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INTERNAL CHEMICAL ENERGY: ORIGIN OF CHONDRULES.

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Understanding of the origin of the meteoritic chondrules has been plagued by the absence of a plausible energy source for producing molten droplets, whose subsequent crystallization into olivine and pyroxene can resemble these ubiquitous mm-sized objects. I here advocate a most plausible and common source ... internal chemical energy released when chemically unequilibrated accumulates begin rapid chemical reactions. I will argue that ample energy is available. This idea appears to have been overlooked in the literature, probably because of the chemical preoccupation with the idea of a solar thermal condensation sequence. If the solar system were to have begun as a hot gas, the condensation sequence would have progressed through a series of equilibrated chemical forms lacking in internal energy. But if it begins instead as a collection of interstellar dust with associated accreted gas molecules that begins and remains cold (10-50 K), that dust inherits the chemical energy of its unequilibrated components. The accumulation of dust is likely to lead to the sedimentation of cm-sized objects , which Cameron (1) proposed to be the origin of the parents of the chondrules; however, he suggested collisions of such objects at 1-2 km s^{-1} as the heat source for their melting. I suggest instead that the subsequent mild heating suddenly precipitates a runaway of chemical reactions within the chondrules leaving them quite hot and crystalline and depleted in the more volatile elements, whose evaporation from their surfaces provides quick cooling. The astrophysical challenge would then be the description of the precondensed cold matter, so that its energy content could be assessed, whereas the chemical challenge would be to determine whether a more satisfactory description of chondrule details results from this approach.

Though the details are far from certain, the dynamic plausibility can be inferred from published works. Cameron (1) calculated that cold dust would accumulate to cm-sized loosely structured balls before the latter could sediment. Because the central disk temperature is near 400 K near the asteroids (2), the sedimentation is accompanied by gradual heating. The first energy source is likely to be the recombination of free radicals in the grain mantles, which Greenberg (3) argued would lead to interstellar-grain explosions injecting large hydrocarbon molecules into the interstellar medium. These and other low-T reactions can heat the cm-sized accumulates to the point that considerable larger energy can be released by the formation and crystallization of olivine, pyroxene, and metal. The scenario explains the size of the chondrules. The finer dust to settle later (matrix) will have been gradually heated in dispersed form rather than as accumulates. I suggest that the chondrule formation occurs in a sudden thermal runaway, so that the redox conditions are determined by the bulk composition of the accumulate, rather than by the ambient solar gas. The FeO contant of the silicates will therefore differ from chondrule to chondrule, as observed, but zoning will not be present because the quick cooling of the small object does not allow the separation of MgO from FeO.

To evaluate this idea one must take a completely different view of the chemical state of the early solar system than the one prevalent in meteoritic literature. Consider by way of example pure MgO, and imagine that MgO molecules in the interstellar medium slowly accrete into a dust particle consisting of randomly oriented MgO molecules frozen at 10 K. This state is already liquid, at least in the sense that is possesses no long-range order. When it is heated enough for the MgO molecules to orient themselves, a spontaneous liquid → solid phase transition occurs, accompanied by the liberation of 18.5 kcal/mole of heat. If C_p=0.2 cal/gm^0 for MgO, this heat would (if adiabatic) leave the MgO crystal with its temperature elevated by $\Delta T=2300$ C. The crystallization would proceed as a wave, leaving a hot crystal behind as it propagates into frozen liquid. The final product will look like it was once molten, even though it never was so in the usual sense of the word.

A considerably larger amount of heat is released if chemical energy, rather than merely crystallization is involved. A model of interstellar dust consisting of a frozen assembly of equal numbers of MgO, SiO and FeO molecules has the advantage of being relatively nonbiased, but plausible, and also of being similar to the interstellar-dust model of Duley *et al*.(4). If I take these molecules to be bound by 0.5 eV each in an amorphous paste, then $(MgO + SiO+FeO)_{am}=(MgO + SiO + FeO)_{gas}$ - 34 kcal/mole. I further calculate (using thermodynamic data from 49th ed. of CRC handbook) that

$$(MgO + SiO + FeO)_{am} \rightarrow MgSiO_3 + Fe + 340 \text{ kcal/mole} \qquad (1)$$

which is such a great energy that it would resemble a melt until that energy could be radiated (probably by volatile evaporation). If one instead takes the already crystalline cubic structure proposed by Duley *et al*. (4),the final products are the same but $\Delta H=40$ kcal/mole, which is still enough that the recrystallization should leave the appearance of a melt, although it was really only a hot crystallization wave.

The final composition above is commonly called "reduced", and is the basic constituent of enstatite chondrules and chondrites. In this model, it was never actually chemically reduced, but instead formed with a limited closed-system oxygen supply. I propose this to be the actual origin of enstatite meteorites and chondrules, as well as of the Earth, which also has free iron. Occurrence of FeS, CaS, etc. occurs because these sulfides are predicted (5) to already exist in the initial amorphous accumulate. They are not "injected" by subsequent reduction processes.

At accumulation temperatures of 50 K, significant amounts of O_2 may freeze out into the paste of accumulating grains. In that case, the basic thermal runaway is

$$(MgO + SiO + FeO + O_2)_{am} \rightarrow (Mg,Fe)_2\,SiO_4 + 500 \text{ kcal/mole} \qquad (2)$$

as found in highly oxidized chondrules. The intermediate conditions, characterized by a spectrum in FeO/FeO+MgO in both olivine and pyroxene, are generated by closed systems having differing initial ratios $MgO:SiO:FeO:O_2$. But they represent varying oxidations of the enstatite mixture. It is, of course, possible to regard the enstatite mixture as being a *reduced* version of eq. (2) that arises because the accumulates trapped an active reducing agent (*e.g.* C_2H_2) in individual grain surfaces. In either case, rapid exothermic crystallization (not recrystallization) accompanied by volatile flow and evaporation from the chondrule seems more likely to produce the actual textures of chondrules than does solidification of a homogeneous melt. In either case, the difference between enstatite and ordinary chondrites traces to a difference in volatile content of the grain surfaces at the time of accumulation, not to some exotic differences in an imagined thermal condensation sequence.

In the development of the significance to meteoritics of the chemical state of precondensed matter, I have previously given more emphasis to thermal condensates than to nebular condensates (6), but even these thermal condensates should be made amorphous by the severe interstellar sputtering and redeposition of the magnesium silicates (7,8). Krätschmer and Huffman (9) have shown that severely sputtered olivine reproduces the interstellar 10μ silicate absorption much better than do crystalline silicates, confirming the earlier discovery (10) for amorphous olivine. Thus, even the thermal condensate portion has energy to contribute.

Space limitations prohibit discussion of many exciting consequences and related problems, especially:(a) origin of Ca-Al-rich inclusions, (b) heating of planetesimals and parent bodies, (c) oxygen isotope systematics, and (d) so-called "alteration products". But perhaps the deepest significance of the correctness of these ideas would lie not in facilitating an understanding of chondrules but rather in dealing the final blow to the idea of a hot solar condensation sequence.

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