

AVOIDING TURBULENCE IN METASTABLE EUTECTIC CONDENSATION OF REFRACTORY FE-MG-AL-O TERNARY VAPOR SYSTEMS IN MICROGRAVITY. F. J. M. Rietmeijer¹, A. J. Brearley¹, E. Dobra¹ and J. A. Nuth², ¹Department of Earth and Planetary Sciences, 1-University of New Mexico, MSC3-2040, Albuquerque, NM 87131-0001 USA, ²Astrochemistry Laboratory, Solar System Exploration Division, NASA Goddard Space Flight Center, Code 69, Greenbelt, Maryland 20771, USA.

Introduction: The question to be answered by a series of vapor phase condensation experiments is to predict when refractory grains will condense from the vapor. The results will be especially important for the formation of refractory materials in astrophysical environments. The first vapor phase experiments during the early “1980-ties” demonstrated that laboratory-based vapor phase condensation experiments carried the promise of finding the answers to this question [1, 2]. What would we be looking for in the condensed matter (smokes) from vapor phase condensation experiments using ATEM analyses of the smoke nanograins? That is how we discovered DME (Deep Metastable Eutectic) vapor phase condensation [3] which turned out to be a defining systematic phenomenon in the vapor phase condensation experiments [4, 5]. Is there a reality test that DME’s “are real”?

As a test we used the compositions of nanometer dust particles in the coma of comet P/Halley, which well-established [6], but their origin(s) were poorly understood. Our results from magnesiosilica vapor phase condensation experiments that contained Fe-nanograins proved to be a perfect match with the Halley nanograin dust compositions [7]. This success came with a caveat, *i.e.* it required an “external” iron source other than vapor condensation.

Gas-to-solid condensation experiments in a Fe-Mg-SiO-H vapor revealed that this process yielded solids with magnesiosilica (MgO.SiO) and ferrosilica (Fe-oxide.SiO₂) compositions that coincide with metastable eutectics in the MgO-SiO₂ and (FeO/Fe₂O₃)-SiO binary phase diagrams plus simple metal oxides (MgO, SiO₂ and FeO or Fe₂O₃) [8]. But no solids with mixed Mg-Fe-O compositions formed during vapor phase condensation of this Fe-Mg-SiO-H vapor.

We conducted a series of vapor phase condensation experiments and ATEM analyses of smoke nanograins to explore this so-called Deep Metastable Eutectic (DME) phenomenon (Table 1). The condensed nanograin size distributions in each of these experiments showed an inflection point between 10 to 15 nm. The larger condensate grains up to ~100 nm (our cut-off) in all of these smokes defined another normal grain size population in the smokes [5].

The only exceptions were Ca-bearing refractory smokes (Table 1). This behavior is thought to be a thermal experimental artifact caused by convection

Table 1: Vapor compositions used in DME condensation experiments; DME grain size, and range of the population of the smallest condensate grains. The data are from nine different experiments

System	Grain size (nm)	
	DME	Range
Mg-Si-O	1	1 - 15
Fe-Si-O	4	4 - 35
Fe-Si-O	2	2 - 8
Fe-Si-O	10	10 - 30
Fe-Al-O	2	2 - 25
Al-Si-O	7	7 - 12
Al-Fe-Si-O	5	5 - 30
Ca-Si-O	20	20 - 60
Ca-Fe-Si-O	16	16 - 27

inside the condensation chamber that may have delayed growth of the first nanograins due to size-dependent re-evaporation [9] that then caused (local) supersaturation prior to “equilibrium” cooling that in these refractory vapor experiments was delayed by forced disequilibrium. The result were so-called “Prigogine dissipative structures” [Fig.1]. These dissipative structures arise as a result in changes in the rates of condensation and/or evaporation prior to reaching thermal equilibrium [10]

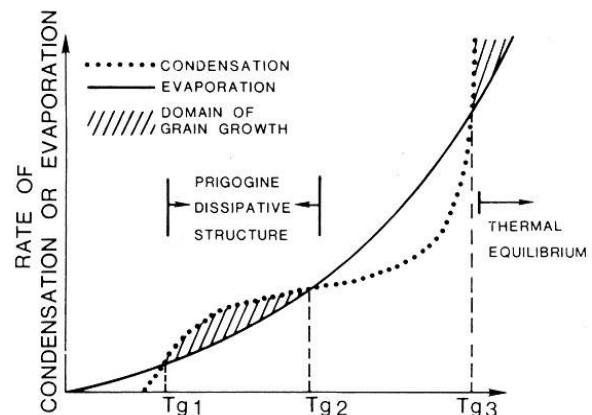


Fig. 1: Rates of condensation or evaporation as a function of grain temperature in the state of disequilibrium of temperature domains of grain growth and emergence of a Prigogine dissipative structure [10]; reproduced from [10].

Our vapor phase condensation experiments actually catch “Prigogine Dissipative Structures” that are represented by the population of the smallest condensate grains in the condensed smokes (Table 1).

It was discovered that these grains invariably had compositions that were in between the eutectic compositions in the corresponding phase diagram. This behavior is referred to as DME condensation that is constrained by the presence of stable eutectics (Fig 2).

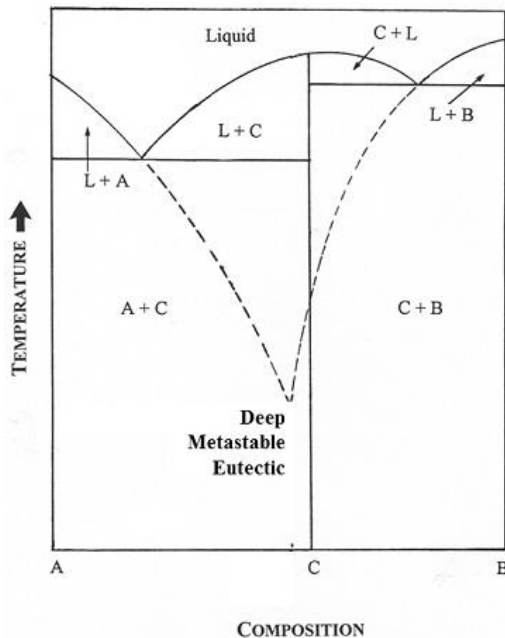


Fig. 2: Grain formation processes in oxygen-rich circumstellar outflows are another test of the metastable eutectic condensation hypothesis]. The hypothetical phase diagram A-B has two eutectics [reproduced from [11]. When equilibrium phase boundaries (solid lines) are extended towards lower temperatures (dashed lines) they will intersect at the intermediate composition “AB.” AB is the composition of a deep metastable eutectic compound. The hypothetical binary equilibrium phase diagram A-B showing the position of a (deep) metastable eutectic with composition “AB” defined at the intersect of metastable extensions of the liquidus surface (dashed lines) and located in between compositions of both eutectic points on the liquidus surface (solid lines).

This work will test the DME hypothesis by condensing solids from the Fe-Mg-Al-O ternary vapor system. Since FeO-MgO miscibility also applies to this system, the primary condensates from such a vapor should consist of pure amorphous Fe-aluminates and Mg-aluminates. No mixed Fe-Mg-spinel should be

detected as primary condensates if this hypothesis is correct, just as none were detected for the FeO-MgO-SiO system. Confirmation of this hypothesis would be a major step in establishing a simple, chemical kinetic model for the nucleation, growth and annealing of circumstellar oxide dust. Since strong convective flows in the terrestrial laboratory make it almost impossible to measure the growth and aggregation of freshly condensed refractory grains (Fig. 3).

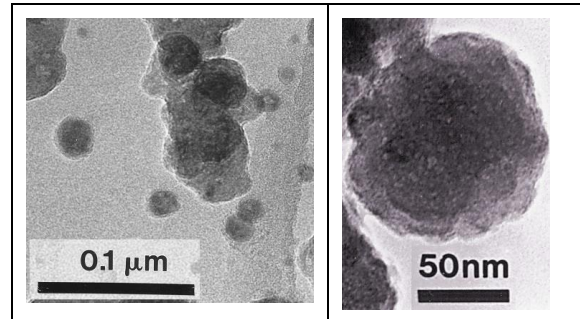


Fig. 3: TEM images of vapor phase condensed Al-Si-O smoke grains. *Left*: Al-Si-O clusters with widely varying sizes formed on the collector plate inside the condensation chamber. *Right*: a dense Al-Si-O cluster grain of DME grains that is probably an experimental artifact of turbulent condensation and agglomeration, that is avoided in microgravity.

Condensations experiments conducted in microgravity will eliminate such flows and measure the efficiency of grain growth from simple SiO, AlO and FeO vapors, the sticking coefficients for dust coagulation via analyses of the grain morphology, and size distributions of collected condensates that are returned to earth after each sounding rocket experiment.

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