CHEMICAL COMPOSITION OF COMETARY ICE AND GRAIN, AND ORIGIN OF COMETS

Tetsuo Yamamoto
Institute of Space and Astronautical Science
Komaba 4-6-1, Meguro-ku, Tokyo 153, Japan

ABSTRACT. The chemical composition of the ice and grains in a cometary nucleus is discussed by applying the condensation theory. The equilibrium condensation theory of a gas having the elemental abundances in the solar system is briefly reviewed. The composition of solids predicted by the equilibrium condensation theory is compared with that of the ice and grains in the nucleus; the latter is inferred from the observations of cometary molecules and grains. On the basis of the results of this comparison, a scenario for the formation history of comets is proposed, and discussion is given on the temperature and region of the primordial solar nebula where comets formed.

1. INTRODUCTION

Cometary matter observed in the coma is classified into molecules and grains. The molecules originate from the icy component of a cometary nucleus, i.e. the low-temperature condensate, and the grain is the refractory component, i.e. the high-temperature condensate. In this article, I discuss the chemical composition of the ice and grains in a cometary nucleus, and the origin of comets as viewed from the chemical evolution of the materials that formed comets.

2. ELEMENTAL AND CHEMICAL COMPOSITION OF ICE AND GRAIN PREDICTED BY THE EQUILIBRIUM CONDENSATION THEORY

A rough estimate of the mass ratio of the icy and refractory components is obtained by noting that the ice is composed mainly of C, N, O (and H bonded with them), whereas the grain is composed mainly of Si, metallic elements (and O and H bonded with them). The elemental abundances in the solar system (Cameron, 1981) yields the ratio χ as

$$\chi = \sum_{i\,=\,\text{metals and Si}} A_i X_i \Big/ \sum_{i\,=\,\text{CNO}} A_i X_i = 0.25,$$

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where A_i and X_i are the atomic weight and abundance of the element i. It is to be noted that this value is consistent with the dust-to-gas mass ratio (χ = 0.1 to 1) derived in several comets (Ney, 1982). The detailed evaluations of the elemental composition of comets have been given by Delsemme (1978, 1982) and by Greenberg (1982).

The theoretical basis for the study of the chemical composition is provided by the equilibrium condensation theory, which predicts the composition of solids condensed out of a gas having the elemental abundances in the solar system as a function of the temperature and pressure in thermal equilibrium. This composition is regarded as model composition of solids in cosmic environment, and serves as a working hypothesis for the study of their chemical composition. The condensation calculations were carried out extensively more than a decade ago, particularly in connection with the study of the chemical history of meteorites in the early solar system (see a review by Grossman and Larimer, 1974).

Figure 1(a) shows a condensation diagram of high-temperature condensates versus the total gas pressure. As the gas cools down, tungten (W), corundum (Al_2O_3), Ca-Ti compounds, Mg-silicate, and iron (Fe) condense between 2000 and 1000 K. Of these, the most abundant condensates are Mg-silicate and iron. According to detailed calculations, Mg-silicate condenses in the form of solid solution of Mg and Fe such as pyroxyne (Mg_x, Fe_{1-x})SiO_3 or olivine (Mg_x, Fe_{1-x})$_2SiO_4$, and iron

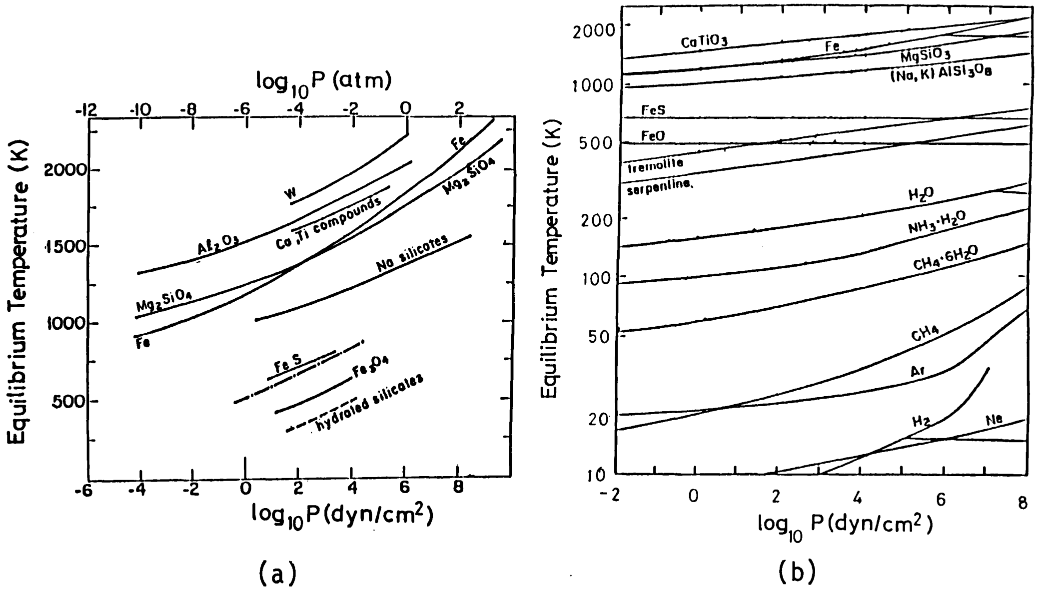

Fig. 1: Equilibrium condensation diagrams of a gas having the elemental abundances in the solar system for high-temperature condensates (a: from Salpeter, 1977) and low-temperature condensates (b: from Lewis, 1974). The equilibrium temperature is the temperature at which a bulk solid and a vapor is in thermal equilibrium, and P is the total pressure of the gas mainly of H_2.

in the form of iron-Ni alloy. At a low temperature around 500 K, Mg-silicate transforms into hydrated silicate, and iron into FeS and then into magnetite, Fe_3O_4 at a lower temperature.

Figure 1(b) shows a condensation diagram at low temperature. H_2O, NH_3 and CH_4 and their clathrate hydrates are the major low-temperature condensates, which condense at temperatures lower than about 200 K.

3. COMPARISON WITH THE OBSERVATIONS

3.1. High-Temperature Condensate

The data usually used to discuss the composition of grains have been the infrared and visible spectra. See a recent review by Ney (1982). As discussed by many authors, there are at least two kinds of grains of different composition in the coma; one is silicate, and the other is metallic or carbonaceous grains. The 10 μm peak is attributed to silicate, and the blackbody-like base line in the near to middle infrared to metallic or carbonaceous grains. The presence of silicate is consistent with the equilibrium condensation theory, which predicts that the silicate will be Mg-silicate or its low temperature form, hydrated silicates. Rose (1979) discussed the detailed composition on the basis of the laboratory measurements of infrared spectra of various silicates including meteoritic ones. He listed three kinds of silicates that fit the infrared data: (1) olivine or anorthite, (Ca, Al)-silicate, (2) amorphous olivine, or (3) mixture of amorphous olivine and anorthite. As for the blackbody-like grains, Fe-Ni alloy, FeS, or magnetite is predicted by the condensation theory. Carbonaceous grains condense in the gas with the C/O ratio larger than unity (Gilman, 1969).

There is another approach to the problem of the grain composition. Comets approaching very close to the sun, usually less 0.2 AU, exhibit many emission lines of metallic atoms in the coma. Comet Ikeya-Seki (1965 VIII) is an excellent example. In this comet, metallic elements such as Ca, V, Fe, Ni, Cr, Co, Cu, Mn, Na, and K were observed, and the upper limits of the line intensities for Al, Ti, Mg, Si and Li were determined.

Arpigny (1978, 1979) calculated the abundances of the elements of "iron sequence" (Ti, V, Ni, Cr, Co, Cu, Mn) relative to Fe in the coma of this comet. Comparing the abundances of these and other metallic elements, he found that (1) many of them were essentially of solar abundances, but that (2) the refractory elements such as Al, Ti, Ca, Si, and Cr were depleted compared with those of less refracrory elements such as Cu, when referred to Fe and the solar abundances. He interpreted that the depletion was due to the result that most of the refractory elements were locked in the grains. These results imply that the composition of cometary grains is not far from that predicted by the equilibrium condensation theory. There is, however, a problem that Si and Mg were not observed in the comet. Both are expected to form Mg-silicate near the condensation temperature of iron. In addition the 10 μm emission band observed in many comets is the strong evidence that Si is one of the elemental components of cometary grains. These atoms may sublime from the grain in molecular form as suggested by the sublimation experiments of the solid with the solar composition (Hashimoto, 1983).

A similar depletion pattern has been observed in the interstellar gas as well (e.g. Morton, 1974), suggesting that cometary grains have some relation to interstellar grains.

3.2. Low-Temperature Condensate

The composition of the icy component of the nucleus has been studied by the chemical models, which simulate molecular reactions in the coma. The models in which various abundances of the parent molecules (i.e. the icy component) are assumed have been proposed, and it has been found that the models that adopt the interstellar molecule abundances reproduce the molecular abundances observed in the coma better than the models adopting the major parents H_2O, NH_3 and CH_4 as predicted by the equlibrium condensation theory (Giguere and Huebner, 1978; Huebner and Giguere, 1980; Mitchell et al., 1981; Swift and Mitchell, 1981; Biermann et al., 1982). This result shows that the icy component of the nucleus was formed through non-equilibrium condensation. Comparing the abundances of cometary molecules produced from the ice having the interstellar molecule composition with the observed abundances, Yamamoto et al. (1983) have pointed out that the icy component of a cometary nucleus is approximately regarded as a condensate of interstellar molecules, but is depleted of very volatile species such as CO and N_2 (and of course, H_2) compared with their interstellar abundances.

As pointed out by Feldman (1983, 1985), neutral molecules exhibit similar relative abundances from comet to comet except for CO. CO has been detected only in two comets, West (1976 VI) and Bradfield (1979 X), and its production rates relative to H_2O, Q_{CO}/Q_{H2O}, are estimated to be 0.27 for Comet West and 0.011 for Comet Bradfield, whereas it is suggested that the abundances of CO_2^+ and presumably CO_2, which is less volatile than CO, are comparable in the comets (Feldman, 1985). For both of the comets, it is shown that CO is a parent molecule from the spatial distribution of the emission. The abundance similarity of the neutrals suggests that cometary nuclei are homogeneous in structure, and formed in limited thermal environment. On the other hand, the difference in the CO abundance may imply that the temperature of the formation environment is around the sublimation temperature of CO in the ice, since in this case some comets retain CO in their icy component and other comets do not.

4. ORIGIN OF COMETS: THE CHEMICAL VIEWPOINT

The evolution of the ice and grains that formed comets is divided into the two stages of (1) the interstellar molecular cloud and (2) the primordial solar nebula, which formed by gravitational contraction and fragmentation of the parent molecular cloud (Yamamoto, 1985b).

In the parent cloud, molecules in the cloud condensed onto the surface of refractory grains to form ice mantle on it; such grains will be characterized by the model of interstellar grains in a dense molecular cloud as proposed by Greenberg (1983). The origin of the refractory core would be the places where the gas would not have been far from thermal equilibrium such as stellar atmospheres and the gas ejected from stars, as suggested by the grain composition discussed in section 3.1. The outer ice mantle condensed in the cloud is composed of molecules including very volatile species except H_2 and He (Greenberg and d'Hendecourt, 1985) because of very low temperature of the grain surface. The ice mantle would have the composition similar to that of interstellar molecules. The size of these grains is on the order of 0.1 μm (Greenberg, 1983), much smaller than the size of a cometary nucleus. The grains coated with the mantle of frozen interstellar molecules will be the raw material of cometary nuclei.

At the primordial solar nebula stage, the grains would have suffered heating since the temperature in the solar nebula was expected to be higher than that in the molecular cloud. The degree of sublimation due to heating depended on the distance from the sun, varying from sublimation of the refractory core in the inner region to almost no sublimation of volatiles in the outer region. The formation region can be determined by finding out the region where the ice mantle suffered sublimation to the degree so as to achieve the ice composition discussed in section 3.2. Cometary nuclei will be the planetesimals or their agglomerates (Donn, 1981) made of such processed grains in this region. The formation mechanism of planetesimals, whose size is on the order of cometary nuclei, is considered to be gravitational fragmentation of the dust layer in the solar nebula (Safronov, 1972; Hayashi, 1972; Goldreich and Ward, 1983). It is to be noted that cometary nuclei thus formed are expected to be homogeneous both in composition and structure.

On the basis of this scenario, Yamamoto (1985a) evaluated the temperature of the formation region of cometary nuclei in the solar nebula by calculating the grain temperature in the dust layer. Table I lists the sublimation temperature T_{subl} for possible species composing the ice mantle. The species marked by the circles are the parent molecules reported to have been detected in the comets, and those marked by the triangles are the plausible parents of the detected species shown in the parentheses.

From the composition of icy component of the nucleus described in section 3.2, the temperature of the formation region is conservatively estimated to be between $T_{subl}(CO_2)(\cong 70\ K)$ and $T_{subl}(N_2)(\cong 20\ K)$. If the abundance difference of CO from comet to comet is attributed to that of CO in the ice of the nucleus, the temperature of the formation region is expected to be around $T_{subl}(CO)(\cong 25\ K)$. It is interesting to note that this temperature is close to the temperature suggested by A'Hearn and Feldman (1985) and Greenberg (1985), who discussed the source of S_2 observed in Comet IRAS-Araki-Alcock (1983d).

The formation region of cometary nuclei based on these temperature conditions was discussed by Yamamoto (1985a), who investigated the formation region by adopting the two temperature distributions in the solar nebula: (A) the radiative equilibrium distribution (Hayashi model, 1981) and (B) the adiabatic distribution (Cameron model, 1978). The result is summarized in Fig. 3, which shows the sublimation temperature

Table I. Sublimation temperature

	T_{subl} *)	adopted abundance log [X]/[H_2]	detection reported	ref.
H_2O	152	-4	o	1, 2
HCOOH	112	-7	Δ (HCO)	3
CH_3OH	99	-8		
HCN	95	-7	o	1, 4
CH_3CN	91	-8	o	5
SO_2	83	-8		
NH_3	78	-7	o	6
CS_2	78	-8	Δ (CS)	7
HC_3N	74	-8	o	8
CO_2	72	-5		
CH_3C_2H	65	-8		
H_2CO	64	-8	Δ (HCO)	3
C_2H_2	57	-7		
H_2S	57	-7	Δ (H_2S^+)	3
C_2H_4	42	-7		
CH_4	31	-4.5		
CO	25	-4	o	9
O_2	24	-5		
N_2	22	-4		
H_2	5	0		

*) gas density $n = 10^{13}$ cm^{-3}

references.

(1) Irvine and Schloerb, IHW Newsletter (January 13, 1986).
(2) Jackson et al., NASA SP-393, p. 272 (1976).
(3) Cosmovici and Ortolani, Nature 310, 122 (1984).
(4) Huebner et al., Icarus 23, 580 (1974).
(5) Ulich and Conclin, Nature 248, 121 (1974).
(6) Altenhoff et al., Astron. Astrophys. 125, L19 (1983).
(7) Jackson et al., Astron. Astrophys. 107, 385 (1982).
(8) Hasegawa et al., Icarus 60, 211 (1984).
(9) See the text.

T_{subl} for the major possible species composing the ice mantle as a function of the nebular density along with the temperature distributions of the two nebular models. From the upper limit of the temperature, $T_{subl}(CO_2)$, the formation region has to be outside the Saturnian region (> 14 AU) for (A) or outside the Uranian region (> 15 AU) for (B). The outer limit determined by the lower limit of the temperature, $T_{subl}(N_2)$, is beyond the planetary region for both cases; the distance from the sun is 110 AU (A) or 79 AU (B). If the formation region is around the the region of CO sublimation, the distance becomes 82 AU (A) or 65 AU (B), again beyond the planetary region.

If cometary nuclei actually formed beyond the planetary region in the solar nebula, an interesting possibility arises that a large number

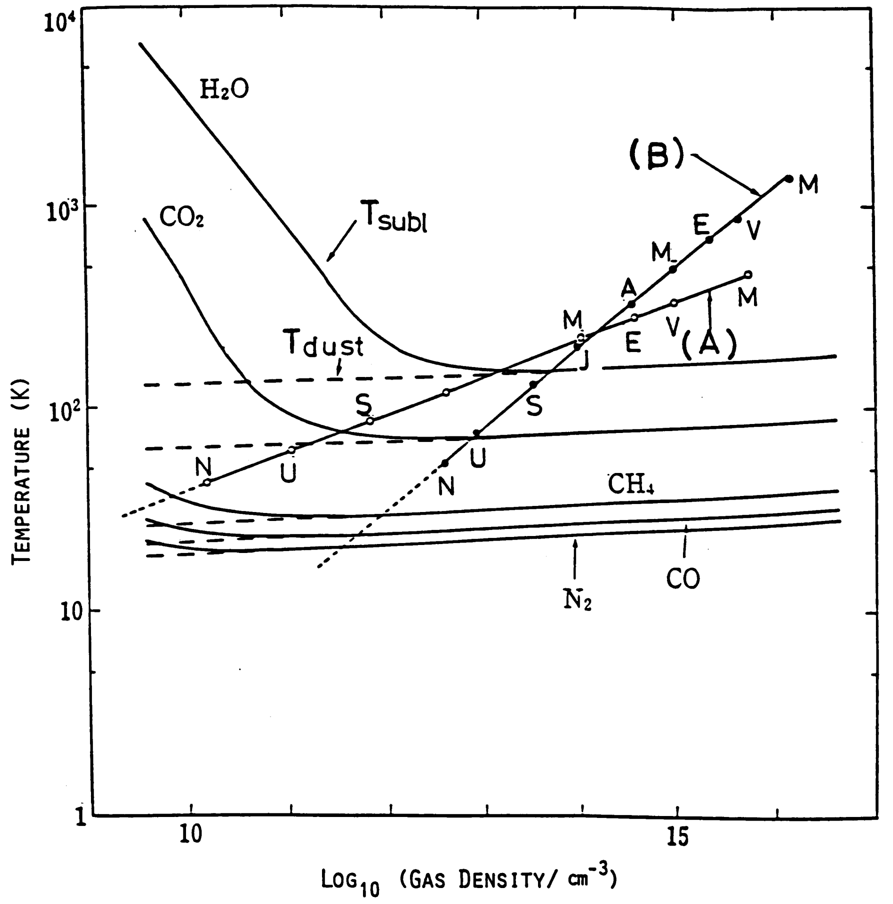

Fig. 2: The sublimation temperature T_{subl} and the corresponding grain temperature T_{dust} versus the gas density of the solar nebula. The temperature distributions for the two nebular models are shown with formation regions of the planets (M, V, E, ...): (A) the radiative equilibrium distribution and (B) the adiabatic distribution.

of comets has been reserved there up to the present time, although they will not be on their original orbits but have diffused both outwards and towards the directions perpendicular to the ecliptic plane through gravitational perturbation due to, for instance, the encounters of passing stars and giant molecular clouds during the age of the solar system. Weissman (1985a, b) has suggested, from the study of the evolution of cometary orbits, the presence of the "inner" Oort cloud extending from just beyond the Neptune's orbit to 10^4 AU or more, where it merges into the "classical" Oort cloud. In developing a theory of formation of planetary systems around stars, Nakano (1985) has pointed out that planetesimals may still remain beyond the planetary region, and has discussed the present status of these planetesimals.

These lines of the theoretical studies independently suggest the presence of a cometary cloud beyond the outer planets. It is an exciting problem to prove or disprove the presence of this hypothetical cometary cloud, a swarm of remnant planetesimals in the early solar system, for obtaining a deeper understanding not only of the origin of comets but also of the origin and evolution of the solar system.

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DISCUSSION

IRVINE: I am skeptical about the reported observations in comets of CH_3CN and HCN (prior to Halley), and also of H_2S^+ and HCO.

P.D. FELDMAN: I agree that the detection of H_2S^+ and HCO is dubious. It is unlikely that H_2S^+ would show up so close to the nucleus (in comet IRAS-Araki-Alcock). For HCO, there are good spectra from several other observers, but no one has confirmed this identification.

D'HENDECOURT: How much the observations of H_2O/CO ratio really reflect the abundance of these molecules in the original comet? CO will evaporate much further out from the Sun than H_2O.

YAMAMOTO: CO on and near the surface of the nucleus will sublime at large heliocentric distances. When a comet comes close to the Sun and can be observed, sublimation becomes active, and as a result CO in the inner nucleus comes to sublime along with H_2O. Considering that cometary nuclei will have homogeneous structure as suggested by the similarity of the relative abundances of neutral molecules observed in the coma, I think that the observed H_2O/CO ratio probably does not differ much from the original one in the nucleus.

HUEBNER: If molecules are to condense on grains in the presolar nebula then the partial pressure of each molecular species must be equal to its vapour (sublimation) pressure. This would then determine the sublimation temperature, if thermodynamic equilibrium is valid. It probably is not valid and one should use sticking coefficients.
YAMAMOTO: I assume that molecules condense on grains in the parent molecular cloud and that the volatile molecules on grain surface such as N_2 sublime in the solar nebula; condensation and sublimation

occurred in different environment. Condensation in the molecular cloud is a non-equilibrium process as the supersaturation ratios (= partial pressure/vapour pressure) of most of the species (except H_2 and He) are much larger than unity because of the very low temperature. In this case, condensation is determined by the kinetic factors such as mean free time of collisions between a molecule and a grain, and sticking coefficient. For sublimation in the solar nebula, I calculated the sublimation temperature from detailed balance between sublimation from grain surface and sticking onto it (cf Yamamoto 1985a).

OMONT: Is the total amount of nitrogen compounds depleted relative to the total amount of nitrogen in the interstellar medium?
YAMAMOTO: Yes. The total amount of nitrogen compounds relative to OH (or H_2O) is 10^{-3} to 10^{-2} in comets, probably comparable with or a little larger than the abundance of interstellar NH_3 relative to H_2O.

GREENBERG: Am I correct in assuming that when you were deriving a depletion of CO in comets resulting from sublimation you seemed to be starting with an interstellar ratio of gaseous CO to solid H_2O. This, of course, is larger than the ratio of CO to H_2O in the dust. If you had started with the latter, you would have had an initial $CO/H_2O \cong 0.10$, according to my dust model and I think that this may not be too far from the observed mean in comets or perhaps it is better to say it fits within the wide range of observed values.
YAMAMOTO: I took $CO/H_2O = 1$ by referring to compilations of interstellar molecular abundances (cf Yamamoto et al. 1983), that is, this value is for gaseous CO to gaseous H_2O. It is physically meaningful to take CO/H_2O in dust. If we take your value of $CO/H_2O \cong 0.1$ in dust, we have the expected abundance ratio of $CO/H_2O \cong 1$ in cometary coma, since CO has longer lifetime than H_2O by a factor of 10. This is still larger than the observed value (Feldman 1985). Besides, for CO, we have to note the abundance variation from comet to comet as pointed out by Feldman.

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