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ON THE FORMATION AND PROCESSING OF CARBON AND NITROGEN COMPOUNDS IN CARBONACEOUS CHONDRITES

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Abstract: On the basis of chemical kinetic consideration, we examine processing of carbon and nitrogen compounds that leads to the linear relation between the logarithmic contents of C and N in carbonaceous chondrites: $\log N = a \log C - b$, found by A. SHIMOYAMA *et al.* (Chem. Lett., **10**, 2013, 1987), where $a > 1$ and b are constants. It is shown that the linear relation results from dissociation of organic polymer in the grains before accretion to parent body-sized objects by radiation that penetrates through the grains such as cosmic ray. Condensation of volatile molecules composed of C and N is also examined as a possible process to form precursors of organic compounds in carbonaceous chondrites. From an analysis of a model thermodynamic system, it is conjectured that this process can also realize the linear relation under certain conditions.

1. Introduction

Carbon and nitrogen compounds in carbonaceous chondrites provide a clue to study formation and processing of extraterrestrial organic matter. It is considered that both elements are mainly present in organic matter (*e.g.*, HAYATSU and ANDERS, 1981; CRONIN *et al.*, 1988). Inorganic carbon such as carbonate makes a minor contribution to the total carbon inventory, although there is evidence for other form (KERRIDGE, 1985). Recently SHIMOYAMA *et al.* (1987) analyzed contents of carbon and nitrogen in Antarctic carbonaceous chondrites, and found that their C and N contents follow a linear relation:

$$\log N = a \log C - b\,, \quad (a > 1)\,, \tag{1}$$

where a and b are constants. This relation also holds (SHIMOYAMA *et al.*, 1987) when including non-Antarctic carbonaceous chondrites analyzed and compiled by GIBSON *et al.* (1971) and by KERRIDGE (1985) (see Fig. 1). The constant $a = \mathrm{d}\ln N/\mathrm{d}\ln C$ equals 1.84 for Antarctic carbonaceous chondrites (with the correlation coefficient $r = 0.97$), and $a = 1.48$ (with $r = 0.89$) when including non-Antarctic ones. Note that $a > 1$. The smaller r for non-Antarctic chondrites may be due to their terrestrial contamination higher than that in Antarctic ones (SHIMOYAMA *et al.*, 1987). The relationship (1) places a constraint upon formation mechanisms and subsequent processing of carbon and nitrogen compounds found in carbonaceous chondrites.

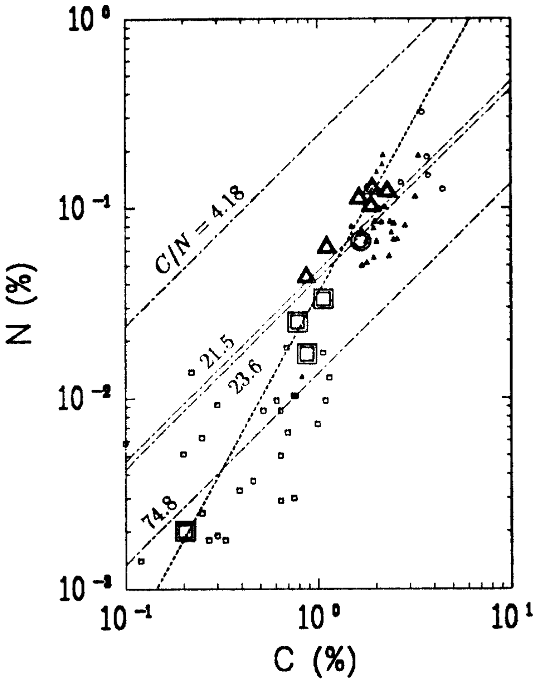

Fig. 1. Carbon and nitrogen contents of carbonaceous chondrites. Circles stand for C1 chondrites, triangles for C2, and squares for C3. The Antarctic ones analyzed by Shimoyama *et al.* (*1987*) *are indicated by large symbols. Data of non-Antarctic chondrites are taken from* Gibson *et al.* (*1971*) *and* Kerridge (*1985*). *In plotting the data, Cold Bockeveld B1 and B2 are omitted because of their terrestrial contamination* (Kerridge, *1985*), *and half of the tabulated contents of Essebi is adopted for the bulk contents since the tabulated values are the ones for matrix, which occupy about a half of the total mass. Least-squares fit to the Antarctic chondrites is shown by the dotted line, which indicates the relation* $\log N = 1.84 \log C - 1.46$. *The dash-dotted lines indicate the lines of the solar C/N ratio of 4.18, and the mean C/N ratio for C1, C2, and C3 chondrites.*

Shimoyama *et al.* (1987) pointed out that the Antarctic carbonaceous chondrites filled the gap between C2 and C3 chondrites so far observed in non-Antarctic carbonaceous chondrites (Gibson *et al.*, 1971) (see Fig. 1). The C/N ratio continuously increases according to the order of

$$(C/N)_{\rm solar} < (C/N)_{\rm C1} \leq (C/N)_{\rm C2} \leq (C/N)_{\rm C3}\,. \tag{2}$$

Geometric means of C/N are 21.5 for C1 chondrites, 23.6 for C2, and 74.8 for C3. The continuous variation of the C/N ratio suggests that C1, C2, and C3 chondrites originate from common materials and the difference results form the difference in the degree of processing.

In this paper, we examine processes that lead to relation (1) on the basis of chemical kinetic consideration. We assume that the measured contents of carbon and nitrogen in carbonaceous chondrites are those of organic polymer, and examine the following two processes separately: (1) irradiation of energetic particles or photons (Section 2), and (2) thermal processes (Section 3). For irradiation, we consider the following two types; (i) a hard type: radiation that penetrates through the grains which are raw materials of carbonaceous chondrites, *e.g.* cosmic ray; (ii) a soft type: radiation that is absorbed at the surface layers of the grains, *e.g.* ultraviolet photons. It should be kept in mind that irradiation is effective to grain-sized bodies before accretion to parent body-sized objects, since the radiation cannot penetrate into large bodies. Among hard radiation, cosmic ray with energy less than ~1 GeV will be effective, since its flux and the energy deposited in passing through matter of unit depth are higher than cosmic ray of higher energy. In thermal processes, we examine relation between the contents of carbon and nitrogen in condensation of volatiles prior to polymer formation. Volatiles of organic composition can be converted into refractory organic polymer by subsequent alteration (*e.g.*, Greenberg, 1982).

2. Irradiation

It is known from experiments that irradiation of energetic particles and photons onto organic polymer produces volatiles by dissociating it together with leaving fragment polymer (WYANT, 1964) on one hand, and produces organic polymers or their precursor radicals from volatiles (STRAZZULLA *et al.*, 1985; GREENBERG, 1982; LACY *et al.*, 1984) on the other hand. We formulate this process as follows: Let us classify C-bearing and N-bearing compounds in the grains into refractory polymers and volatiles. We denote the contents of C or N in the polymers by X_i^{P}, and those in the volatiles by $X_i^{\mathrm{V}} \equiv X_i - X_i^{\mathrm{P}}$, where X_i is the total content of the element $i(=\mathrm{C}$ or $\mathrm{N})$. It is to be noted that the measured carbon and nitrogen contents in carbonaceous chondrites correspond to $X_{\mathrm{C}}^{\mathrm{P}}$ and $X_{\mathrm{N}}^{\mathrm{P}}$, respectively. The coefficient of polymer disscociation is denoted by α_i, and that of polymer production from volatiles by β_i. Both α_i and β_i are parameters involved in the model, and are proportional to the flux of radiation and the reaction cross sections, which are related to G-values used in radiation chemistry.

2.1. Hard type

For hard radiation, the rate of decrease of the polymer content is proportional to the polymer content in the volume of a grain, X_i^{P}, while the rate of increase in the polymer content is proportional to the volatile content X_i^{V}. Thus the time variation of X_i^{P} under irradiation is expressed by

$$\frac{\mathrm{d}X_i^{\mathrm{P}}}{\mathrm{d}t} = -\alpha_i X_i^{\mathrm{P}} + \beta_i (X_i - X_i^{\mathrm{P}}) \,. \tag{3}$$

First we examine the case where all the carbon and nitrogen atoms are initially in the form of volatiles. The initial condition for this case is $X_i^{\mathrm{P}}(t=0)=0$, and the solution of eq. (3) is given by

$$X_i^{\mathrm{P}} = \frac{\beta_i X_i}{\alpha_i + \beta_i}[1 - \exp-(\alpha_i + \beta_i)t] \,. \tag{4}$$

The solution (4) does not satisfy the relation (1) in general, since $\mathrm{d}\ln X_{\mathrm{N}}^{\mathrm{P}}/\mathrm{d}\ln X_{\mathrm{C}}^{\mathrm{P}}$ depends on t. At an early stage of irradiation when $(\alpha_i+\beta_i)t \ll 1$, eq. (4) reduces to $X_i^{\mathrm{P}} \simeq X_i \beta_i t$ and we have $\mathrm{d}\ln X_{\mathrm{N}}^{\mathrm{P}}/\mathrm{d}\ln X_{\mathrm{C}}^{\mathrm{P}} \simeq \mathrm{const.} = 1$. In this case the contents ($X_{\mathrm{C}}^{\mathrm{P}}$, $X_{\mathrm{N}}^{\mathrm{P}}$) vary along a line of $C/N=$const., and fail to satisfy (1) since the inclination of the line a must be >1.

Next we examine another case where all the C and N atoms are initially in the form of refractory polymers, *i.e.* $X_i^{\mathrm{P}}(0)=X_i$. The solution of eq. (3) is given by

$$X_i^{\mathrm{P}} = \frac{X_i}{\alpha_i + \beta_i}[\beta_i + \alpha_i \exp-(\alpha_i + \beta_i)t] \,. \tag{5}$$

The ratio β_i/α_i is determined by the ratio of the G-values. According to the experimental results in radiation chemistry, many organic compounds have much lower G-values for production than those for dissociation (WYANT, 1964), although the species of organic compounds in carbonaceous chondrites are not well characterized. We assume $\beta_i \ll \alpha_i$ in view of these results. Then we obtain, from eq. (5),

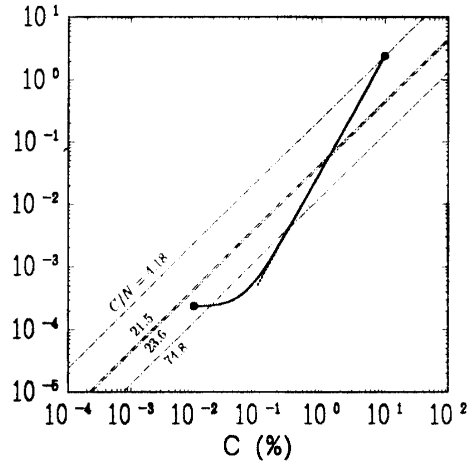

Fig. 2. A locus of the polymer contents $(X_C{}^P, X_N{}^P)$ *for hard radiation, and the dotted line showing the relation (1) for* $0.1 < C(\%) < 10$. *Parameter values adopted here are:* $(\beta_C/\alpha_C, \beta_N/\alpha_N) = (10^{-3}, 10^{-4})$, $\alpha_N/\alpha_C = 1.84$, *and* $(X_C{}^P(0), X_N{}^P(0)) = (9.96, 2.38)$.

$$\frac{\mathrm{d}\ln X_{\mathrm{N}}{}^{\mathrm{P}}}{\mathrm{d}\ln X_{\mathrm{C}}{}^{\mathrm{P}}} \simeq \frac{\alpha_{\mathrm{N}}}{\alpha_{\mathrm{C}}}\,, \tag{6}$$

which is constant, and satisfies the relation (1). A comparison of eq. (6) with eq. (1) leads to $a \simeq \alpha_{\mathrm{N}}/\alpha_{\mathrm{C}}$, the ratio of the dissociation coefficients, which should be larger than unity.

A locus of $(X_{\mathrm{C}}{}^{\mathrm{P}}, X_{\mathrm{N}}{}^{\mathrm{P}})$ with increasing t calculated from eq. (5) is shown in Fig. 2, in which $\alpha_{\mathrm{N}}/\alpha_{\mathrm{C}} = 1.84$ is adopted. Starting from the initial contents $(X_{\mathrm{C}}{}^{\mathrm{P}}(0), X_{\mathrm{N}}{}^{\mathrm{P}}(0)) = (X_{\mathrm{C}}, X_{\mathrm{N}})$, the polymer contents $(X_{\mathrm{C}}{}^{\mathrm{P}}, X_{\mathrm{N}}{}^{\mathrm{P}})$ move downward left (*i.e.* $X_{\mathrm{C}}{}^{\mathrm{P}}, X_{\mathrm{N}}{}^{\mathrm{P}} \downarrow$) along the line given by eq. (6) as t increases. Deviation from the linearity becomes appreciable when $\exp-(\alpha_i+\beta_i)t \leq \beta_i/\alpha_i$. Finally the contents $(X_{\mathrm{C}}{}^{\mathrm{P}}, X_{\mathrm{N}}{}^{\mathrm{P}})$ converge to the steady-state contents of $(X_{\mathrm{C}}{}^{\mathrm{P}}(\infty), X_{\mathrm{N}}{}^{\mathrm{P}}(\infty)) \equiv (\beta_{\mathrm{C}}X_{\mathrm{C}}/(\alpha_{\mathrm{C}}+\beta_{\mathrm{C}}), \beta_{\mathrm{N}}X_{\mathrm{N}}/(\alpha_{\mathrm{N}}+\beta_{\mathrm{N}}))$ at $t=\infty$. The steady-state content $X_i{}^{\mathrm{P}}(\infty)$ is of the order of $(\beta_i/\alpha_i)X_i$, which is a small fraction of the total content X_i, since $\beta_i \ll \alpha_i$. From the condition that the solution (5) realizes the linear relation in the measured interval of the C- and N-contents, we can place a constraint upon the values of $\beta_{\mathrm{C}}/\alpha_{\mathrm{C}}$ and $\beta_{\mathrm{N}}/\alpha_{\mathrm{N}}$. If we assume the initial C/N ratio to be solar ($C/N=4.18$; ANDERS and EBIHARA, 1982), β_i/α_i should be less than $\sim 10^{-3}$. Larger values are allowed, however, if the initial C/N ratio is less than the solar value. In this model, the difference of the C- and N contents in C1, C2, and C3 chondrites is interpreted as the result of the difference in the amount of radiation that hit original grains containing refractory organic polymers; the precursor grains that formed C1 chondrites underwent least irradiation, and those that formed C3 chondrites most irradiation.

The C/N ratio expected from this model is calculated, in the interval where the linearity holds, to be:

$$\frac{X_{\mathrm{C}}{}^{\mathrm{P}}}{X_{\mathrm{N}}{}^{\mathrm{P}}} \simeq \frac{X_{\mathrm{C}}}{X_{\mathrm{N}}} \exp(\alpha_{\mathrm{N}}-\alpha_{\mathrm{C}})t\,, \tag{7}$$

which increases with t for $\alpha_{\mathrm{N}}/\alpha_{\mathrm{C}} > 1$. Thus the C/N ratio increases continuously from C1 to C3 chondrites, which is consitent with the experimental result (2).

Relative amount of radiation that hit precursor grains of C1, C2, and C3 chondrites can be estimated from eq. (7) by using initial contents of C and N, and the observed

C/N ratio. The initial C/N ratio X_C/X_N is not *a priori* given by this model. If we take the inital C/N ratio to be solar ($C/N=4.18$), and the mean C/N ratio of 21.5 for C1, 23.6 for C2, and 74.8 for C3 given in Section 1, then the relative amount of radiation is estimated to be $t_1 : t_2 : t_3 \sim 1:1:2$.

2.2. *Soft type*

For soft radiation, the time variation of the polymer contnt X_i^P is proportional to the contents of C and N in the surface layers of the grains. The rate equation is similar to eq. (3), but X_i^P's in the right-hand side are replaced by $(X_i^P)^{2/3}$, and X_i by $X_i^{2/3}$:

$$\frac{dX_i^P}{dt} = -\alpha_i (X_i^P)^{2/3} + \beta_i [X_i^{2/3} - (X_i^P)^{2/3}] \, . \tag{8}$$

The coefficients α_i and β_i in this case are those for soft radiation. The solution of eq. (8) is given by

$$\frac{\alpha_i+\beta_i}{3} t = X_i^P(0)^{1/3} - (X_i^P)^{1/3} + \frac{X_i^P(\infty)^{1/3}}{2} \ln \left| \frac{(X_i^P(0)^{1/3} - X_i^P(\infty)^{1/3})\{(X_i^P)^{1/3} + X_i^P(\infty)^{1/3}\}}{(X_i^P(0)^{1/3} + X_i^P(\infty)^{1/3})\{(X_i^P)^{1/3} - X_i^P(\infty)^{1/3}\}} \right| , \tag{9}$$

where $X_i^P(0)$ is the initial content of i in the polymer, and $X_i^P(\infty) \equiv \{\beta_i/(\alpha_i+\beta_i)\}^{3/2} X_i$, the steady-state content at $t=\infty$. From solution (9), we obtain

$$\frac{d \ln X_N^P}{d \ln X_C^P} = \frac{\alpha_N+\beta_N}{\alpha_C+\beta_C} \cdot \frac{X_C^P\{(X_N^P)^{2/3} - X_N^P(\infty)^{2/3}\}}{X_N^P\{(X_C^P)^{2/3} - X_C^P(\infty)^{2/3}\}} \, . \tag{10}$$

It can be shown that, for the case of the initial C-bearing and N-bearing compounds being volatile (*i.e.* $X_i^P(0)=0$), we have $d \ln X_N^P / d \ln X_C^P \simeq 1$ from eq. (10), as in the case of hard radiation, which again does not satisfy the experimental result (1) of $a>1$.

For the case where the initial compounds are polymers with $X_i^P(0)=X_i$, we have $d \ln X_N^P / d \ln X_C^P \neq$ constant, and thus soft type radiation does not generally reproduce the linear relation in the wide range of the measured contents of C and N. An example

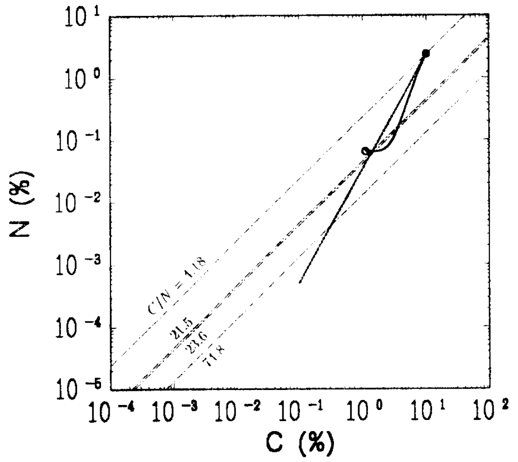

Fig. 3. A locus of the polymer contents (X_C^P, X_N^P) for soft radiation, in which the parameter values adopted are: (β_C/α_C, β_N/α_N)=(0.3, 0.1), and ($d \ln X_N^P / d \ln X_C^P)_{t=0}$=1.84). Others are the same as in Fig. 3.

of the locus is shown in Fig. 3.

3. Thermal Processes

Abundant elements H, C, N, and O are present in solid state mainly as ices of various species in the primordial solar nebula (*e.g.*, FEGLEY and PRINN, 1989) and in molecular clouds (*e.g.*, HAGEN *et al.*, 1980). Laboratory experiments show that organic compounds are produced by processing of the ices (*e.g.*, GREENBERG, 1982). If this is the case for organic compounds in carbonaceous chondrites as well, the contents of C and N consisting of organic compounds in carbonaceous chondrites will reflect composition of the ices condensed prior to the formation of organic compounds. It is thus worth examining relationship between the contents of C and N expected from condensation of the ices, although we have kept it in mind that the C- and N contents may change during subsequent alteration into organic compounds. We discuss an idealized model based on the following assumptions: (i) The system is in chemical equilibrium. (ii) The ices are composed of the same molecular species as the species in gas phase, *i.e.* no chemical reaction at and after condensation. (iii) The ices are regarded as ideal solution, which means that the free energy of formation is expressed by the sum of the free energy of formation of each solid species and that stemming from the entropy of mixing. This model can be also applied to condensation and sublimation of materials other than ice if the above assumptions are valid for the materials in consideration.

In chemical equilibrium between gas and solid under a given total gas pressure P_t, total Gibbs free energy of the system must take minimum, which is attained when the Gibbs free energies of both phases are equal for each species. We denote quantities that are concerned with the gas phase by the suffix "g" and those concerned with the solid phase by "s". The equilibrium condition at a temperature T is expressed by

$$G^{\circ}_{k\mathrm{g}}+k_{\mathrm{B}}T\ln P_k=G^{\circ}_{k\mathrm{s}}+k_{\mathrm{B}}T\ln\frac{n_{k\mathrm{s}}}{\sum\limits_{l=1}^{L} n_{l\mathrm{s}}}\,, \quad (k=1,\ldots,L)\,, \tag{11}$$

where $G^{\circ}_{k\mathrm{g}}$ and $G^{\circ}_{k\mathrm{s}}$ are the Gibbs free energies per molecule of the k-th species at a standard pressure, P_k the partial pressure of the k-th species in the gas phase, $n_{k\mathrm{s}}$ a concentration of the k-th species in the solid phase, L the number of species in consideration, and k_{B} the Boltzmann constant. The second term on the right-hand side indicates the free energy resulting from the assumption (iii) stated above. The change in the Gibbs free energy $\Delta G^{\circ}_k\equiv G^{\circ}_{k\mathrm{s}}-G^{\circ}_{k\mathrm{g}}$ is related to the vapor pressure of the pure solid of the k-th species, $P_{k\mathrm{e}}$, as

$$\Delta G^{\circ}_k=k_{\mathrm{B}}T\ln P_{k\mathrm{e}}\,. \tag{12}$$

P_k is expressed in terms of P_t by

$$P_k=P_t\frac{n_{k\mathrm{g}}}{\sum\limits_{l} n_{l\mathrm{g}}}\,, \tag{13}$$

where $n_{k\mathrm{g}}$ is the concentration of the k-th species in the gas phase. With the use of eqs.

(12) and (13), we obtain

$$\frac{n_{k\mathrm{s}}}{\sum_l n_{l\mathrm{s}}}=\frac{P_\mathrm{t}}{P_{k\mathrm{e}}}\frac{n_{k\mathrm{g}}}{\sum_l n_{l\mathrm{g}}} \tag{14}$$

from eq. (11). We define γ_k, the relative abundance of the k-th species, by

$$\gamma_k\equiv\frac{n_{k\mathrm{g}}+n_{k\mathrm{s}}}{n_\mathrm{tot}} \tag{15}$$

with

$$n_\mathrm{tot}\equiv\sum_l (n_{l\mathrm{g}}+n_{l\mathrm{s}})\ , \tag{16}$$

and f_k, the fraction of the k-th species in a solid phase, by

$$f_k\equiv\frac{n_{k\mathrm{s}}}{n_{k\mathrm{g}}+n_{k\mathrm{s}}}\ . \tag{17}$$

Then we have

$$n_{k\mathrm{s}}=f_k\gamma_k n_\mathrm{tot}\ ,\quad n_{k\mathrm{g}}=(1-f_k)\gamma_k n_\mathrm{tot}\ . \tag{18}$$

Thus eq. (14) is rewritten as

$$\frac{f_k}{Y}=\frac{P_\mathrm{t}}{P_{k\mathrm{e}}}\cdot\frac{1-f_k}{1-Y}\ ,\quad (k=1,\ldots, L)\ , \tag{19}$$

with

$$Y\equiv\sum_l f_l\gamma_l\ . \tag{20}$$

The fraction f_k of the k-th species condensed into a solid is given by solving the simultaneous eq. (19).

The content X_i of an element i in a solid solution is given by

$$X_i=\sum_l \nu_{il}f_l\gamma_l\ , \tag{21}$$

where ν_{il} is the stoichiometric coefficient for the element i contained in the l-th species. If the element i contained in one species k as are the cases discussed later, ν_{il} is zero for species other than $l=k$. In this case, we obtain

$$X_i=\frac{P_\mathrm{t}}{P_{k\mathrm{e}}(i)}\cdot\frac{\nu_{ik}\gamma_k Y/(1-Y)}{1+\dfrac{P_\mathrm{t}}{P_{k\mathrm{e}}(i)}\cdot\dfrac{Y}{1-Y}}\simeq\frac{P_\mathrm{t}}{P_{k\mathrm{e}}(i)}\cdot\frac{\nu_{ik}\gamma_k Y}{1-Y}\ , \tag{22}$$

from eqs. (19) and (21), where $P_{k\mathrm{e}}(i)$ is the vapor pressure of k-th species containing the element i. The last expression is valid at temperatures where

$$\frac{P_\mathrm{t}}{P_{k\mathrm{e}}(i)}\cdot\frac{Y}{1-Y}\ll 1\ , \tag{23}$$

holds, which implies that the fraction of the k-th species in solid solution is much less

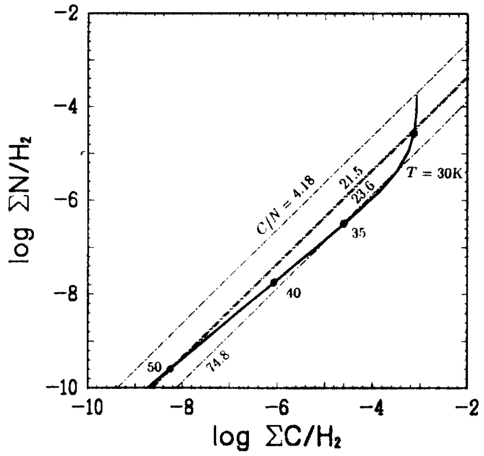

Fig. 4. Variation of the contents of C and N in solid solution with the temperature T, which is shown along the curve, for the H_2-H_2O-CO-N_2 system for the total pressure of 10^{-5} atm. The dot-dashed lines are the lines of C/N = constant whose values are attached to the lines.

than that of other (less volatile) species ($f_k\gamma_k \ll Y \simeq$const., *i.e.* dilute solution). If we note that $P_{k\mathrm{e}}(i)$ is approximately expressed in terms of the heat of sublimation ΔH_k at $T=0$ K by

$$\ln P_{k\mathrm{e}}(i) \simeq -\Delta H_k(i)/k_\mathrm{B}T + \mathrm{const.}\,, \tag{24}$$

according to the Clapeyron-Clausius relation, we obtain

$$\frac{\mathrm{d}\ln X_i}{\mathrm{d}\ln X_j} \simeq \frac{\Delta H_k(i)}{\Delta H_l(j)} = \mathrm{const.} \tag{25}$$

This result indicates that the inclination a reflects volatility of the species that contain the elements in consideration.

We consider a system composed of molecular species of H_2, He, H_2O, CO and N_2, and of a solid solution of H_2O, CO, and N_2, where the elemental abundance is solar. This composition is realized in the solar nebula (FEGLEY and PRINN, 1989). It is to be noted that carbon is contained mainly in CO and nitrogen in N_2, and that both species are musch volatile than H_2O. Figure 4 shows the variation of the contents of C and N with the temperature T for a total gas pressure $P_\mathrm{t} = 1 \times 10^{-5}$ atm. The contents of C and N in the solid solution increase with decreasing T. It can be seen from the figure that the linear relation between log C and log N is realized in the temperature range of $T \geq 35$ K. In this case, the inclination equals $a=0.84<1$, contrary to the experimental results, since $\Delta H_{N_2} < \Delta H_{CO}$. It is possible, however, that $a>1$ when N-bearing species are less volatile than C-bearing species. This situation is realized, for example, in an H_2O-NH_3-CH_4 system, proposed for the subnebulae of the Jovian planets (FEGLEY and PRINN, 1989).

To make quantitative comparison with the experimental results, however, we will have to take into account chemical fractionation of C-bearing and N-bearing species in the formation process of complex organic polymer from ices as stated previously. Furthermore, the study of realistic thermodynamic systems including chemical reactions (*e.g.*, HAYATSU and ANDERS, 1981) is necessary, together with the study of alteration processes that produce organic compounds from ices consisted of H, C, N, and O.

4. Conclusion

(1) Dissociative processing of organic polymers in grains by hard radiation (*e.g.* cosmic ray) can reproduce the experimental results (1). This processing should work for individual grains prior to their accretion onto parent bodies of carbonaceous chondrites, and not for bodies of meteorite size. The inclination a of the experimental line indicates the ratio of the coefficients of dissociation α_N/α_C, which should be larger than unity. In this kind of processing, α_i must be much larger than the coefficient of production β_i in order to realize the linearity in the measured interval of C and N contents, implying that the observed contents of C and N in carbonaceous chondrites are a small fraction of the initial contents X_i. If we assume the initial C/N ratio to be solar, β_i/α_i is estimated to be less than $\sim 10^{-3}$.

The theory can be tested by an irradiation experiment of powdered carbonaceous chondrites with protons and measurements of the difference in the contents of carbon and nitrogen in the pre- and post-irradiated powders.

(2) The experimental result (1) with the inclination $a > 1$ does not arise form polymerization of volatiles by irradiation. This process, however, could be responsible for the formation of the original organic refractories or their precursor radicals from volatiles. It is characteristic that the evolution of the contents of C and N in this case keep $C/N \simeq$const. for both hard and soft radiations.

(3) It is possible that thermal processes can lead to the relation (1) under certain conditions, especially when the C-bearing and N-bearing species are minor components in solid solution. The inclination a is determined by the ratio of the heat of sublimation of C-bearing compounds to that of N-bearing compounds. To confirm this conjecture, however, thermal processes including chemical reactions should be studied for more realistic thermodynamic systems.

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