula must also consider protoplanetary disk evolution due to mass and angular momentum transport as this will lead to constantly evolving pressure-temperature (P-T) conditions within the disk. Further, particles themselves will be transported through the disk, throughout the time that they are able to react with the nebular gas. These two effects will lead to particles experiencing a varied P-T history, rather than sitting in uniform conditions as assumed in previous work [2,5]. Here we explore how the varied P-T histories of Fe grains affect the rates and levels to which they are sulfidized within the solar nebula and the implications that this complicated nebular chemistry has for models of planet formation.

Motivation and Previous Work: That H₂S is expected to have reacted with Fe grains in the solar nebula, and thus its concentration decrease over time, has been invoked by some to explain the relatively low abundance of S in the Jovian atmosphere [6]. The Galileo probe measurements revealed that the Jovian atmosphere was enriched in heavy elements, including the most volatile species, relative to H₂ suggesting that volatile-rich planetesimals were accreted while Jupiter accreted its gaseous envelope. Gautier et al. [6] suggested that the most volatile species, including noble gases, were incorporated into these planetesimals in the forms of clathrates with water ice. If clathrates could form, not only would noble gases be incorporated into the planetesimals, but so would H₂S. However, because S in the Jovian atmosphere is not as abundant as expected if clathrates formed in a gas of solar composition, the authors suggested that H₂S was depleted in the region where the clathrates formed as a result of loss to FeS formation.

This mechanism was shown to work, in principle, by Pasek et al. [7], who modeled the diffusion of H₂S in a dynamically evolving protoplanetary disk. In these calculations, H₂S was assumed to be lost from the gas in the region where temperatures fell into the range

500-690 K as at temperatures above this range FeS was not stable and would not form, whereas at temperatures below this range reaction kinetics were too slow to allow significant reaction between the Fe and H₂S. Any H₂S that diffused into the region of the nebula that fell in this temperature range was assumed to immediately react with Fe and the sulfur permanently locked up as FeS. As a result, the concentration H₂S decreased to 50% its solar abundance at ~5 AU on a timescale of 10⁵ years.

The assumption of instantaneous reaction between Fe and H₂S in the nebula is a simplification, as reaction timescales are estimated to be on the order of 100-10⁶ years [7]. During this time, solid iron grains can diffuse or be advected significant distances in an evolving disk. Further, the chemical timescales are estimated assuming midplane P-T conditions, but particles are expected to spend more than 50% of their lifetime at heights of $z > 0.8H$ (H being the local disk scale-height) where temperatures and pressures are significantly less, resulting in longer chemical timescales [8]. Finally, any FeS that is formed will, itself, migrate around the disk and potentially decompose if it moves into a region of the nebula at $T > \sim 690$ K. This would reintroduce H₂S into the disk, which itself could be transported through the disk again.

New Approach: Here a new model for the chemical evolution of iron grains within a dynamical protoplanetary disk using the particle-tracking methods of [8-10] is applied to explore how H₂S evolves in a dynamic disk. The evolution of the disk is calculated using the standard α -viscosity formalism, allowing for the surface density and the temperature of the disk to be determined by internal viscous stresses and dissipation. In addition, the concentration of H₂S is also calculated using the same model, accounting for advective and diffusive redistribution.

Iron particles of a given size are initially distributed across the disk at a constant H₂/particle mass ratio. As we cannot expect to track every single iron grain in the disk, each model grain is treated as a "super-grain" with its evolution representing the evolution of a number of other grains equal to: $N_{rep} = M_{Fe} / (m_{grain} n_{part})$, where M_{Fe} represents the total amount of Fe in the disk, m_{grain} is the mass of a single grain tracked in the model, and n_{part} is the total number of grains tracked in the model. The radial and vertical motions of the particles are calculated using the methods of [8-10], with motions determined by disk advection, gas drag, vertical settling, and radial+vertical diffusion.

The method of the calculations is as follows: First the protoplanetary disk structure is defined, with H₂S vapor and Fe grains distributed uniformly according to their solar abundances [11], with all S assumed to be found as H₂S and all Fe in the form of grains. The disk then is allowed to evolve for one timestep, with the H₂S distribution being updated accordingly. The particle motions are then calculated, with each particle evolving for the length of the same timestep used in the disk evolution calculation.

Once the particles reached their new locations at the end of the timestep, they were allowed to react with the gas. Sulfidization ($\text{Fe} + \text{H}_2\text{S} \rightarrow \text{FeS} + \text{H}_2$) and decomposition (reverse reaction) rates were calculated using rates in [4,7], noting again that the decomposition reaction was not accounted for in [7]. The net gain or loss of H₂S due to reactions with the particles (accounting for their “super particle” status) was then determined and subtracted from the appropriate location in the H₂S vapor profile. The calculations were then repeated for enough timesteps to reach 10⁶ years of model time.

Results: Figure 1 shows the level of sulfidization (f value) for $\sim 10^4$ 1 μm -radius particles and where they reside within a model disk that began with a surface density of $\Sigma(r) = 20,000(r/1\text{AU})^{-1.5}$ g/cm² out to 10 AU and $\alpha = 10^{-3}$ after 10⁶ years of evolution. Particles were assumed lost to the sun if they migrated inside of 0.5 AU. Grains that remain unsulfidized have $f=1$, while grains which did react with sulfur have $0 \leq f < 1$.

Figure 2 shows the normalized H₂S profile in the nebula at various times throughout its evolution. Unlike [7], the H₂S distribution does not drop to zero, and instead is depleted by at most a factor of 2-5 at any location in the disk. This is due to the non-instantaneous nature of the chemical reactions, the resupply of H₂S at high temperatures, and the fact that grains are lofted to heights above the disk midplane.

Discussion: Sulfidization of Fe grains will remove H₂S from the nebular gas as expected by [6]. However, the level of depletion in the disk evolves as a function of time and location. At later times, the H₂S depletion will reach the levels required for the clathrate model of [6] in the very inner disk, where Jupiter was unlikely to form. The levels were reached further out in the disk at earlier times: at 2.5×10^5 years, H₂S was depleted down to 65% its solar value at ~ 6 AU, with an ambient temperature of ~ 105 K. These numbers are likely to be strong functions of disk properties and evolution histories. The model is now being applied to evaluate which disk histories allow for sufficient depletion of H₂S in the gas phase and how those disk conditions (e.g. mass, temperature profile) compare to those needed to form Jupiter. We will also examine the effects of localized heating events, such as those

responsible for CAI and chondrule melting. Comparisons to variations in S-abundance in various meteorite types are also planned.

References: [1] Lodders K. (2003) *Astrophys. J.*, 591, 1220-1247. [2] Lauretta D. S. et al. (1996) *Icarus*, 122, 288-315. [3] Tachibana S. and Tsuchiyama A. (1998) *Geochim. Cosmochim. Acta*, 62, 2005-2022. [4] Lauretta D. S. et al. (2001) *Geochim. Cosmochim. Acta*, 65, 1337-1353. [5] Fegley Jr. B. (2000) *Space Sci. Rev.*, 92, 177-200. [6] Gautier D. et al. (2001) *Astrophys. J.*, 550, L227-L230. [7] Pasek M. A. et al. (2005) *Icarus*, 175, 1-14. [8] Ciesla F. J. (2010) *Astrophys. J.*, 723, 514-529. [9] Ciesla F. J. (2011) *Astrophys. J.*, 740, #9. [10] Ciesla F. J. and Sandford S. (2012) *Science*, 336, 452-454. [11] Lodders K. (2010) *Princ. Persp. Cosmochem. Astrophys. Spac. Proceed.* p.379.

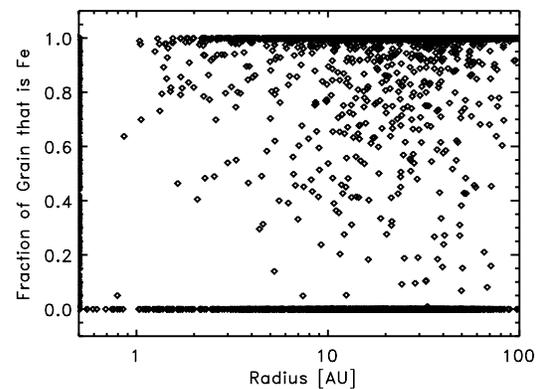


Figure 1: Plot of the level of sulfidization (f) of the 10^4 grains used in the model described here versus their final radial location after 10^6 years of evolution.

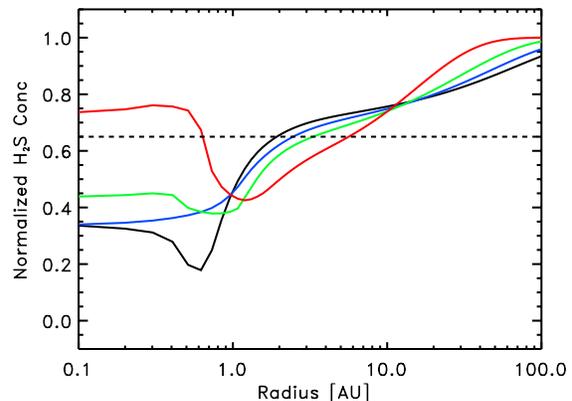


Figure 2: Normalized H₂S distribution in the model solar nebula considered here at 2.5×10^5 (red), 5×10^5 (blue), 7.5×10^5 (green) and 10^6 years (black). The dashed line at $f=0.65$ corresponds to the depletion level desired by [6] to explain the lower observed levels of S in the Jovian atmosphere.