### Journal of Physics Communications

### PAPER

OPEN ACCESS

CrossMark

RECEIVED 12 July 2020

REVISED 19 August 2020

ACCEPTED FOR PUBLICATION 26 August 2020

PUBLISHED 7 September 2020

Original content from this work may be used under the terms of the Creative Commons Attribution 4.0 licence.

Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI.



# Did nuclear transformations inside Earth form nitrogen, oxygen, and water?

### Mikio Fukuhara<sup>1,2</sup>

New Industry Creation Hatchery Centre, Tohoku University, Sendai 980-8579, Japan Waseda University Research Organization for Nano & Life Innovation, Green Device Laboratory, Tokyo 162-0041, Japan **E-mail: mikio.fukuhara.b2@tohoku.ac.jp** 

Keywords: nitrogen, oxygen, water, Earth, nuclear transmutation

### Abstract

Nitrogen and sea water concentrations have increased continuously from the Archean era. A rapid rise in oxygen concentration has been recorded from around 0.8 billion years ago. These phenomena cannot be completely explained by the collision of planetesimals with abundant nitrogen and the late veneer heavy bombardment of comets and meteorites with abundant water, and photosynthesis of heterotrophic plants, respectively. The formation of nitrogen, oxygen, and water are postulated to be the result of an endothermic nuclear transformation of carbon and oxygen nuclei confined in the carbonate aragonite lattice of Earth's mantle or crust at high temperatures and pressures. This process was likely influenced by excited electrons generated by stick sliding during the evolution of supercontinents, mantle convection triggered by collisions of major asteroids, and nuclear fusion in Earth's core. Thus, we show that calcium carbonates may have played a crucial role in the generation of atmosphere and sea water in Earth's history.

### 1. Introduction

One of the most important aspects of Earth's evolution is the formation of the atmosphere followed by continuously altering composition of the gases contained in it. The atmosphere is also affected by the amount and temperature of sea water. The atmospheric composition and amount of water (H<sub>2</sub>O) are of obvious importance given their necessity for life on Earth. However, the processes that alter the atmospheric composition changes of three important atmospheric gases, carbon dioxide (CO<sub>2</sub>), nitrogen (N<sub>2</sub>), and oxygen (O<sub>2</sub>), and the increase of the sea level in Earth's history from 4 billion years (Ga) ago to present time. The composition changes of atmospheric CO<sub>2</sub> [1–3], N<sub>2</sub> [4–6], and O<sub>2</sub> [7–9] through geologic time, and the increase in sea level from around 2.3 Ga to the present, are shown in figures 1(a) and (b). The data were sourced from nine [1–9] and four research groups [10–13], respectively.

A parabolic decrease in  $CO_2$  composition in the Archean era is accompanied by a gradual accumulation of  $N_2$  until 2 Ga [1–3]. The majority of  $CO_2$  removal can be explained by inorganic geochemical processes such as dissolution in a hot ocean environment and subsequent formation of carbonates as  $CO_2$  reservoirs in seas by virtue of the weathering of igneous rocks [14]. The  $N_2$  pressure increased parabolically until 2 Ga and converged to approximately 78% (the value seen presently), although the precise process leading to the change in  $N_2$  concentration is not well established yet. Given that these changes cannot be solely contributed to the collision of planetesimals with abundant nitrogen (given the evidence provided by the higher N/Kr ratio [15]), we must look to transformation within the Earth's crust and mantle for a more credible answer.

On the other hand, changes in  $O_2$  compositions can be divided into three time sections [7–9]: very low content before 2 Ga, rapid accumulation (popularly known as the 'Great Oxidation Event' (GOE) [9]) in the Precambrian era from 2 to 0.42 Ga, and saturation content from 0.42 Ga to the present time. The formation of the small amount of free  $O_2$  on the sea surface by ultraviolet photochemical reactions [16] in the Archean era can be expressed as



Figure 1. Changes in (a) concentrations of atmospheric  $CO_2$ ,  $N_2$ , and  $O_2$ , and (b) volumetric ratio of sea water from billions of years ago to the present time.

$$2H_2O + 4h\nu \rightarrow 4e - +4H^+ + O_2$$
 (1)

According to the present consensus [17], the generation is attributed to photosynthetic activity. After the first oxygenic photosynthesis by anaerobic cyanobacteria from 3.5 Ga onwards, aerobic photosynthesis by plants, algae, and cyanobacteria (eubacteria) produced  $O_2$  and carbohydrates from  $H_2O$  and  $CO_2$  after 2.7 Ga. The photosynthetic reactions can be represented by a single well-recognized formula [18]:

$$6CO_2 + 6H_2O + \text{light energy}(2, 870 \text{ kJ mol}^{-1}) \rightarrow C_6H_{12}O_6(\text{glucose}) + 6O_2.$$
 (2)

According to equation (2) 1 M of  $CO_2$  produces 1 M of  $O_2$ . In other words, this reaction does not vary in volume.

Studies on sea level changes have shown varied results. Kuenen [10] and Rubey [19] asserted a gradual accumulation, Conway [11] estimated a linear increase, whereas Twenhofel [12] and Walther [13] showed a parabolic rise from 0.5 Ga (figure 1(b)). Turekian [20] accepted the hypothesis of an initial 'watering'. However, Craig [21] conducted an isotope analysis of  $\delta^{28}$ O and  $\delta$ D values in natural waters from many parts of the world, and showed evidence of circular water activity below the sea surface, far from Earth's interior. This evidence excludes speculations of sea level variations by Conway [11], Twenhofel [12], and Walther [13]. Even if the level has remained constant after the Archean era, high amounts of water impose restrictions on phased bioactivity. As per the present consensus, sea water originated from the bombardment of a late veneer of asteroids with water. These asteroids were supposedly 'main belt' asteroids orbiting between Mars and Jupiter, and they delivered some amount of water to the primitive dry Earth [22]. However, while it sounds interesting, this mechanism is unlikely to be the dominant source of sea water [23].

As can be seen from figure 1(a), since the formation of  $O_2$  is likely related to the decrease in  $CO_2$  and increase in  $N_2$  levels, we consider two ratios in Earth's history:  $O_2/CO_2$  and  $O_2/N_2$ . Figures 2(a) and (b) present the timedependent ratios of  $O_2/CO_2$  and  $O_2/N_2$ , calculated using data from the nine research groups [1–9] listed in figure 1(a), and considering the main events in Earth's history. The time-dependent curves of  $O_2/N_2$  show systematic behaviour compared with the unstable ones of  $O_2/CO_2$ , suggesting that  $O_2$  formation was probably associated with the formation of  $N_2$ .



However, as figures 2(a) and (b) show, three questions regarding the generation of O<sub>2</sub> remain. First, how is it that a small amount of CO<sub>2</sub> in the atmosphere could have created a relatively large amount of O<sub>2</sub> via photosynthetic activity of heterotrophic plants for 2.7 billion years? According to equation (2), when all of the CO<sub>2</sub> (0.039 vol.%)[24] is consumed,  $2.37 \times 10^{15}$  kg (1.66  $\times 10^{15}$  m<sup>3</sup>) of O<sub>2</sub> should be generated. However, this amount corresponds to only 0.199 vol.% (=1.66  $\times 10^{15}/8.34 \times 10^{17}$  m<sup>3</sup>) of total O<sub>2</sub> present in the atmosphere today. Second, consider the strong dependency of the O<sub>2</sub>/N<sub>2</sub> ratio in figure 2(b) in comparison with the O<sub>2</sub>/CO<sub>2</sub> dependency in figure 2(a). The third question refers to the constant ratio of O<sub>2</sub>/N<sub>2</sub> in the atmosphere from 0.3 Ga to the present time. Since no directly observed geophysical data are available to explain the formation of these gases, these questions need to be analysed using circumstantial evidence and our understanding of Earth's atmosphere.

#### 2. How Earth's atmospheric pressure can affect gas concentrations

According to Newton's law of universal gravitation, every point mass of matter in the Universe attracts every other point mass with a force directly proportional to the product of the two masses and inversely proportional to the square of their separation. The atmospheric gases on Earth's surface play a role here; the gravitational force is directly proportional to the product of the masses of Earth and the atmospheric gases, and inversely proportional to the square of the distance between them [25]. The altitude of the troposphere, which contains roughly 80% of the mass of Earth's atmosphere, extends to 12 km (0.19% of Earth's radius [25]). We consider time-dependent gas pressure for three main gases, CO<sub>2</sub>, N<sub>2</sub> and O<sub>2</sub>, assuming a total pressure of 1 atm, including pressure of Ar. We use the revised data of Kasting [3] and Bertaux [5] to generate the curves for CO<sub>2</sub> and N<sub>2</sub>, respectively, and the data from NESTA [8] are used to create the O<sub>2</sub> curve. The integrated N<sub>2</sub> without escape curve is calculated using the following equation of the integrated N<sub>2</sub> curve between 3.6 and 1.2 Ga.

$$P_{\rm CO2} = 0.2775 \ln(|-t|) + 0.7215$$

where t denotes time in billion years. Note that  $R^2$  for the integrated N<sub>2</sub> without escape curve is 0.9953. Since the pressure and composition of the first atmosphere before late heavy bombardment in the Archean era are unclear,





we restrict our discussion to the formation period of  $N_2$  and  $O_2$  from 4 Ga to the present time. The calculated result is shown in figure 3(a). When the atmospheric pressure of  $N_2$  and  $O_2$  exceeds 1 atm, they escape to space from the upper atmosphere, aided by Earth's centrifugal force.

To investigate the formation of  $O_2$  by photosynthesis, the differential pressure of  $CO_2$  required for the formation of  $O_2$  is calculated from the integrated  $O_2$  pressure in figure 3(a). The result is presented in figure 3(b). The findings for the other groups appear in figures A1 and A2 in appendix A. According to equation (2), a considerable amount of  $CO_2$  is required for the formation of  $O_2$  in the time period from 0.7 to 0.2 Ga, which also corresponds to the appearance of animals on continents. Judging from the small amount of integrated  $CO_2$ , the rapid generation of  $O_2$  cannot be explained by photosynthetic activity alone. Thus, the consensus on the origin of the abundance of  $O_2$  on Earth appears to be misplaced.

We offer an alternative intuition, namely, the possibility of nuclear transmutation of  $N_2$ ,  $O_2$ , and  $H_2O$  inside the Earth.

## 3. Dynamic nuclear transmutation between C and O atoms in calcium carbonate in Earth's lower mantle

In previous papers [26, 27], the formation of  $N_2$  was interpreted to be the result of an endothermic nuclear transmutation of C and O atom pairs in the calcium carbonate aragonite lattice of Earth's lower mantle from the Archean era to the present time. Since it is expected that the formation of nitrogen is distinctively associated with the formation of the carbonaceous rocks, we considered the dynamic reaction that the carbon and oxygen atoms in carbonate crystals interact to form nitrogen. The processes came about as a result of physical catalytic help of excited electrons ( $e^*$ ) arising from plate tectonics and geoneutrinos ( $\nu_e$ ):

4

$${}^{12}\text{C} + {}^{16}\text{O} + 2e^* + 2\nu_e \rightarrow 2^{14}\text{N} + {}^{4}\text{He}$$
 (3)

When we reconsider transmutation reactions under high temperature and pressure, the following reaction comes to mind [26].

$$^{14}N + {}^{4}He \rightarrow {}^{16}O + {}^{2}D - 3.11 \text{ MeV}$$
 (4)

Deuteron fusions are generally expressed by the following reactions [28].

$$(D + {}^{2}D \rightarrow {}^{4}He + \gamma + 23.8 \text{ MeV}$$
 (5)

$$^{2}D + ^{2}D + ^{2}D \rightarrow ^{1}H + ^{4}He + n + 21.62 \text{ MeV}$$
 (6)

where  $\gamma$  and n are the photon and neutron, respectively.

From equations (4)–(6), we get

$${}^{12}C + {}^{16}O + 2e^* + 2\nu_e \to 2^{14}N + {}^{4}He + \gamma \to {}^{14}N + {}^{16}O + {}^{1}H + n - 5.29 \text{ MeV}$$
(7)

Thus equation (7) is an important reaction in the context of geoscience.

$$2^{12}C + 2^{16}O + 4e^* + 4\nu_e \to 2^{14}N_2 \uparrow + {}^{16}O_2 \uparrow + {}^{1}H_2{}^{16}O \uparrow + 2n - 10.58 \text{ MeV}.$$
(8)

This reaction is facilitated by the electropionic attraction related to excited electron capture and neutral pion catalysis [29]. Equation (8) distinctly indicates formation of gases with a N<sub>2</sub> to O<sub>2</sub> to H<sub>2</sub>O molar ratio of 2:1:1. The reaction shows endothermic nuclear transmutation, and the higher the increase in temperature and pressure, the faster the reaction rate (see appendix B). If O<sub>2</sub> in equation (8) reacts with hydrogen (H<sub>2</sub>) liberated from Earth's core, additional water is formed.

$${}^{16}\text{O}_2 + 2^1\text{H} \to {}^{1}\text{H}_2{}^{16}\text{O}.$$
 (9)

Thus, we ascribe the larger than explained concentrations of N<sub>2</sub>, O<sub>2</sub>, and H<sub>2</sub>O to possible endothermic nuclear transmutations of C and O nucleus pairs confined along three  $[1\sqrt{30}]$  directions in a (111) plane of rhombohedral calcium carbonate crystals existing in the Earth's mantle or crust [26, 27] (appendix A, figure A3).

### 4. O<sub>2</sub> generated from processes other than photosynthesis

If the integrated N<sub>2</sub> gas curve in figure 3(a) is reasonable, we can unconditionally calculate the generation amounts  $m_o$  of  $O_2$  in Earth's history, using amounts of N<sub>2</sub> ( $m_N$ ) from the integrated N<sub>2</sub> without escape curve over 1 atm, because N<sub>2</sub> is inert.  $m_o = m_N \times 0.5 \times 32/28$  from equation (8), and the result is presented in figure 4(a). The curve shows a logarithmic increase from 3.6 Ga to the present time. The calculated value of  $1.06 \times 10^{20}$  kg [24] is 387 times that of O<sub>2</sub> content in the present atmosphere (2.74  $\times 10^{17}$  kg). The extra O<sub>2</sub> could react with the large amount of H<sub>2</sub> liberated from Earth's core, according to equation (9), resulting in the formation of additional water.

### 5. Possible generation of H<sub>2</sub>O by nuclear transmutation

According to equation (8), we can calculate integrated water  $m_{w1}$  as  $m_N \times 0.5 \times 18/28$ . The additional water content referred to above is calculated using equation  $m_{w2} = (m_O - O_2) \times 18/16$ , where  $O_2$  is calculated oxygen in figure 4(a). The mass of each major gas component present in the dry atmosphere is as follows:  $3.9 \times 10^{18}$  kg  $(3.12 \times 10^{18} \text{ m}^3)$  for  $N_2$ ,  $1.2 \times 10^{18}$  kg  $(8.34 \times 10^{17} \text{ m}^3)$  for  $O_2$ , and  $2.6 \times 10^{15}$  kg  $(1.66 \times 10^{15} \text{ m}^3)$  for  $CO_2$ . Thus, as per Earth's gravity, the total mass of atmospheric gas at 1 atm is  $5.169 \times 10^{18}$  kg, including  $6.67 \times 10^6$  kg  $(3.72 \times 10^{16} \text{ m}^3)$  0.934 vol %) for Ar [24]. The pressure exerted by each gas is calculated using its weight. These results are shown in figure 4(b), along with the curves for  $m_{w1}$  and  $m_{w2}$ . The results clearly show an expected and continuous rise in seawater level from 3.6 Ga to the present time. However, the total weight of water at the present time is around  $3.9 \times 10^{20}$  kg, which constitutes 28.9% of the current amount of sea water,  $1.34 \times 10^{21}$  kg [30]. If Earth contains around  $10^{21}$  kg of  $N_2$ , the total amount matches with Kuenen's estimation [10], although he used no data for the period before 2.3 Ga. The formation of water from the Proterozoic era contributed to the explosion in growth of cyanobacteria and vegetation.

#### 6. Geological conditions for generation of N<sub>2</sub>, O<sub>2</sub>, and water

Considering the C–O distance (~0.079 nm [31]) required for the dynamic nuclear transmutation of calcium carbonates in the lower mantle, higher temperature and pressure are necessary conditions for these reactions [26, 27]. Since the relationship between critical temperature *T* and critical pressure *P* for the nuclear





transmutation is expressed as 7,253  $\times$  e<sup>-0.014P(31)</sup>, the formation of N<sub>2</sub>, O<sub>2</sub>, and H<sub>2</sub>O would be possible at temperatures  $\geq$ 2,510 K and pressures  $\geq$ 58 GPa. The region with this temperature–pressure profile corresponds to a zone known for diamond formation in an upper portion of Earth's lower mantle (see figure 4(S) in appendix B). Carbonaceous rocks are recognized to have been predominantly formed by weathering on land and inorganic reactions in the primitive sea during the Archean era, and by biological reactions in the Coral Sea during the Cambrian era. They are distributed on and near Earth's surface [32], and the large quantities of fresh calcium carbonate rocks are thought to have been delivered to the upper mantle by plate tectonics [33].

Next, we consider another attraction mechanism responsible for accelerating the confinement of C and O nuclei in the calcite lattice. We refer to physical catalysis by a neutral pion, which is recognized as a nonexchangeable component in a strong nuclear field [29] (see appendix B). The excited electrons and neutrinos in equation (3) are thought to have been generated by stick sliding [34, 35] due to plate tectonics and the sliding of carbonate crystals, and are trapped in the interior portions of Earth, respectively. Neutrinos are known to be generated in the Universe by the Sun [36] or the flares of t-tauri stars [37] in the Archean era, and they are also generated by the collision of free electrons derived from pressure inonization, namely, the deuteron-thermonuclear fusion in Earth's inner core [38–40]. Furthermore, the excited electrons may be also generated from existence of heavy holes which have rapid, small amplitude oscillations [41] in the lower temperature crust, rather than higher-temperature mantle. Therefore, we note that there is a very real possibility that nuclear transformation combined with physical catalysis may have occurred in Earth's crust and crust.

### 7. Asteroid collision-induced atmospheric evolution and liberation of atmospheric gases to space

The rise in the concentrations of  $N_2$ ,  $O_2$ , and  $H_2O$  in figures 2(b), 3(a), and 4, respectively, indicate the possibility that, from 3.5 Ga onwards, the liberation of  $O_2$  into the atmosphere from 1.7 Ga onwards and the rapid increase in its levels in 0.5 Ga were caused by the collisions of asteroids in 3.4 Ga [42], the Vredefort collision in South Africa in 2 Ga [43], and the Sudbury collision in Canada in 1.85 Ga [44], and the Bedout collision in 0.3 Ga [45].

The extraterrestrial impacts aided the growth of the continents and intrusion of granitic magmas, and shifted mantle conversion patterns [46–48].

Inert N<sub>2</sub> and active  $O_2$  was discharged into the atmosphere by volcanic activities and mantle plumes up to 1.2 and 0.5 Ga. Thereafter, the gases started escaping to space from 1.3 and 0.3 Ga, respectively. Indeed, recent research has reported that large amounts of terrestrial N<sub>2</sub> [49, 50], noble gases [50], and  $O_2$  [51] have been transported as far as the Moon by the Earth's wind. This is because atmospheric mass gets constantly dispersed into space, and without a continuous resupply of these gasses from Earth' interior portions, our atmosphere could not keep the pressure needed to support life on Earth. If N<sub>2</sub>, O<sub>2</sub>, and H<sub>2</sub>O were derived from C and O<sub>2</sub>, geologically active planets with abundant C and O<sub>2</sub> atoms could have generated these compounds. This is a very interesting possibility, since C and N<sub>2</sub> are two elements essential for all life forms [52].

### Acknowledgments

We would like to thank Editage (www.editage.jp) for English language editing.

### **Competing financial interests**

The author declares no competing financial interests. Correspondence and requests for materials should be addressed to MF (fukuhara@niche.tohoku.ac.jp).

### Appendix

### Appendix A. Verification for formation effect of oxygen by photosynthesis

In photosynthetic reaction of equation (2), the consumption of one molar carbon dioxide produces equivalent molar oxygen. To investigate formation effect of oxygen by photosynthetic reaction, the differential pressures of CO<sub>2</sub> required for formation of O<sub>2</sub> are calculated from integrated O<sub>2</sub> curves by Kasting [7] and Holland [9], using an equation of reduced pressure  $P_{CO2} = P_{O2} \times 1.977/1.429$ , where 1.977 kg m<sup>-3</sup> [53] and 1.429 kg m<sup>-3</sup> [54] are density of CO<sub>2</sub> and O<sub>2</sub> gases, respectively. These data are shown in figures A1 and A2, respectively.

Both amounts of  $CO_2$  required for formation of  $O_2$  in time period from 1.9 Ga onwards in Kasting [7] and 0.9 Ga onwards in Holland [9] are larger than those of integrated  $CO_2$ .  $CO_2$  is reversely in excess at 0.2 Ga and present time. Thus the rapid generation of oxygen cannot be explained by photosynthetic activity only.







**Table A1.** The densities of CaCO<sub>3</sub>, C–O bond distances, depths from surface, temperature and  $k_1/k_2$  at pressure of 50, 60, 70, 80, 90 and 100 GPa.

Pressure (GPa)	50	60	70	80	90	100
Density (Mg m <sup>-3</sup> )	8.38	9.51	10.64	11.77	12.91	14.04
C–O bond distance (nm)	0.1009	0.0968	0.0932	0.0900	0.0873	0.0850
Depth (km)	1,100	1,340	1.570	1,800	2,030	2,270
Temperature (K)	2,778	2,443	2,109	1,774	1,439	1,105
$k_1/k_2$	3.68	4.80	5.91	7.03	8.14	9.26





### Appendix B. Previous works for nuclear transformation

In previous papers [17, 26], from wondering fact that the main 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 [55], the origin of nitrogen is interpreted to be the result of the endothermic nuclear transmutation:

$$^{12}C + {}^{16}O \rightarrow 2^{14}N \quad Q = -10.47 \text{ MeV}$$
 (A1)

This reaction is due to two-body confinement of carbon and oxygen nuclei in CaCO<sub>3</sub> carbonate lattice of the mantle, which is mediated by nuclear attraction caused by the catalysis of neutral pions,  $\pi^{\circ}$ , and taking into consideration the energy-momentum equilibrium [29]:

$${}^{12}C + {}^{16}O - 2\pi^{\circ} = 2^{14}N, \tag{A2}$$

which is based on an assumption that a parabolic increase in nitrogen content correlated to an abrupt decrease in carbon dioxide during the Archean era [1–3]. Pions are responsible for all low- energy nuclear interactions [56]; the pions within the nucleus allow the nucleonic species to bind together and transmute with each other [31]. Kenny [57] has termed equation (A2) as the 'electropionic reaction'. The neutral pion mass balance in equation (A2) is provided by emission of two excited electrons [58], which are derived from the carbonate lattice:

$$e \to e^*, \gamma,$$
 (A3)

$$\gamma + \gamma = \pi^{\mathbf{o}},\tag{A4}$$

where  $\gamma$  is photon.

On the other hand, the electron reacts with deuteron to form neutral pion and an electron neutrino [29, 59]:

$$e - +^2 D \rightarrow \pi^o + \nu_e + \gamma$$
 (A5)

Fukuhara [59] has reported the following formula for the formation of helium:

$$^{2}D + ^{2}D + 4 e^{*} = ^{4}He$$
 (A6)

From equations (A3)–(A6), we actually got the following weak dynamic interaction:

$${}^{12}\text{C} + {}^{16}\text{O} + 2e^* + 2\nu_e \to 2^{14}\text{N} + {}^{4}\text{He.}$$
 (A7)

This reaction is facilitated by the electropionic attraction that is related to excited electron capture and neutral pion catalysis [29]. Since equation (A7) is a non-equilibrium (irreversible) equation, it does not obey the rules of parity and momentum balance. In the irreversible endothermic reaction proposed by Glansdorff and Prigogine [60], remarkable enhancement is expected on the basis of the thermal factor of  $\exp(-\Delta G/kT)$ , where  $\Delta G$  is the change of Gibbs energy for the whole system.

### Appendix C. Configuration of calcium carbonate required for endothermic nuclear transformation

Oxides, carbonates, sulfides were representative of the rocks in the early Earth. Among these rocks, the carbonaceous rocks are clearly  $CO_2$ -reservoirs. The main parts of carbonaceous rocks are distributed on and near the Earth's surface as dolomite  $CaCO_3 \cdot MgCO_3$ , and the remainder of rocks sinks into the deep mantles by plate tectonics. We select calcite  $CaCO_3$  as a candidate material available for the nuclear transformation in this study. Figure A3 is configuration of  $CO_3$  group atoms in (111) calcite crystal planes.

The scheme for nuclear transmutation of  $CaCO_3$  resembles that observed for the Pd/complexes, which are composed of Pd and CaO thin films, and the Pd substrate, after the Pd complexes are subjected to  $D_2$  gas permeation [61]. Although they have not described directly the transmutation effect brought about by the Ca element, they could not obtain a similar result using MgO in place of CaO [62]. Therefore, we believe that Ca is the common driving element for nuclear transmutation. In particular, the possibility exists that the electrons are derived from calcium elements with many electrons, such as  $3s^2$ ,  $3p^6$  and  $4s^2$  [63].

### Appendix D. Nuclear transformation in aragonite phase at high pressure and high temperature

Since we can estimate that the calcite form, which is thermodynamically stable under pressure below 100 GPa and temperature between 3,000 and 1,000 K, is orthorhombic aragonite from *P*-*T* phase diagram reported by Suito *et al* [64], we used an extrapolated formula  $\rho = 0.113 P + 2.71$  for high pressure density  $\rho$  of aragonite, due to the lack of crystal lattice data for aragonite subjected to pressure P > 7.7 GPa.

The densities of CaCO<sub>3</sub>, C–O bond distances, depths from surface and temperatures at pressure of 50, 60, 70, 80, 90 and 100 GPa can be estimated. From Thomas-Fermi (TF) approximation to the screening effect of the CaCO<sub>3</sub> lattice below the surface of Earth, we can assume the corresponding C–O bond distance  $2r_1$  [65]. The temperatures are estimated from [66]. These are presented at table A1, along with  $k_1/k_2$ .

The reaction rate *k* can be expressed by Arrhenius equation:

$$k = (k_b f_N^2 / h f_c f_O) e^{-E/RT}$$
(A8)

where the  $f_C$ ,  $f_O$ , and  $f_N$  are the partition functions of <sup>12</sup>C, <sup>16</sup>O, and <sup>14</sup>N, respectively, and  $k_b$ , R, and E are the Boltzman constant, gas constant, and activation energy of the reaction, respectively, we can obtain the effect of temperature on the reaction rate k. When  $f_C = f_O = f_N$ , a ratio of the rates at temperature  $T_0$  and  $T_1$  can be described as follows:

$$\frac{k_1}{k_2} = \frac{T_1}{T_2} e^{\frac{E}{R} \left( \frac{(T_1 - T_o)}{T_0 T_1} \right)}$$
(A9)

In comparison with  $T_0 = 300$  K and  $T_1 = 1,105, 1,439, 1,774, 2,109, 2,443$  and 2,778 K, we get  $k_1/k_2$  in table A1. From the effect of temperature on the reaction rate *k* (equation (A8)) under the repulsive interaction potential between the atoms, we get the following example of shrunken distance at 80 GPa:

$$2r_2 \cong 2 \times 0.682r_1 = 2 \times 0.862 \times 0.86128 \times r_o = 0.1554 \text{ nm}$$
(A10)

This radius (0.078 nm) is somewhat smaller than the critical radius (0.079 nm [31]) required for a dynamic nuclear reaction. Thus, the relationship between the critical temperature T and the critical pressure P for the reaction is expressed as

$$T = -33.47 \text{ P} + 4,\,451.5 \tag{A11}$$

Since the region with this temperature–pressure profile corresponds to the zone of diamond formation in mantle [67], the relationship in equation (A11) is shown in figure A4, along with the diamond formation lines, the actual temperature-pressure profile (T = 10.95 P + 1,875) of the Earth [68], and estimated *P*-*T* phase boundary line between aragonite and disordered calcite [64]. The critical temperature and pressure, which correspond to the critical radius for the dynamic reaction, are 2,510 K and 58 GPa, respectively, leading to depth of 1,290 km [69]. Therefore, it is possible that the dynamic reaction occurs in an upper portion of the lower mantle. Even if the convection current in the upper mantle is viscous fluid, the nuclear reaction could be promoted. In fact, fusion reaction <sup>6</sup>Li (d,  $\alpha$ ) <sup>4</sup>He and <sup>2</sup>H(d, p) <sup>3</sup>H with ~6.83 × 10<sup>6</sup> K were measured in liquid Li acoustic (ultrasonic) cavitation [70]. The conditions required for the formation of nitrogen, oxygen and water are more stringent than those for diamonds.

Here, we must describe the natural conditions for diamond formation. It is well known that natural diamonds crystallize directly from kimberlite rock melts that are rich in calcite and found at depths of 1,500 km or more inside the Earth. The melts are essentially saturated in carbon dioxide gas at high pressures over 30 GPa

and temperatures  $\geq$  763 K in deep in the Earth's mantle, since the <sup>12</sup>C/<sup>13</sup>C ratio of a diamond shows that it arises from carbon dioxide [71]. Furthermore we note that platesets of poly-cyanogen (C<sub>3</sub>N) are dispersed in kimberlite rocks [72]. The type Ia diamond contains maximally 2%-mil nitro-gen [73]. Thus, the nitrogen impurities in diamonds may be the result of nuclear transmutation of CO<sub>2</sub> in the melt. Furthermore, it is known that nitrogen is distributed extensively throughout the silicate phase of the Earth's crust and mantle [74].

#### **ORCID** iDs

Mikio Fukuhara https://orcid.org/0000-0002-6139-7596

#### References

- [1] Owen T, Cess R D and Ramanathan V 1979 Nature 277 640
- [2] Ohmoto H 1994 Kagaku (Science) 14 360
- [3] Kasting J F 1987 Precamb. Res. 34 205
- [4] Budyko M I, Ronov A B and Yanshin A L 1985 *History of the Earth's Atmosphere* 16128 (Berlin, F.R. Germany: Springer) p 2
- [5] Bertaux J-L 1983 Le Grand Atlas de L'Astronomie. ed J Andouze and G Israel 63Encyclopedia Universalis (Obunsha, Tokyo: Japanese language ed.)
- [6] Bimblecombe P and Davies T D 1981 The Cambridge Encyclopedia of Earth Sciences 276(New York: Cambridge University Press)
- [7] Kasting J F 1993 Science 259 920
- [8] Manning CLM 2012 The slow build up of Oxygen in the Earth's Atmosphere (Kentucky: Kentucky Foundation) http:// windows2universe.org/earth/past/oxygen\_buildup.html
- [9] Holland H D 2006 Phil. Trans. R. Soc. B 361 903
- [10] Kuenen PH 1933 Geology of Coral Reefs. The Snellius Expedition 5, Geological Results, Pt. A (Utrecht: Keminken Zoon) 126
- [11] Conway E J 1943 Roy. Irish Acad., Pr., XLVIII, Sect. B 9 161
- [12] Twenhofel W H 1929 Geol. Soc. Am. Bull. 40 385
- [13] Walther J 1911 Am. J. Sci. 31 55
- [14] Hamilton PJ, O'Nions RK, Evensen NM, Bridgwater D and Allaart JH 1978 Nature 272 41
- [15] Pepin R O 1989 Atmospheric compositions: key similarities and differences Origin and Evolution of Planetary and Satellite Atmospheres Atreya S K et al (Tucson: The University of Arizona Press) 291–305
- [16] Bates D R and Nicolent M 1950 J. Geophys. Res. 55 301
- [17] Dutkiewicz AV, George SC, Ridley J, Vlk H and Buick R 2016 Geology 36 437
- [18] Whitmarsh J et al 1999 The photosynthetic process Concepts in Photobiology: Photosynthesis and Photomorphogenesis ed G S Singhal et al (Boston: Kluwer Academic) 11–51
- [19] Rubey W W 1951 Bull. Geo. Soc. Am. 62 1111
- [20] Turekian K K 2001 Origin of the Oceans (New Haven: Academic Press, Yale University) 2055-8
- [21] Craig H 1963 The isotopic geochemistry of water and carbon in geothermal areas Nuclear Geology on Geothermal Areas, Spoleto ed T E Pisa (Consiglio Nazionale Delle Ricerche Lab. di Geol. Nucleare) 17–53
- [22] Altwegg K et al 2014 Science 347 1261952
- [23] Jewitt D and Young E D 2015 Sci. Am. 312 36
- [24] Koeneman R and Kohnke J M 2020 The Composition of the Earth's Atmosphere (Kaiser Science) https://kaiserscience.wordpress.com/ earth-science/weather/atmosphere
- [25] Serway R A 1990 Physics for Scientists & Engineers with Modern Physics 3rd edn 151 (Philadelphia: Saunders College Publishing) 358
- [26] Fukuhara M 2004 Nuovo Chimento 27C 99
- [27] Fukuhara M 2014 J. Mod. Phys. 5 75
- [28] Allis W P 1960 Nuclear Fusion (Princeton: D. Van Nostrand Company) 1
- [29] Fukuhara M 2003 Fus. Sci. and Technol. 43 128
- [30] Shiklomanov I A 1993 Water in Crisis. ed P H Gleick (New York: Oxford University Press) 13
- [31] Jones S E et al 1989 Nature 338 737
- [32] Allègre C J and Schneider S H 1994 Sci. Am. 271 44
- [33] Bercovici D 2011 Mantle conversion Encyclopedia of Solid Earth, Geophysics ed H Gupta (Netherland: Springer) 1–27
- [34] Brune J N, Brown S and Johnson P A 1993 Tectonophysics 218 59
- [35] Tsutsumi A and Shirai N 2008 Tectonophysics 450 79
- [36] McFadden L-A and Johnson T V 1999 Encyclopedia of the Solar System. ed P R Weissman, L-A McFadden and T V Johnson (San Diego: Academic) 68
- [37] Unsold A 1974 Der Neue Kosmos 2nd ed. (Berlin: Springer) 226
- [38] Fukuhara M 2016 Sci. Rep. 6 37740
- [39] Fukuhara M 2017 Sci. Rep. 7 46436
- [40] Fukuhara M 2020 AIP Advances 10 035126
- [41] Jiang Z F, Li R D, Zhang S-C and Liu W M 2005 Phys. Rev. B 72 045201
- [42] Gough E 2016 30 km Wide Asteroid Impacted Australia 3.4 Billion Years Ago (Universe Today) http://universetoday.com/128993/
- [43] King H Vredefort Impact Crater (Geoscience News and Information) http://geology.com/articles/vredefort-dome.shtml
- [44] Prigg M 2014 The mystery of the Sudbury Basin Solved: The Second Largest Crater on Earth was Caused by a Massive COMET Hitting the Planet 1.8 billion Years Ago, Mail Online, http://dailymail.co.uk/sciencetech/article-2839876/.
- [45] Honda S, Yuen D A, Balachandar S and Reuteler D 1993 Science 259 1308
- [46] Grieve R A F 1980 Precam. Res. 10 217
- [47] Taylor R S and McLennan S M 1996 Sci. Am. 274 60
- [48] Simpson S 2010 Sci. Am. 302 60
- [49] Wieler R, Humbert F and Marty B 1999 Earth Plan. Sci. Lett. 167 47
- [50] Ozima M, Seki K, Terada N, Miura Y N, Podsek F A and Shinagawa H 2005 Nature 436 655

- [51] Terada K, Yokota S, Saito Y, Kitamura N, Asamura K and Nishino M N 2017 Nat. Astro. 1 41550
- [52] Nealson K 2006 (Los Angeles: University of South California) Private communication
- [53] Pierantozzi R 2003 Carbon dioxide Kirk-Othmer Encyclopedia of Chemical Technology (https://doi.org/10.1002/ 0471238961.0301180216090518.a01.pub2)
- [54] Baxter G P and Starkweather H W 1924 Proc. Natl. Acad. Sci. U.S.A. 10 479
- [55] Bohr A and Mottelson B R 1969 Nuclear structure Single-Particle Motion vol 1 (Amsterdam: W. A. Benjamin) 168
- [56] Measday D F and Miller G A 1979 Ann. Rev. Nucl. Part. Sci. 29 121
- [57] Kenny J P 1991 Fusion Technology 19 547
- [58] Feymann R P 1961 The Theory of Fundamental Processes (San Francisco: Benjamin/Cummings Publishing Company) 37
- [59] Fukuhara M 2005 Possible coexistence of electron and electron neutrino in nucleus and its effect on D-D cold fusion into helium Proc. of 6th Meet. of Jap. CF Res. Soc. (Tokyo Inst. Technology) (Tokyo, 27–28 April) pp 53–7
- [60] Glansdorff P and Prigogine I 1971 Thermodynamic Theory of Structure, Stability and Fluctuations (London: Wiley Interscience, London) 1
- [61] Iwamura Y, Sakano M and Itoh T 2002 Jap. J. App. Phys. 41 4642
- [62] Iwamura Y 2004 Mitsubishi Heavy Industries, private communication
- [63] Fukuhara M 2007 Jap. J. Appl. Phys. 46 3035
- [64] Suito K et al 2001 Am. Mineral. 86 997
- [65] Kennard O 1968 International Tables for X-ray Crystallography, International Union of Crystallography (Birmingham: Kynoch Press)) 276
- [66] Glasstone S and Lewis D 1960 Elements of Physical Chemistry (Princeton: D. Van Nostrand Company) 632
- [67] Bundy F P 1985 Solid State Physics under Pressure: Recent Advance with Devices (Dordrecht: Scientific Publishing Company)) 1
- [68] Ohtani E and Irifune T 1994 Basic Measurement and Application of Solid Planet Material Science ed H Takeda, H Kitamura and M Miyamoto (Tokyo: Science House) 187
- [69] Warneck P 1998 Chemistry of the Natural Atmosphere 2nd edn (New York: Academic) 605
- [70] Toriabe Y, Yoshida E, Kasagi J and Fukuhara M 2012 Phys. Rev. C 85 054620
- [71] Craig H 1953 Geochim. Cosmochim. Acta 3 53
- [72] Frondel C and Kennedy G C 1977 Encyclopedia of the Geological Sciences ed D N Lapedes (New York: McGraw-Hill) 155
- [73] Satoh S, Yazu S, Tsuji K, Hara A, Urakawa N and Yoshida A 1986 Sumitomo Electric Technical Review 129 99
- [74] Brandes J A, Boctor N Z, Cody G D, Cooper B A, Hazen R M and Yoder H S Jr 1998 Nature 395 365