Nucleation Constraints Lead To Molten Chondrule Precursors in the Early Solar System[†]

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This paper is on an application of nucleation theory and an empirical extension of the quasichemical solution theory of Guggenheim to an ancient bit of nature and to a long standing problem on the origins of meteorites. Chondritic meteorites, which were formed 4.6 billion years ago, contain solid chondrules, which appear to have been liquid silicate droplets formed in a rainlike process in the solar nebula with a gas of solar composition. Such droplets have been experimentally subcooled up to 500 °C below their liquidus temperatures. Just as in terrestrial clouds of subcooled water, silicate droplets should readily form metastably below their liquidus temperatures. Calculations of the condensation of these metastable droplets from a solar gas with an expected nucleation constraint on the formation of metallic iron lead to calculated compositions of chondrules at two different temperatures for each particular pressure that emulate the compositions of two types of porphyritic chondrules in the well-studied meteorite Semarkona, a minimally metamorphosed (or automorphosed), i.e., "most primordial", chondrite. The two types of chondrules, types IA and II, have very different concentrations of FeO, which are well above the equilibrium concentration because of the nucleation constraint on the formation of metallic iron. The high supersaturation of iron in the gas leads to much higher levels of iron oxide in the chondrules than would be present at equilibrium by driving the reaction $H_2O + Fe \leftrightarrow H_2 + FeO$ to the right. These concepts, if correct, can explain a major part of the chemistries of primordial meteorites and the different relative sizes of the metallic cores of planets. The physics of nucleation of metallic iron and of the crystallization of silicates governs the chemistry of iron compounds such as FeO and leads to metastable liquid chondrule droplets. These significant physicochemical processes explain the origins of two chondrule classes.

Introduction

Nucleation theory is seldom applied to many events in the universe. On earth, volcanic eruptions, metastability of crystals in lava flows, geysers, spattering water in frying, industrial explosion due to bubble nucleation, clouds, etc. are understood by nucleation theory. In this paper, we apply the seldom-utilized nucleation theory outside our world to the formation of molten condensates in the solar nebula, which are the precursors of the solid chondrules in chondritic meteorites formed 4.6 billion years ago. Chondrules appear to have been liquid silicate droplets formed, we believe, in a rainlike process in the solar nebula from a gas of solar composition. We calculate the chemistry of the liquid condensates using an empirical extension of the quasichemical solution theory of Guggenheim and well-known physical nucleation constraints on the chemistry.

This paper is dedicated to Professor Howard Reiss who introduced the first author to the concept of a constrained equilibrium in which all of the individual simple equilibria are at equilibrium except those which have a significant kinetic barrier, e.g., because of a very low probability of nucleation of the products. This concept and nucleation theories are applied in this paper to the formation of condensed phases in the early solar nebula. The results are consistent with important properties of the planets and chondritic meteorites. For example, it explains the decreasing relative sizes of the inner planetary metal (iron-rich) cores in going from Mercury to Mars. Mercury probably was formed close to the center of the solar nebula where the gas pressure was relatively high, while Mars was far from the center where the pressure was relatively low. Metallic iron tends to condense more readily at higher temperatures at the high pressure than at the lower temperatures at lower pressures. The very large surface free energies (surface tension for liquids) of metallic iron (1800 ergs/cm^2 for the liquid and higher for the solid) lead to large barriers to iron condensation which are smaller at higher temperatures than at lower temperatures.[1] With nucleation constraints, the iron atoms in the nebula become supersaturated and have concentrations in the gas that are much larger than those at true equilibrium.[1] This drives a reaction in the reducing environment of a hydrogen-rich solar gas, $Fe + H_2O \leftrightarrow H_2 + FeO$, which introduces more FeO into the silicates than would be present at full equilibrium. This property leads to the formation of much more metallic iron near the nebular center (where nucleation constraints on metallic iron formation are very low) than further out (where nucleation constraints on metallic iron formation are very high) with a continuum of the ratios of iron to iron oxide getting smaller the farther from the center. This variation in Fe/FeO ratios is reflected by the relative core sizes of the four innermost planets, and it is found in the chondritic meteorites in which these ratios are large for enstatite chondrites, small (of the order of 1) for ordinary chondrites, and close to zero for many carbonaceous chondrites. Chondritic meteorites provide important clues on the origins of condensed matter in the solar system.

The presence of nonequilibrium effects in chondrites and in the chondrules they contain[1-5] greatly enhances the information content and the number of clues locked in chondrite properties. Because of nonequilibrium effects, all chondrules differ from each other somewhat like fingerprints. This variety of compositions of individual crystals, textures of the crystals, and physicochemical and mineralogical properties leads to a wealth of information in unequilibrated chondrites. In this paper, we examine probable nonequilibrium effects on the origin of chondrules that have led to the freezing in of a large variety of chondrule properties. In ordinary unequilibrated chondrites (which have a significant amount of both Fe metal and FeO in silicates and in which chondrules have accreted to form meteoritic rocks at low enough temperatures to not equilibrate the different chondrule compositions and where there is minimal metamorphism), there is a spectrum of chondrule compositions,[6-15] which has defied a simple explanation. We will use a simple example of difference in chondrule properties to illustrate a new approach, which we hope will ultimately provide a unified explanation of the mode of formation of chondrules on the basis of the high probability that two well-understood nucleation constraints[1] are important in nebular condensation.

There are different types of chondrule classes in ordinary chondrites with different chemistries and different crystalline textures.[6-15] Such differences have been thought to require the unlikely possibility of different compositions of pockets of the likely turbulent nebular gases from which these different classes originated.[6,7] Turbulence will mix the gases on a large scale making the gas compositions uniform on a large scale. It seems to be impossible to bring in many other compositions of chondrules from parts of the nebula far away. Thus, a subsequent assemblage of these chondrules from different locations, which was postulated, we believe, is bound to be very unlikely. For any one type of chondrite, this requires a "just so" mixing to get the compositions for that type of meteorite. We will show that two of the most important classes of chondrules, the porphyritic classes (having well-formed separate crystals) IA and II that are present in all unequilibrated ordinary chondrites,[7-15] could have readily originated from the same gaseous environment and location by crystallization and freezing in of reactions of chondrules with the nebula at different temperatures to be later assembled into a parent body at yet lower temperatures. Because of nucleation constraints, different types of chondrules with different compositions can be formed in the same environment. One of these bodies, the source of the LL 3.0 chondrite Semarkona, was assembled at a temperature that was low enough (<525 K[6]) to preserve the record of the original compositions and crystal textures. (The designation 3.0 indicates the lowest degree of metamorphism (changes by external heat) or automorphism (changes by internal heat) encountered in ordinary chondrites.)

Our study utilizes a unique computer program, the F*A*C*T* system,[16] for the calculation of the complex chemical equilibria in a cooling nebular gas, which can handle 1000 chemical species in different phases including the nebular gas, liquid, and solid condensates, which includes a large number of separate compound phases and multicomponent solid and liquid solutions.[17] Especially important in our calculations is a method for the calculation of the thermodynamic properties of molten multicomponent silicates[18-22] based on an empirical modification of the quasichemical theory of Guggenheim, which permits us to accurately determine the compositions of condensates as a function of temperature. In addition, we consider two kinds of expected nucleation constraints on the process of condensa-

TABLE 1: Mean Bulk Compositions (mol %) (After McCoy et al.[6]) and Calculated Liquidus Temperatures, T_L (K), of 11 Type IA and 11 Type II Porphyritic Chondrules in Semarkona (LL 3.0)[a]

	T_L (K)	SiO_2	Al_2O_3	FeO	MgO	CaO	Na_2O	Mg/Si ratio
IA	2012	39.64	2.03	0.89	53.68	3.32	0.45	1.354
II	1810	41.19	1.45	11.38	42.61	1.86	1.51	1.034

[a] We have left out minor constituents such as TiO_2, Cr_2O_3, MnO, and K_2O, which total less than 1% of the assemblage, and we have normalized the remaining constituents to total (within a round-off error) 100 mol %.

tion,[1] (a) on the nucleation of metallic iron and (b) on the formation of solid silicates. The constraint a leads to large supersaturations of iron in the solar gas (because of the very high surface tension of liquid iron, 1800 erg/cm^2, and the even larger surface free energy of the solid supersaturation of iron; the liquid supersaturation is of the order of 250 at 1400 K), which raises the activity of iron and iron oxide at all temperatures in the reaction $Fe + H_2O \leftrightarrow H_2 + FeO$. This raises the FeO solution activity and the FeO content in the chondrule silicates considerably higher than the equilibrium values and makes the FeO content of chondrules a fairly strong function of temperature at any one pressure. The constraint b leads to the direct condensation of metastable subcooled liquid silicates (analogous to the metastable liquid water in terrestrial clouds) with the degree of subcooling needed to produce a liquid being a function of the nebular gas pressures.[1-4] Both constraints are based on well-understood physical nucleation phenomena. In the laboratory, we have preserved metastable liquid silicates with types IA and II compositions for up to 40 min while cooling above 1500 K. If correct, these processes lower the nebular temperatures of the formation of molten chondrule precursors. No metastable liquid silicate droplet has ever crystallized spontaneously in all our experiments (>100) above 1500 K indicating that the metastable droplets have a long lifetime, certainly greater than about 18 h and close to 300−500 °C of subcooling. If our experiments are funded, we plan to test the stability for much longer runs in the apparatus.

To simplify the presentation, we will not discuss in detail the many different processes proposed by others for the formation of chondrules. We will focus on our mechanism, which requires a nebular environment with far simpler properties (e.g., uniform composition) than are necessary for all other chondrule-forming mechanisms. We will present some currently popular concepts for comparison with our results.

We have chosen to focus largely on type IA and II chondrules because they are abundant[12] and they are similar in texture (both are porphyritic but do have differences in composition). They have very different compositions (Tables 1 and 2), which at full equilibrium cannot be formed from the same gaseous environment. The calculations for these two classes provide a simple and clear picture of the potential significance of our mechanism. An important result is that the compositions of the chondrules in a nebula at a given pressure are governed by the temperature. With a nucleation constraint on iron condensation, the temperature dependence of the FeO content of chondrules is large until a major fraction of Fe has condensed as FeO. Thus, as is described below, the FeO content correlates with the content of the relatively volatile silica as measured by the inverse of the MgO/SiO_2 ratio. MgO is much more refractory than FeO or SiO_2, and a decrease in this ratio means an increase in the total amount of SiO_2 in the chondrule.

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TABLE 2: Calculated Bulk Compositions (mol %) and Temperatures (K) of Metastable Subcooled Liquids with Compositions Similar to Type IA and Type II Chondrules at Several Pressures (atm)[a]

T (K)/P	SiO_2	Al_2O_3	FeO	MgO	CaO	Mg/Si ratio
1780/5	37.14	4.15	0.85	48.90	8.96	1.317
1600/5	40.30	1.19	11.40	44.54	2.56	1.105
1662/1	38.61	2.07	0.89	53.97	4.46	1.398
1515/1	40.59	1.18	11.38	44.31	2.54	1.092
1538/0.1	40.34	1.67	0.90	53.50	3.60	1.326
1411/0.1	40.80	1.17	11.38	44.13	2.52	1.082
1436/0.01	41.73	1.52	0.89	52.58	3.28	1.260
1321/0.01	40.86	1.16	11.44	44.02	2.51	1.077
1348/0.001	42.88	1.44	0.88	51.69	3.10	1.205
1240/0.001	40.93	1.16	11.39	44.01	2.51	1.075

We have left out minor constituents such as TiO_2, Cr_2O_3, MnO, and K_2O, which total less than 1% of the assemblage, and we have normalized the remaining constituents to total (within a round-off error) 100 mol %.

Calculations of Condensation in a Solar Nebula

We consider an origin of these chondrules from a gas of solar composition[23] to show that the compositions of both types of chondrules could have been produced in the same location and the same gas. We used the F*A*C*T* computer program, which calculates equilibria in complex multicomponent systems. The major condensed phases are liquid solutions. The relevant data on geologic materials of Berman and Brown[26] and Berman et al.[27] have been reanalyzed and modified very slightly so as to be consistent with the geologic equilibria used by these authors as well as with binary and ternary phase diagrams, which include these geological materials including silicate liquids. For these calculations, we consider condensation from a solar gas[23] consisting of H, C, O, Si, S, Fe, Ca, Na, Al, and Mg at pressures ranging from 10^{-3}–5 atm.

In addition, we impose the two nucleation constraints on the calculation described earlier. The first is a large nucleation constraint on the condensation of metallic iron, which has been observed experimentally,[28] and nucleation theory indicates[1] large supersaturations, $(p_{Fe}/p_{Fe}(eq))$, the ratio of the partial pressure of supersaturated Fe atoms to the equilibrium partial pressure. The second constraint is on the formation of solid silicates, which has a larger barrier to nucleation than liquid silicates, which leads to the direct condensation of metastable subcooled liquids. From the presence of olivines with fayalite (Fe_2SiO_4) rich cores, Jones[7] deduced that type II chondrules were supercooled. In fact, the compositions of the cores, Fa_{20} (i.e., $Fe_{0.4}Mg_{1.6}SiO_4$), are consistent with expectations for deeply subcooled metastable liquids; with large subcooling, the chemical driving force for the initial crystallization is so great that the composition of the first and rapidly crystallized olivines is diffusion-controlled, and if Fe^{2+} and Mg^{2+} have similar mobilities, one would expect that their compositional ratios in the first olivine crystals formed would be about the same as those of the original melts. From the data in Table 1, we can deduce a composition of about Fa_{20} in these first formed crystals in the average of the type II compositions in Semarkona. This provides support for an origin from metastable subcooled droplets and explains the presence of some of the "relict" crystals of olivine with this composition in Semarkona chondrules. Thus, an understanding of the physical processes explains the unusual chemistries of many unusual crystals and possibly the compositional differences between different chondrules.

Calculations

Our calculations will be compared with the average bulk compositions of type IA and II chondrules given by McCoy et al.[6] In Table 1, we present these compositions, calculated by leaving out the minor constituents (which total less than 1% of the chondrule) TiO_2, Cr_2O_3, MnO, and K_2O, converting to mole percent, and normalizing the compositions to total 100%. As a reference point for our calculation of condensation from a solar nebula, we have calculated the equilibrium liquidus temperatures for these measured type IA and II average compositions as 2012 and 1810 K, respectively. Calculations were made for condensation from a solar nebula with kinetic constraints blocking the condensation of Fe atoms and solid oxides. This leads to higher than equilibrium activities of FeO and to metastable liquid silicates. Calculated compositions of metastable nebular condensates are given in Table 2 at five pressures ranging from 0.001 to 5 atm at temperatures where the FeO contents of the condensate were close to those of one or of the other of the two chondrule types. We can see that some of the compositions deviate significantly from the measurements. In particular, the results for the low FeO contents at 5 atm have too much CaO and Al_2O_3 and those at 0.001 atm can be ruled out because of the low Mg/Si ratios and related differences between the absolute values of SiO_2 and MgO mole fractions. The other values at pressures between 0.01 and 1 atm are probably within the combined uncertainties in the measurements and calculations. It is likely that somewhat higher pressures (and temperatures) are possible (e.g., 2 atm). Thus, we see that under conditions defined by nucleation theory one can obtain the compositions of both type IA and type II chondrules from a solar gas. The question of origins transforms to the question, How can these compositions be preserved?

Preservation of Droplet Compositions and Chondrule Formation

These liquids are metastably subcooled below their liquidus temperatures (about 350–576 K below for the low-FeO type IA material and about 295–489 K below for the high-FeO type II material). The first author has observed subcooling of types IA and II chondrule compositions up to 500 °C in recent laser experiments. It is well-known in pyrometallurgy that liquid silicates react far more readily with gases than solid silicates do. This should be especially true where the reactants and/or products are complex chain or network silicates. Thus, nucleation and crystallization of the metastable liquids in Tables 1 and 2 at or below one of the temperatures listed in Table 2 would lead to their preservation once they are solidified (except for alkalis like Na which appear to participate in secondary reactions with silicates in CAI (refractory) meteorite inclusions[5] or in chondrules[38]). Nucleation of solids is known to be initiated by shock waves, bursts of light, and other strong radiation.

As has been shown in experimental studies,[29-38] many preformed nuclei are necessary to form the porphyritic textures. It is unfortunate that most of the experiments that have been performed were specifically designed to define pathways for the origin of chondrules by a specific mechanism of reheating solid preformed silicates to partly melt them and then cooling them rapidly. Aside from the difficulty of determining the heating mechanism that would do this, there is an overwhelming difficulty based on the observations of Hewins[29] and Hewins and Radomsky[30] that porphyritic chondrule textures appear to be formed only when the initial temperatures are somewhat below the liquidus temperature for that composition. When one considers that most chondrules are porphyritic[12] with all the

chondrules having a very broad range of compositions and hence of liquidus temperatures, this constraint would appear to be impossible to fulfill. For example, we calculate a difference of 202 °C between the liquid temperatures for the average compositions of types IA and II chondrules. Because there is a distribution of compositions within each of these types, the range of liquidus temperatures of individual chondrules is likely to be much higher than 202 °C. To precisely heat rapidly the large majority of droplets to just below the liquidus of each droplet seems to us to be highly improbable, especially considering the somewhat different sizes of the droplets even for a single location, let alone in the many different locations in which, it has been postulated, the different ordinary chondrites formed. As we discuss below, the crystallization of metastable silicate droplets well above the glass transition temperature leads to an enthalpy release from the crystals formed that heats the droplets to somewhat below the liquidus temperature. This fulfills the constraint of Hewins and Radomsky for the formation of porphyritic chondrules and suggests that recalescence of metastable droplets is probably of significance in the formation of porphyritic chondrules. Recalescence is a process of crystallizing a metastable liquid with a release of enthalpy in three stages: (1) temperature rises often to just below the liquidus temperature if crystallization is rapid; (2) crystallization slows and the temperature returns to somewhat above the ambient temperature; (3) crystallization is slow. These three stages are likely to form different kinds of crystals of a particular mineral with the first stage probably forming at least some of the relict crystals. The number of crystals formed is governed by the nucleation rates of new crystals relative to the rates of growth of already nucleated crystals. These relative rates could govern the differences between forming porphyritic chondrules (probably high nucleation rates) and other types such as, for example, barred olivine chondrules (probably high growth rate). Such differences are possibly governed by the temperatures of initial nucleation. (Many people have observed recalescence when they see metal ground on a rotating grindstone. The "sparks" seen are due to the nucleation of solid metal from the molten droplets flying off the grindstone. The "sparkle" is due to the release of the enthalpy of fusion, which rapidly raises the temperature of the crystallizing droplet to a bright glow, and the rapid cooling by radiation suppresses the glow.) Other influences on crystal nucleation and growth are related to the turbulence likely to occur in a cooling nebula. Locations in turbulent cells are likely to have cyclical pressure changes in their motions, which lead to cyclical temperatures in any given environment. Cyclical temperatures are known to catalyze nucleation rates and enhance the ripening rates of crystals. These concepts need to be examined in the future.

Conclusions

The application of nucleation theory and of the chemical concept of constrained equilibria simplifies the possible path for the formation of chondrules and primordial stony meteorites and solves long-term problems which have not been well-understood. The predicted nucleation constraints on the formation of metallic iron and on the formation of solid silicates lead to the formation of metastable molten chondrules with more than the equilibrium content of FeO. Because nucleation is a stochastic process, not all metastable molten chondrules will crystallize simultaneously. Long time scales or energy perturbations (e.g., shock waves, etc.) could lead to crystallization of the metastable liquids. The first to crystallize could be the type IA chondrules, which constitute a significant fraction of the

chondrules in Semarkona. The type II chondrules would crystallize at lower temperatures (about 100−150 °C lower) and contain more FeO and SiO_2 than type IA as calculated. Thus, the formation of chondrules and the variety of compositions found are likely to be best and most simply understood by the concept of constrained equilibria and the application of nucleation theory. The physics of nucleation governs the chemistry of iron in chondrites. The use of such physical chemistry could be very important in nature. For example, bubble nucleation theory should be very important in understanding volcanism and the origin of metastable hydrated silicates we observed in a thin volcanic flow. Constraints on bubble nucleation in the molten lava at 900−1100 °C kept the explosive mineral (with a water fugacity of at least 1 kbar) from exploding or just decomposing.

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