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Possible origin of nitrogen in the Earth's atmosphere

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Summary. — The details on the origin of nitrogen, which exists so abundantly in the Earth's atmosphere, are missing. An attempt to give a possible answer to the question was interpreted to be the result of endothermic nuclear transmutation of carbon and oxygen atom pairs in carbonate lattice of mantle containing crust, $^{12}\text{C} + ^{16}\text{O} + 2e^* + 2\nu \rightarrow 2\ ^{14}\text{N} + ^4\text{He}$, with help of electropionic attraction effect (48% shrinkage) due to the excited electron capture and neutral pion catalysis. The excited electrons were generated by rapid fracture or sliding of carbonate crystals due to volcanic earthquake, and plenty of neutrinos were derived from the universe, mainly from the young sun. The formation of nitrogen would have continued for 1.3 billion years from 2.5 to 3.8 billion years ago in the Archean era, until the active volcanism or storm of neutrinos ceased. The possible nuclear transmutation rate of nitrogen atoms could be calculated as 2.3×10^6 atom/s.

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1. – Introduction

When we examine the composition of the atmosphere of all solar planets (Mercury, Venus, the Earth, Mars, Jupiter, Saturn, Uranus, Neptune and Pluto) and their sixty-one satellites, we notice that the nitrogen composition of these planets and satellites is considerably low ($\sim 6\%$) with the exception of the Earth, Titan and Triton, as presented in table I [1,2]. The Earth has the extremely high composition of nitrogen of 78.8%. The amount of Titan and Triton would be smaller by $2 \times 10^{-6}\%$ and $5 \times 10^{-6}\%$ than the Earth's one, respectively, as can be assumed from their mass and nitrogen composition and atmospheric pressure [3]. Thus it is clear that the Earth is an exceptional planet, so far as an amount of nitrogen is concerned.

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TABLE I. – *Major atmospheric compositions, pressures and masses of solar planets and their satellites* [1,2].

Planet (Satellite)	Pressure (Pa)	Mass (kg)	Major atmospheric composition (%)						Pressure (atm) Ar
			N ₂	CO ₂	O ₂	H ₂	He	CH ₄	
Venus	9.2×10^7	4.87×10^{27}	3.4	96.4					6.7×10^{-3}
Earth	1.0×10^5	5.98×10^{27}	78.1	0.2	20.9				9.3×10^{-3}
Mars	700	6.42×10^{26}	2.7	95.3	0.1				7×10^{-5}
Jupiter	...	1.90×10^{30}				89.8	10.0	0.2	
Saturn	...	5.69×10^{29}				96.3	3.3	0.4	
Uranus	...	8.68×10^{28}				85	15	2	
Neptune	...	1.02×10^{29}				81	19		
Pluto	?	1.29×10^{25}							
(Titan)	1.5×10^5	1.35×10^{26}	65–98			0.5–1		0.5–20	
(Triton)	1.6	2.14×10^{25}	~ 100						

The significant question on the origin of nitrogen abundance in the Earth has not been entirely resolved, and has been overlooked without consensus. Some research groups [4,5] investigated the nitrogen contents of various rocks, but no previous paper has treated this subject, as far as we know. For the origin of nitrogen, the present consensus, based on chemical and radioactive isotopic analyses, is that nitrogen has been discharging for long time from the Earth's interior portions (crust, mantle and core) to the atmosphere through volcanic and hydrothermal activity [6], or that it has come from catastrophic degassing due to the impact of planetesimals [7]. However, since it is considered that the solar interior rocky planets (Mercury, Venus, the Earth and Mars) had formed by accretion of solid planetesimals in the same region of the developing solar system at almost the same time [8], we cannot image that the high composition of nitrogen in the Earth is originated in the planetesimals, as far as Mars has poor nitrogen atmosphere. Indeed, there is no evidence that the planetesimals had abundance of nitrogen.

On the other hand, although the nitrogen composition of Venus is $\sim 3.4\%$, the total amount of nitrogen of Venus is almost the same as that of the Earth [9], because of Venus's high atmospheric pressure (~ 93 bar). Table I also shows that the amount of argon of the Earth compares well with that of Venus, thus suggesting a very similar composition, while that of Mars is much smaller. These results suggest a similar evolution for atmospheric nitrogen and argon in the Earth and Venus. Thus we can put severe constraints on the possibility of a biological cause as the origin of nitrogen in the deoxidizing atmosphere of the early stage in the Earth. In fact, a terrestrial nitrogen biogeochemical cycle, which is responsible for the release of nitrogenous volatiles from soils and aquatic environments, is premised on the existence of nitrogen-fixing bacteria such as phototrophic cyanobacteria in natural waters, heterotrophs in soils and symbionts associated with plants, in oxidizing atmosphere [10,11]. One aim of this study is to investigate the origin of nitrogen that is the constituent element of such organic matters. Even if matter existed in the Archean

era, it is difficult to explain the striking increase of nitrogen without transmutation of nitrogen by bacteria. In this study, we focus our discussion on the Earth for the formation mechanism of nitrogen, because of deficient geological data of Venus. Furthermore, since there is no direct available geophysical data for the formation of nitrogen, the study of this problem must begin with an idea on how nitrogen created on the basis of the history of the Earth's atmosphere, using circumstantial evidence.

Our theory is based on the assumption that the presence of nitrogen increases with decreasing percentage of carbon dioxide before the generation of oxygen due to photosynthesis. Our interest lies in studying the possible formation of nitrogen by the dynamic weak interaction among carbon and oxygen atoms in carbonate crystals in the upper mantle containing crust. In this paper, a new interaction model is proposed with the help of electropionic attraction due to the electron capture and neutral pion catalysis. We have already reported a neutral-pion-catalyzed fusion based on the collective resonance (excitation) effect of electrons derived from palladium [12],



where ${}^2\text{D}$ is the deuteron. The central problem of this work is to explain how carbon and oxygen atoms react with each other when there exists a potential barrier so high that it should not be possible for both nuclei to come near enough to interact.

2. – Appearance of nitrogen in early earth

To estimate the generation time of nitrogen in Earth's history, we first note a composition change of various atmospheric gases from the primitive Earth to the present one. It is generally agreed that the terrestrial planets have formed by accretion of solid materials that condensed from the solar nebula about 4.56 billion years ago [13]. It is believed that the present, secondary atmosphere was generated from volatile compounds (mainly CO_2 , H_2O) contained within the solid planetesimals from which the Earth formed, after the primary, captured atmosphere was released [14]. From determination of the ratios of the isotopes in the mantle rocks [15], the composition of the primitive atmosphere around 4.3 billion years ago was most certainly dominated by carbon dioxide, with abundant water. The 10 to 20 bar carbon dioxide atmosphere had existed during the first hundred million years of the Earth's history [16]. Trace amounts of methane, ammonia, sulfur dioxide and hydrochloric acid were also present, but there was no oxygen [17]. The evolving ammonia and methane gases were highly reactive and had short lifetimes in the atmosphere. Even if ammonia was resolved into nitrogen and hydrogen or the reduction of nitrate (NO_3^-) to nitrogen gas occurred by denitrifying bacteria under anaerobic conditions, we could not explain the occurrence of abundant nitrogen. After about 4.3 billion years, carbon dioxides had dissolved mainly in the hot ocean and formed carbonates by virtue of the weathering of igneous rocks [18], because the ocean already existed 3.8 billion years ago [19].

We here describe three research groups' results for variation of carbon dioxide. Owen *et al.* [20] have calculated the CO_2 surface pressure PCO_2 from 4.25 billion years ago to the present time for the evaluation of enhanced CO_2 greenhouse effect. Their result showed 0.31, 0.07 and 0.009 bar 4.25, 3.5 and 2.0 billion years ago, respectively. Ohmoto [21] has proposed that the atmospheric composition of carbon dioxide in the Archean era was fairly high (0.3, 0.1 and 0.04 bar 3.4, 2.5 and 2 billion years ago, respectively) from sulfur isotope evidence of pyrites formed by bacterial reduction of seawater

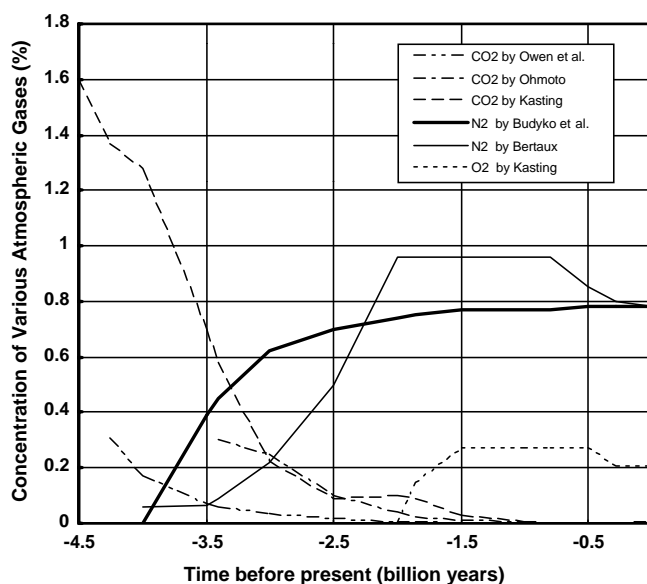


Fig. 1. – The composition change of atmospheric CO₂, N₂ and O₂ gases from the primitive to present times of the Earth.

sulfate. Kasting [22] has reported the variation of carbon dioxide pressure from 4.5 billion years ago to the present time, associated with atmospheric temperature. His results are 2, 0.7 and 0.1 bar 4.5, 3.5 and 2.5 billion years ago on the average, respectively. The common point of the three research groups is a parabolic decrease of carbon dioxide in the Archean era.

On the other hand, Mojzsis *et al.* [23] have reported that organic matter existed as many as 3.8 billion years ago, from carbon-isotope measurement of the Isua formation in West Greenland. Since all present living organisms of the Earth consists of biological macromolecular sources such as amino acid sugars, proteins and nucleic acids, the existence of organic matters provides strong evidence for the presence of nitrogen. Budyko *et al.* [24] have assumed that the nitrogen atmosphere started to form from about 4 billion years ago, then increased steadily by more than 50%, about 80% and about 95% of the present content 3.5, 3.0 and 2.0 billion years ago, respectively, based on the calculation of carbon dioxide. Bertaux [25] has simulated the evolution of terrestrial atmosphere; the composition of nitrogen was 0.06, 0.22 and 0.96% 4.0, 3.0 and 2.0 billion years ago, respectively. Thus we can image a striking increase of nitrogen pressure in time scale from around 4.0 to 2.0 billion years ago, although the precise composition change of nitrogen is not well established yet. Since atmospheric nitrogen is an inert gas, its quantity has remained constant through the ages.

In addition, oxygen was essentially absent in the primitive atmosphere: the prevalence of anoxic conditions during the first 2.6 billion years of the Earth's history is documented by the preservation in ancient detritals of mineral like uranite and pyrite [26], which are unstable in oxidizing environments. Kasting [27] has estimated an abrupt increase in oxygen from around 2 billion years ago. The generation of atmospheric oxygen from around 2 billion years ago is mainly due to the result of photosynthetic activity by the organic matter [28], even if a considerable amount of oxygen dissolved in the hot seawater

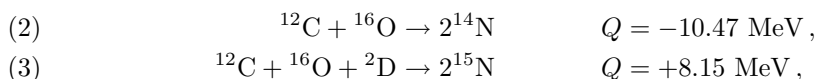
of the Archean era. From the above-mentioned data, the composition change of the three kinds of representative atmospheric gases in the history of the Earth is summarized in fig. 1. Although there are other processes such as a t-tauri phase loss of the early atmosphere, the many different atmospheric escape mechanisms, and the complexities of the different outgassing timescales, fig. 1 will be considered roughly right.

When we look carefully at the variation of the various gases, we find that the decrease in carbon dioxide composition is accompanied by a gradual accumulation of nitrogen in the atmosphere, and nitrogen is the atmosphere's principal component at the end of the epoch of the oxygen-free atmosphere. This view agrees with the estimation by Bydyko *et al.* [24]. Thus, consumption of the carbon dioxide seems to correlate with formation of nitrogen. Therefore, we restrict our discussion to the formation period of nitrogen between 2.5 and 3.8 billion years ago in the Archean era.

3. – Dynamic interaction between C and O atoms in magnesium carbonate and formation of nitrogen

We next consider a necessary condition for the formation of nitrogen. As far as the origin of the nitrogen of the Earth was not contributed to by the collision of planetesimals with abundance of nitrogen, we must inquire into the early Earth. As can be seen from the calculation by Budyko *et al.* [24], the appearance of nitrogen is strongly connected to the disappearance of carbon dioxide. Generally speaking, the disappearance comes from the formation of rocks containing carbon dioxides in the hot sea. Therefore it is expected that the formation of nitrogen is distinctively associated with the formation of the carbonaceous rocks. Indeed, the core with the rocks had already formed at an epoch that ranges from 4.44 to 4.41 billion years ago due to the separation from magma-ocean [27]. Oxides, carbonates, sulfides were representative of the rocks in the early Earth. Among these rocks, the carbonaceous rocks are clearly CO₂-reservoirs. The main parts of carbonaceous rocks are distributed on and near the Earth's surface, and the remainder of rocks sinks into the deep mantle by plate tectonics. Thus we next consider the dynamic reaction of the carbonates in the mantle containing crust, associated with formation of nitrogen.

Here it should be noted that Jones *et al.* [29] suggest a possibility of cold nuclear fusion in the mantle water reserve of the Earth and in the core of Jupiter, from the observations of high (~ 16 times) helium isotope concentrations ratio ($^3\text{He}/^4\text{He}$) at the volcano area [30] and radiation of excess heat. By analogy we infer that the carbon and oxygen atoms in carbonate crystals interact to form nitrogen. In fact, to one's surprise, the mean binding energy per nucleon of stable nitrogen nucleus (7.5 MeV) is lower than those (7.7 and 8.0 MeV) of carbon and oxygen ones, respectively [31]. The binding of carbon and oxygen nuclei is called "nuclear transmutation". These reactions are as follows for ^{14}N and ^{15}N isotopes [32]:



where the Q values represent the amount of energy absorbed or released per reaction. Equations (2) and (3) are endothermic and exothermic reactions, respectively. This means eq. (2) accelerates, according to increasing temperature. In the present Earth, the natural abundance of the ^{14}N isotope is about 99.635%, whereas that of the ^{15}N is only

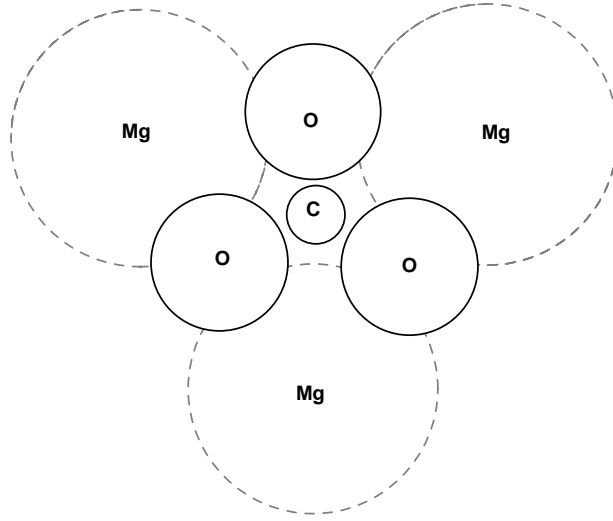


Fig. 2. – Configuration of CO₃ group atoms in (111) magnesite crystal planes. The CO₃ group lies exactly midway between the planes on which the Mg atoms lie, and in consequence the (111) planes have a structure Mg...CO₃...Mg [35]. The large, middle and small open circles are Mg, O and C atoms.

about 0.365% [33]. Thus we consider the ¹⁴N isotope formation alone in eq. (2), as the main nitrogen isotope of the Earth.

To investigate the reaction in carbonate rocks, we must select a stable carbonate with smaller lattice constant under high pressure. In general, the stability of the carbonate MCO₃ (M: alkaline element) comes from that of the CO₃ group configuration, which increases with increasing ionic radius of M, while the lattice constant of MCO₃ increases with increasing radius of M. Since the carbonate deposited near the surface of the Earth is a candidate material available for the nuclear transmutation, dolomite, CaCO₃·MgCO₃ must be taken up as the main carbonate of the Archean era [34]. However, we select a magnesium carbonate MgCO₃ (dolomite) instead of dolomite due to the lack of crystal lattice data of dolomite under a high pressure over 50 GPa.

The magnesite is an analogous material of calcite CaCO₃ with rhombohedral crystal form. We note a triangle configuration of the CO₃ group atoms on a (111) plane of MgCO₃ crystals (fig. 2) [35]. The three oxygen atoms in the CO₃ triangle are symmetrically situated around the carbon atom. When the magnesite is compressed at a high pressure of 27 GPa [36], corresponding to the pressure condition of the lower-upper mantle boundary, the O-O distance is 0.248 nm. From the triangle configuration of CO₃, the C-O distance ($2r_1$) along three $[1\sqrt{3}0]$ directions can be calculated as 0.143 nm. Since the ideal C-O distance ($2r_0$) is 0.147 nm [37], the shrinkage ratio is 2.6%. However, the distance $2r_1$ is too large in comparison with the distance (~ 0.079 nm [29]) required for the dynamic nuclear reaction. Therefore, we must consider another attraction mechanism accelerating the confinement of carbon and oxygen nuclei in the magnesite lattice.

4. – Formation of ^{14}N

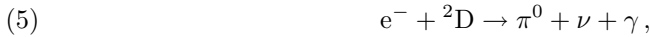
Here we consider an electropionic attraction effect for the possible nucleonic reaction. Because the probability of nuclear tunneling through the barrier of the internuclear Coulomb repulsion is very small, we must consider an acceptable explanation for the dynamic mechanism. The nitrogen ^{14}N is a seven- np state whose isospin wave function is antisymmetric, where n and p are the neutron and proton, respectively. The carbon ^{12}C nucleus is composed of six protons and six neutrons. The first-order internuclear force among protons and neutrons in the carbon nucleus is mediated by eighteen charged pions, π^\pm and eighteen neutral pions π^0 . Similarly, the oxygen nucleus is composed of eight protons and eight neutrons. These sixteen nucleons in the oxygen ^{16}O nucleus are combined by the attractive mediation of thirty-two charged and thirty-two neutral pions.

On the other hand, twenty-five charged and twenty-four neutral pions mediate the nitrogen nucleus. Therefore when the confinement, *i.e.* transmutation, of the carbon and oxygen nuclei occurs at the necessary close proximity, the formation of two nitrogen nuclei from carbon and oxygen nuclei requires transportation of one proton and one neutron from the oxygen nucleus to the carbon one. According to the theory of fundamental processes [38], it is necessary for the reaction to move seven charged pions and six neutral pions from the oxygen nucleus to the carbon one, and to leave two neutral pions in the oxygen one for the modulation of the n , p force in nitrogen nucleus.

Thus the dynamic interaction of interest is presented by the following nuclear reaction:



Pions are responsible for all low-energy nuclear interactions [39]; the pions within the nucleus allow the nucleonic species to bind together and transmute with each other [29]. Kenny [40] has also pointed out that the nucleus is charged by electron capture into neutral pion and then decays to produce heat energy. In the case of the deuteron with lower mass, we have reported the following form [41]:



where ν and γ are the neutrino and photon, respectively. Since the neutral pion does not experience a Coulomb barrier, it can more easily enter within the effective nuclear force field of C-O pairs at close proximity, in comparison with the charged pions. The effect of a neutral pion on the nuclear reaction in solid states has been almost overlooked, as long as we know.

The neutral pion in eq. (4) is provided by a fundamental process, which is an electromagnetic interaction:



From isospin symmetry, the photon in eq. (6) is produced by emission of excited electrons derived from magnesium in the carbonate [38]:

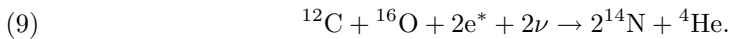


where e^* is an excited electron. The excited electrons can be generated by rapid fracture and sliding [42] of carbonate crystals. Indeed, it is estimated that the continental crust

was often cracked by volcanic earthquake at the Archean era [17]. In a previous paper [12], we reported the following formula for fusion of helium:



When we consider the nuclear reaction (4), using the low-energy nuclear interaction based on the electropionic attraction, we can make use of reactions (5)–(8). Hence, we get the following form for the weak dynamic interaction:



The neutrinos, which have no electric charge, interact very weakly with matter. They can be captured at all interior portions of the Earth. Plenty of neutrinos would have come from the universe, mainly from the sun [43] or flare of t-tauri stars [44] in the Archean era. The storm of neutrinos from the young sun might have attacked the primitive earth, because it is argued that the cyclic thermal expansion of the solar core may be connected to the cyclic occurrence of the glacial epoch [45]. Although the neutral pion is recognized as a nonexchange part in the nuclear strong field, it clearly plays a decisive role in nuclear transmutation of carbon-oxygen nuclei pairs as catalyst of dynamic nuclear interaction. Therefore both the creation of many electrons and the capture of plenty of neutrinos are sufficient condition for nitrogen formation.

According to the symmetry meson theory [46] associated with a binding energy that tends to clump bosons together, we can write the interaction energy of two nucleons at separation r as follows:

$$(10) \quad U(R) = -\frac{C}{r^4},$$

where C is the coupling constant. Since the addition of two neutral pions increases the attraction force by a factor of fourteen in interaction range, we get a condensed C-O distance $2r_2$,

$$(11) \quad 2r_2 \cong 0.517 \times 2r_1 = 0.517 \times 0.974 \times 2r_0 = 0.074 \text{ nm},$$

on the basis of eq. (10). This value would have lead to fusion of carbon-oxygen nuclei before nitrogen formation. The fusion rate R was calculated as $R = 9.254 \times 10^{-6}$ f/s/cc [47].

Although we considered formation of ${}^{14}\text{N}$ in the lower mantle, there is another possibility that the nuclear fusion of the ${}^{15}\text{N}$ isotope occurs at the core of the Earth. This reaction is an exothermic one, accompanied by a release of energy. Indeed, the higher isotope ratio of ${}^{15}\text{N}/{}^{14}\text{N}$ has been observed as an evidence of the high-temperature fluid-rock interactions [48], suggesting nuclear fusion of ${}^{15}\text{N}$ in the core. Even if this reaction had occurred, the amount of products would have been very small, because of the extremely small amount of deuterons in the core.

5. – Nuclear transmutation rate of nitrogen

The mean binding energy/nucleon for the fusion product, *i.e.* activated state product, formed from carbon and oxygen nuclei is 15.6 MeV, being considerably higher than the stability limit of 8.8 MeV of natural nuclei [49]. Since the product is the shape isomer

in the nucleus, it splits into two nitrogen nuclei by spontaneous fission with lifetimes shorter than that of the ground state by a factor of 10^{26} or more [31]. In this stage, four neutral pions must be liberated from the product:



The neutral pions disintegrate rapidly ($< 10^{-15}$ s) into eight photons [37], which convert immediately to heat in the mantle containing crust,



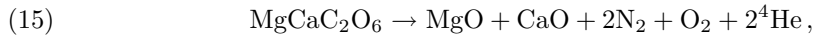
Such nitrogen atoms diffuse toward the grain boundaries of the carbonate crystals, actually dolomites, and then form nitrogen molecules at the crystal boundaries. Then, nitrogen gases discharge by volcanism into the atmosphere. The Earth's nitrogen is present in four pools [50]: lithospheric (10^{23} g), atmospheric (3.9×10^{21} g), terrestrial (4.8×10^{17} g) and aquatic (2.3×10^{19} g). Then we can calculate the formation rate R' for 1.3 billion years from 2.5 to 3.8 billion years ago in the Archean era:

$$(14) \quad R' = \frac{1.34 \times 10^{23}}{14 \times 1300000000 \times 365 \times 24 \times 60 \times 60} \doteq 2.3 \times 10^6 \text{ atom/s}.$$

In this study, we investigated the possibility of nitrogen formation for carbonates existed in the upper mantle containing crust. However, if the plenty of the carbonates had existed in the lower mantle of the early Earth, the reaction rate for nitrogen formation would have accelerated by high temperature and high pressure. The detailed calculation results based on the electrostatic screening attraction effect and high-temperature effect is described in Appendix.

6. – Disappearance of CO_3 group atoms in carbonates and formation of mixed oxides

The formation of nitrogen and helium would continue for 1.3 billion years, until the active plate volcanism or storm of neutrinos ceased. When the CO content in carbonate crystals such as MgCaC_2O_6 or $\text{M}(\text{Mg}, \text{Ca})\text{CO}_3$ was completely exhausted, the following reactions would occur:



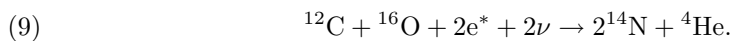
The remainder oxygen atoms and $\text{M}(\text{Mg or Ca})\text{O}$ react with other mixed oxides, especially SiO_2 to form various kinds of oxides under elevated temperature and high pressure. This process is known as carbonate metamorphism. Trace amount of carbon dioxide in the present atmosphere is a final step for formation of nitrogen. The total amount of carbonate associated with the reaction can be estimated as 8.07×10^{23} g and 9.58×10^{23} g for magnesite and calcite, that are 1.4×10^{-7} and 1.6×10^{-7} of the Earth's mass, respectively. The volatile helium gases in reactions (15) or (16) would be released from the Earth's atmosphere to the universe.

For the nitrogen formation of Venus, we guess that a similar formation process had occurred in the Venusian mantle at an ancient period before plenty of water had liberated

from Venus into space. It is because mass and size of Venus and the Earth are nearly the same and the geological interior constitution (a liquid core, a mantle and a rocky crust) of Venus is therefore thought to resemble the Earth's one, except for much less water and no magnetic field [51]. Mars could not hold abundant water and carbon dioxide for the formation of carbonates for the necessary length of time. This means lacking of the carbonate-silicate geochemical cycle system for Mars. Lacking of volcanism and less internal heat in Mars can also account for scanty of nitrogen, because of the smaller surface area and surface gravity of Mars compared with the Earth [52]. In this interesting area, further work such as mass spectroscopic analysis of nitrogen for carbonates under elevated temperature, dynamic high pressure and radiation of neutrinos is called for.

7. – Conclusion

When we examine the composition of the atmosphere of all solar planets and satellites, we notice that the composition of the Earth is extremely rich compared with that of the others. The decrease in the carbon dioxide percentage in the Archean era of the Earth was accompanied by a gradual accumulation of nitrogen in the atmosphere. The formation of nitrogen is distinctively associated with the existence of rocks and mantles containing carbon dioxide. Thus we assumed that the formation of nitrogen was ascribed to a possible endothermic nuclear transmutation of carbon and oxygen nucleus pairs confined along three $[1\sqrt{3}0]$ directions in a (111)-plane of rhombohedral MgCO_3 crystals existing in the upper mantle crust:



The confinement was explained by an attraction effect (48% shrinkage, $2r_2 = 0.047$ nm) based on catalysis of neutral pions thanks to the combined effect of electron emission derived from the carbonate lattice and plenty of neutrinos coming from the universe, especially from the young sun. The critical distance for the transmutation would be less than 0.079 nm. After confinement the activated state product ${}^{12}\text{C}{}^{16}\text{O}$ immediately undergo fission to two ${}^{14}\text{N}$ nitrogen atoms, accompanied by liberation of helium. The formation rate of nitrogen atoms at upper mantle containing of crust could be calculated as 2.3×10^6 atom/s. The formation of nitrogen and helium could continue for 1.3 billion years in the Archean era, until the active volcanism or storm of neutrinos ceased. The total amount of carbonate associated with the reaction can be estimated as 8.07×10^{23} g and 9.58×10^{23} g for magnesite and calcite, being 1.4×10^{-7} and 1.6×10^{-7} of the Earth's mass, respectively.

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The author wishes to express his gratitude to Dr. K. KAWADA for a peg to hang a discourse on.

APPENDIX

1. – Confinement by screening of condensed electrons

First it should be noted that at very high pressure in excess 0.5 Mg/cm^3 the individual electron shell structures of the atoms vanish and are replaced by a statistical distribution

of the electrons in the field of the atomic nuclei [53]. Such electrons in the outer shell behave as if they were free particles [54]. Hence by analogy we infer that the outer-shell electrons of carbon and oxygen atoms in MgCO_3 lattices behave as free electrons, and the resulting screening effect serves as relief of Coulomb repulsive force between carbon and oxygen nuclei. Since such an interelectron Coulomb interaction alters the electron shell behavior under the action of the electromagnetic field, we apply the Thomas-Fermi (abbreviated as TF hereafter) approximation to the screening effect for the MgCO_3 lattice of 55 GPa [36] at around 1200 km below the surface of the Earth. For a many-electron atom, the TF theory [55] gives (in atomic units)

$$(17) \quad V(r) = -\frac{Ze^2}{r}\Phi(r/b),$$

where $V(r)$ is the potential energy, and Z is the atomic number. The density of MgCO_3 at 55 GPa can be estimated as 10.09 Mg/m^3 , using the density-pressure relationship [56]. Thus we can assume the C-O bond distance as $2r_1 = 0.0948 \text{ nm}$, that is 36% less than the equilibrium one ($2r_0 = 0.147 \text{ nm}$) [57]. However, this distance is still large in comparison with critical distance (0.079 nm) [29].

2. – Rate of reaction at high temperature

Since the temperature at around 1200 km below the surface in the present Earth is estimated as about 2473 K [58], we then consider the effect of temperature on the reaction rate k . The rate can be expressed by the Arrhenius equation [59]

$$(18) \quad k = \frac{k_B T f_N^2}{h f_C f_O} e^{-E/RT},$$

where f_C, f_O, f_N are partition functions of ^{12}C , ^{16}O and ^{14}N , respectively, and k_B , R and E are the Boltzmann and gas constants and the activation energy of the reaction, respectively. Since $f_C \doteq f_O \doteq f_N$, we can write the ratio of the rates at temperatures T_0 and T_1 as follows:

$$(19) \quad \frac{k_1}{k_0} = \frac{T_1}{T_0} e^{\frac{E}{R} \left(\frac{T_1 - T_0}{T_0 T_1} \right)}.$$

In comparison with $T_0 = 300 \text{ K}$ and $T_1 = 2473 \text{ K}$, we get

$$(20) \quad \frac{k_1}{k_0} \doteq 8.2.$$

According to the first principle, the following potential form expresses the repulsive interaction between atoms [60]:

$$(21) \quad U(R) = -\frac{B}{r^{12}},$$

where B is an empirical parameter. Taking the effect of temperature on the reaction rate into consideration, we get the shrunken distance

$$(22) \quad 2r_2 \cong 2 \times 0.839r_1 = 2 \times 0.839 \times 0.645 \times r_0 = 0.0795 \text{ nm}.$$

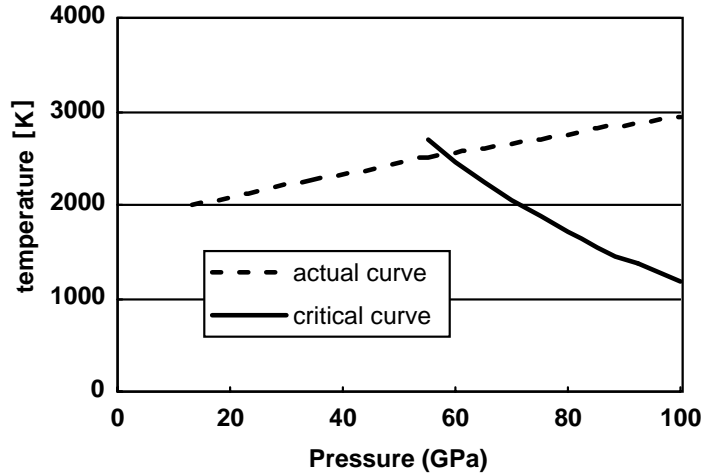


Fig. 3. – Critical temperature-pressure curve for nitrogen transmutation and actual temperature-pressure curve in the mantle.

This radius is still somewhat larger than the critical one (0.079 nm).

Conversely, we can calculate the critical temperature, corresponding to the critical distance for the dynamic reaction. From eqs. (8) and (9), we get $T_1 = 2689$ K. In other words, the nuclear transmutation between carbon and oxygen atoms would occur without help of electropionic attraction, when the temperature of the mantle is higher than 2689 K, that is, the mantle is deeper than 1300 km [58].

3. – Critical pressure-temperature curve for nuclear transmutation

Since the nitrogen transmutation is possible by combined effects of screening of free electrons and thermal activation, we last calculate the critical temperature as a function of pressure, using both screening and high-temperature effects. These results are shown in fig. 3, along with an actual temperature-pressure line in the mantle [61]. A form expresses the relation between the critical temperature T and the critical pressure P for the nuclear transmutation,

$$(23) \quad T = 7386 \times 10^{-0.008P} .$$

The intersection point of the two curves is 58 GPa and 2520 K, corresponding to a shrinkage of 35.4 and 22.4%, respectively. Thus the nuclear transmutation due to the two-body confinement of carbon and oxygen nuclei in the carbonate MgCO_3 lattice of the deeper mantle was explained by a combined effect (50% shrinkage) of screening by free electrons and the temperature enhancing on the reaction rate. This point corresponds to an upper portion of the lower mantle, being 2285 km below the surface in the present Earth [34]. Since temperature in the early Precambrian era was as high as 353 K, the critical curve in the Archean era would be shift to the left side. This means that the geological conditions of the Archean era for nitrogen transmutation would be more suitable than those of the present time.

4. – Comments on recent study

Magnesite is stable at pressures to 80 GPa and temperatures of 2000–2500 K [36, 62, 63], but stability of magnesite throughout the mantle remained unresolved. Recently, Isshiki *et al.* [64] reported that magnesite transforms to an unknown form at pressures above ~ 115 GPa and temperatures of 2100–2200 K (depth of ~ 2600 km) without any dissociation, in an *in situ* X-ray diffraction study, using the double-sided laser-heated DAC technique. However, when we note their X-ray diffraction profiles with increasing pressure at the maximum temperatures, magnesite peaks obtained at 100.1 GPa and 2200 K are lower and broader than those obtained at 60.6 GPa and 2000 K, and 84.3 GPa and 3000 K, although magnesite peaks increase with increasing pressure [65]. The lower and broader peaks at 100 GPa may suggest the possibility of a dynamic interaction of C and O atoms in the magnesite lattice. Unfortunately, they did not analyze nitrogen gases in experimental runs. Further work is needed, such as mass spectroscopic analysis under pressure and temperature conditions over 58 GPa and 2520 K.

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