

**THE ROLE OF ISOTOPIC EXCHANGE REACTION IN OXYGEN ISOTOPE EVOLUTION IN THE PROTOSOLAR DISK.** H. Nagahara<sup>1</sup> and K. Ozawa<sup>2</sup>, <sup>1</sup>Dept. Earth Planet Sci., The Univ. Tokyo (7-3-1 Hongo, Tokyo 113-0033, Japan and hiroko@eps.s.u-tokyo.ac.jp), <sup>2</sup> Dept. Earth Planet Sci., The Univ. Tokyo.

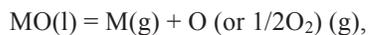
**Introduction:** Calcium-aluminum-rich inclusions (CAIs) and chondrules show wide variability in chemical composition and texture, suggesting notable chemical fractionation before and/or during their formation. CAIs often show evidence for evaporation and/or condensation, whereas very limited fraction of them shows notable mass-dependent isotopic fractionation. An exception is FUN inclusions, which show significant mass dependent oxygen isotopic fractionation with associated Mg and other elemental isotopic fractionation. The inclusions are thought to have suffered evaporation during melting.

It is important to investigate the relationship between chemical fractionation and isotopic fractionation through evaporation and re-condensation during CAI and chondrule formation in order to understand the physical and chemical conditions that resulted in the very little mass-dependent isotopic fractionation. Ozawa and Nagahara [1] explored a model for a silicate melt sphere that was heated instantaneously to an above solidus temperature and then cooled in a closed system. They showed that elemental and isotopic fractionations during evaporation in vacuum are coupled when evaporation Péclet number, the ratio of time scales of diffusion in melt (or solid) and evaporation, is small than ~100, but are decoupled when larger.

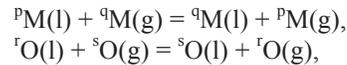
There is a crucial problem that has not been quantitatively investigated yet, which is the role of isotopic exchange reactions. It is easily expected that the effect of isotopic exchange would be larger for oxygen compared to metallic elements, because oxygen is more abundant in the ambient gas than metallic elements.

In this paper we quantitatively examine the role of exchange reactions in mass-dependent oxygen isotope fractionation during evaporation and recondensation of silicate melts in gas with isotopically identical and distinct gases. The results are applied to the CCAM line, mass-dependent oxygen isotopic fractionation of FUN inclusions, and oxygen isotopic variations of Allende chondrules.

**Model:** Evaporation and condensation of metallic element are assumed to take place through a reaction



where M and O represent a metallic element and oxygen, respectively. This equation is appropriate to examine elemental fractionation between melt and gas. Isotope fractionation is affected by following exchange reactions,



where <sup>p</sup>M implies p isotope of the element M. Isotope exchange can take place between specific gas species (e.g. O) and corresponding elements exposing on the surface of the condensed phase (oxygen bonding to Si or Mg). The evaporation and condensation flux of elements and isotopes are described with the Hertz-Knudsen equation, which is extended to isotope exchange reactions.

We examine the role of oxygen isotopic exchange for silicate melt and ambient gas during evaporation and recondensation in a closed system that monotonically cools from high temperature. Silicate dust aggregates are assumed to be instantaneously heated to an above liquidus temperature. There are three free parameters; one is the efficiency of the isotope exchange, the second is the cooling rate of the system, and the third is the amount of initial oxygen in the ambient gas. The efficiency of isotope exchange is critical to evaluate the role of isotope exchange reaction, and we do not know the plausible value. The cooling rate should be evaluated because of the time-dependent nature of all the processes including evaporation, condensation, isotope fractionation, and isotope exchange. Variability in the amount of initial oxygen in gas should be examined, because the oxygen abundance in the ambient gas can be variable in the proto-solar disk due to the existence of ice in addition to silicate dusts.

A silicate melt sphere is assumed to be homogeneous all through the process, and the role of diffusion and crystallization in the sphere are neglected. The system consists of Na-Mg-Al-Si-K-Ca-Ti-Fe-O. Hydrogen is not considered and thus H<sub>2</sub>O is ignored as an oxygen-bearing gas species. Ignorance of H<sub>2</sub>O does not alter the essence of the results, although it affects isotopic fractionation as a function of hydrogen pressure and evaporation/condensation rates. Therefore, discussions are valid as far as we do not discuss the real time scale of the processes.

The model is further applied to evaporation and recondensation processes in isotopically distinct O-bearing gas, where the oxygen isotopic composition of dusts is assumed to be δ<sup>17</sup>O=δ<sup>18</sup>O=-50‰ and that of the initial gas δ<sup>17</sup>O=5‰ and δ<sup>18</sup>O=10‰, which assumes dusts and proto-solar disk gas in conventional model.

The parameters, temperature, size, time, and evaporation/condensation rates are non-dimensionalized.

**Results and Discussions:** Calculations with the kinetic evaporation/condensation model with various values of the three parameters showed the following important results on oxygen isotopic compositions of CAIs and chondrules heated in a protosolar disk.

Evolution of oxygen isotopic fractionation is decoupled with chemical fractionation of silicate melt; oxygen isotopic fractionation goes more rapidly than chemical fractionation, of which timing depends on the isotopic exchange efficiency.

Mass-dependent oxygen isotopic fractionation of silicate melt is suppressed by isotopic exchange during evaporation/recondensation. If isotopic exchange does not work, oxygen isotopic composition of silicate melt spheres after evaporation/recondensation becomes lighter than the initial one if the silicate and gas have identical oxygen isotopic composition due to more abundance of  $^{16}\text{O}$  in the gas. Isotopic exchange homogenizes the compositions effectively to result in the same composition between silicate and gas.

Cooling rate of the system affects the evolution time of oxygen isotopic mass fractionation, but the final composition is not affected. In a system with initially different oxygen isotopic compositions between silicate melt and gas, deviation from a straight mixing line to the  $\delta^{18}\text{O}$ -rich side on a three-oxygen isotope plot is inevitable through evaporation/recondensation and isotope exchange.

Abundance of oxygen in the initial gas largely affects the degree of deviation from a simple mixing line during the cooling process (Fig. 1). In a system with different initial oxygen isotopic compositions between melt and gas, the final oxygen isotope composition of melt is also largely affected by the initial oxygen abundance in gas (Fig. 1). The average oxygen abundance in gas of proto-solar disk relative to that in silicate ( $\sim 10$ ), silicate melt gets oxygen isotopic composition very close to the gas.

Isotopic exchange efficiency plays an important role on evolution and final oxygen isotopic composition of silicate melt heated in gas (Fig. 2). Effective isotopic exchange largely suppresses mass dependent isotopic fractionation, which tends to result in forming a straight “mixing line”, whereas, smaller efficiency causes larger degree of mass-dependent isotopic fractionation, and deviation from a straight “mixing line” becomes larger.

A straight mixing line such as CCAM line requires the presence of oxygen in the ambient gas more than the average proto-solar disk.

The mass-dependent oxygen isotope fractionation of FUN inclusions by  $\sim 40\%$  or more is not achieved by evaporation in a closed system, and almost vacuum condition is required, which was possible by almost complete continuous separation of gas and sili-

cate melt during evaporation. If FUN inclusions were formed from oxygen isotopically light solid, they have suffered a reaction between isotopically heavy gas after vacuum evaporation, where the gas abundance must have been more than 10 times of the average proto-solar gas.

Oxygen isotope composition of Allende chondrules was well reproduced by the present model in the condition of rapid cooling and in gas with oxygen abundance twice to five times of the average proto-solar gas.

In summary, a series of oxygen isotopic composition of CAIs and chondrules reflects the ambient physical conditions such as dust/gas ratio and dust/gas separation of a protosolar disk.

**References:** [1] Ozawa, K. and Nagahara, H. (2001) *GCA*, 65, 2171–2199.

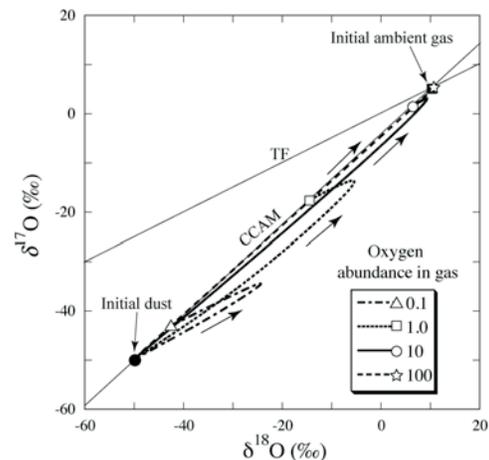


Fig. 1 Effect of abundance of oxygen in gas on evaporating and condensing silicate melt that monotonically cools. The curves show trajectories of isotopic composition of a melt sphere. The final compositions are shown by open symbols.

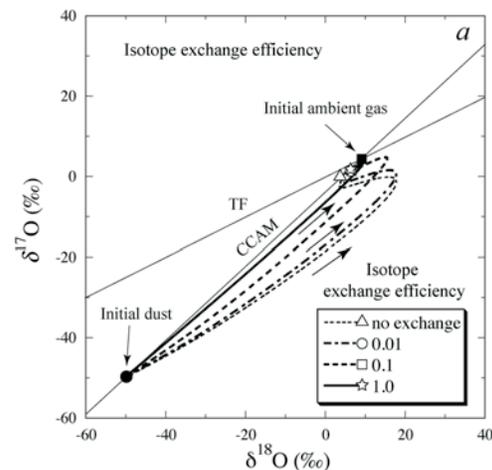


Fig. 2 Effect of isotopic exchange efficiency on trajectory of oxygen isotopic composition of silicate melt cooling in a gas with isotopically distinct composition.