

UNICON – A Universal Condensation model for cosmochemical and planetary-science study

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Abstract

A universal condensation model (UNICON) is designed to study condensation in the early solar system and exoplanets. It is the first open-source condensation program. The model is based on the chemical equilibrium calculation. It provides a robust and easy-to-use framework: users can freely change any parameters and thermodynamic data; users can acquire the phase assemblage of a certain P-T condition by just one command. The model has robust phase transition criteria to guarantee the system would converge to the requested solution. Also, this model has the function of isolating and adding material. This paper shows some examples how this model can solve the cosmochemistry/planetary science problems.

Introduction

Condensation model is a power tool to study the planetary science. The formation of Ca, Al-rich inclusion (CAI), chondrule, and pre-solar grains all request proper condensation models to account for. It can also be used to study the chemistry of protoplanetary disk, which would indicate the chemistry of exoplanets (Carter-Bond et al, 2011). The first condensation was developed by Grossman (1972) to solve the condensation of early solar system, later improved by Yoneda & Grossman (1995). The improved version updated some old thermodynamic data and implemented CMAS (Ca-Mg-Al-Si) melts in the model. Ebel & Grossman (2001) applied this model to study pre-solar grains properly condensed at the supernova ejection. Ebel (2006) further improved the condensation model by updating thermodynamic data, especially for liquid solution. He used the new model to study the condensation of CAI, chondrule, and pre-solar grains. There are also other versions of condensation models: Lodders & Fegley (1993) firstly developed a condensation model CONDOR to study the trace element --- the REE pattern. Later, Lodders (2003) used the CONDOR to study the condensation of all elements, though she did not consider the solid solution; Petaev & Wood (1998) developed a CWPI model, which would isolate a fraction of condensate at each step. They used this model to solve the puzzling condensation sequence between anorthite and olivine in the CAI. All of the above models were written in the FORTRAN programming language and they have not been shared to the public. Therefore, it would be helpful for the public if there is an open-source condensation model that can be applied to all the scenarios. Since different scenarios would require different phase assemblage and thermodynamic data, the model should be designed to modify the thermodynamic data easily. Also, the model should have all the functions that may be used in the modeling, like freely changing P-T condition, isolation, and adding material during the modeling. To guarantee the model to work in different scenarios, a robust criterion for phase transition should be developed. I have developed a condensation model that meets all the requirements. This paper demonstrates how this model works and shows some example that

are solved by my model. This model will be released soon in the Github and people can download and use it.

The principle of equilibrium calculation and modeling

This model adapts the chemical equilibrium as the core engine for condensation. Using the same mathematic principle as Grossman (1972), the model intends to find the species assemblage that would minimize the Gibbs Free Energy of the whole system. The main assumption here is that the scenario to be modelled would eventually achieve equilibrium. This assumption works well when the temperature is high enough, like at the molecular cloud closed to the protosun, stellar interior, and the supernova ejecta. One caveat is that high temperature does not guarantee the equilibrium, for instance, at the scenario where the temperature changes rapidly. Therefore, before using this model, we should confirm the modelled scenario would reach the equilibrium.

Here I will describe the mechanism of the UNICON model. First, the element involving the modelling should be set up. My model allows users to freely choose and add elements. In my prototype, I only include the H, C, O, S, Mg, Al, Si, Ca, and Ti, though more elements could be added. When including an element, its (nominal) total pressure is also set, which defined as:

$$P_{tot} = n_{tot}RT \quad (1)$$

where P_{tot} , n_{tot} , R , T are total pressure, total number density, ideal gas constant, and temperature, respectively. It should be noticed that the total pressure is not the sum of all the gaseous pressure, but a representative for the total number of an element. The reason to use the conservation of (nominally) total pressure rather than number density is that the condensation is often thought to take place where the pressure rather than volume is constant. Nevertheless, my model allows the total pressure to be changed during the modelling, so it could also simulate the isochoric reaction. Nevertheless, users can iterate the whole P-T space, record the phase assemblage at each condition, and study the process they are interested in.

The second step is to input the species that may participate in the condensation process. The species include monoatomic gas, polyatomic gas, solid, solid solution, and liquid solution (melting). In the current version, I haven't implemented in the liquid-solution API, but in principle it should be similar as the solid-solution one. The reason to separate the monoatomic gas from the gas species is that their pressure are free parameters and needed calculated at each P-T condition. For a polyatomic gas, its pressure can be calculated accordingly:

$$\ln P_{AB} = \frac{G_A + G_B - G_{AB}}{RT} (\ln P_A + \ln P_B) \quad (2)$$

where P , G are the pressure and Gibbs free energy, respectively. The pressure here is normalized to 1 atm, where the standard Gibbs energy is measured. The above equation only demonstrates a simple polyatomic gas AB comprising of elements A and B. The more complex gas could be calculated similarly. Therefore, the pressure of all the polyatomic gaseous phases is bounded to that of the monoatomic gaseous phases.

Different from the gas, the solid also needs to be considered separately. When the solid begins to condense, it would also add constraints in the system. The method to determine the starting point of condensation would be discussed later, and here I will focus on the thermodynamics of the solid and solid solution. Since the activity of a pure mineral is one, the chemical equilibrium cannot derive the amount of the solid. Therefore, the amount of solid species are also the free parameters in the model and should be calculated from mass balance. Similar as the nominal total pressure, the amount of solid is represented by nominal pressure, rather than the number of substance:

$$P_{solid} = n_{solid}RT \quad (3)$$

where P_{solid} , n_{solid} are the nominal pressure and number density of a solid, respectively. Also, each solid species would establish the corresponding equilibrium equation to constrain the gaseous species. For a pure mineral, the equilibrium formula is:

$$\frac{G_A + G_B - G_{AB}}{RT} (\ln P_A + \ln P_B) = 0 \quad (4)$$

where G_A , G_B , G_{AB} are standard Gibbs free energy of monoatomic gas A, B, solid AB; P_A , P_B are the pressure of monoatomic gas A and B. AB is the solid made up of elements A and B. Other pure minerals work similarly.

For the solid solution, the situation would be more complicated than the pure solid. Besides the mass balance and equilibrium equation between the solid and gaseous phase, the chemical potential of each endmember should be considered as well. The free parameters of a solid solution are nominal pressure P_{ss} and molar fraction of each endmember X_i . The nominal pressure is defined similar as other nominal pressure:

$$P_{ss} = n_{ss}RT \quad (4)$$

Also, the molar fraction of all the endmembers should be summed up to 1, so any one endmember can be bounded to the fraction of others:

$$X_j = 1 - \sum_{i \neq j} X_i \quad (5)$$

Therefore, if one solid solution has N endmembers, it only needs N free parameters (1 nominal pressure and N – 1 molar fraction). To solve these parameters, N more equations are needed:

$$\sum_i X_i G_i + G_{mix}(X) = \sum_i X_i (G_{i,gas} + RT \ln P_{i,gas}) \quad (6)$$

$$G_i + \frac{\partial G_{mix}(X)}{\partial X_i} - G_n = G_{i,gas} - G_{n,gas} + RT (\ln P_{i,gas} - \ln P_{n,gas}) \quad (7)$$

$$X = (X_1, \dots, X_{n-1}, 1 - \sum_{i < n} X_i) \quad (8)$$

where X is the vector of molar fraction of N endmembers, but can be represented by the first N-1 endmembers; $G_{i,gas}$ is the standard Gibbs free energy of the monoatomic gas building the corresponding endmember; and $G_{mix}(X)$ is the Gibbs energy change of mixing. The above equations contain one bulk equilibrium equation (eqn. (6)) and N – 1 chemical potential equilibrium of endmembers (eqn. (7)), along with the mass balance, so they can solve the N free parameters of a solid solution.

$$P_{tot} = P_{mono-gas} + \sum k_i P_{poly-gas} + \sum k_i P_{solid} + \sum k_i P_{ss} X_{endmember} \quad (9)$$

The above equation is an example of the mass balance of an element, where k_i is the number of the element one phase would contain ($k = 2$ for Al_2O_3 for Al); P is the pressure defined above. Also, including the equilibrium equations (4)(6)(7) set by the condensed solid and solution, the Quasi-Newton method is used to find the root of this large equation set.

Given a possible solution x_0 , the method can converge to the closest root x in the parameter space. If the proposed solution x_0 is too far away from the actual solution x , then this method would fail. However, the self-consistent method can be applied: each time the P-T condition is changed only in a small step, so the solution x_t of the new P-T condition should be closed to solution x_{t-1} of the old P-T condition. If the user wants to acquire the solution of unknown P-T condition (T, P) , the model would start from a known solution x_0 at (T_0, P_0) , change the (T, P) gradually, and calculate the corresponding solution. In the final step, the model will converge to the solution x_t of the wanted P-T condition.

One feature of this model is to automatically detect the condensation and disappearance of solid and solution. It allows the users to acquire the solution of any P-T condition without considering the possible mineral assemblage. Here I will introduce both the appearance and disappearance criterions. For a pure mineral, the appearance criterion utilizes the equilibrium equation. The basic idea is that before the condensation, the Gibbs free energy of the solid should be larger than the chemical potential of the building gas ($G_{i,\text{gas}} + RT \ln P_{i,\text{gas}}$). After the condensation, the Gibbs free energy of the solid should equal to the chemical potential of the building gas. Just merely below the condensation temperature, the chemical potential of the building gas should be a little lower than the Gibbs free energy of the solid. Therefore, the condensation criterion is:

$$\begin{aligned} \text{If } G_{\text{solid}} &\leq \sum(G_{i,\text{gas}} + RT \ln P_{i,\text{gas}}), \\ \text{Then, set } P_{\text{solid}} &= 10^{-5} P_{\text{ref}}, \\ \text{Add constraint: } G_{\text{solid}} &= \sum(G_{i,\text{gas}} + RT \ln P_{i,\text{gas}}) \end{aligned}$$

where P_{ref} is an arbitrary number, usually set up as the total pressure of Si. This criterion states that after changing the P-T condition, if the Gibbs free energy of a solid is lower than the chemical potential of the corresponding gas, then set the nominal pressure of the solid larger than zero and add the equilibrium equation of the solid into the equation set. The exact P_{solid} would be derived from solving the equation set (4)(6)(7)(9) mentioned above.

The criterion of disappearance of solid would be opposite of the appearance criterion. Instead of examining the equilibrium equation, which is required to be zero, the system would check nominal pressure of a solid after solving the equation set. If a solid would disappear from the system, the nominal pressure would become negative after solving for the root. Hence, the criterion is:

$$\begin{aligned} \text{If } P_{\text{solid}} &< 0, \\ \text{Then, set } P_{\text{solid}} &= 0, \\ \text{Delete constraint: } G_{\text{solid}} &= \sum(G_{i,\text{gas}} + RT \ln P_{i,\text{gas}}) \end{aligned}$$

The appearance/disappearance criterion for solid solution would be more complicated. Given different sets of molar fractions of endmembers, the total Gibbs free energy of the solid solution would be different. Therefore, the mole fractions that minimize the total Gibbs free

energy should be calculated in the first place. Therefore, I add a step to solve for the minimal Gibbs free energy of the solid solution before the appearance criterion:

Find the $X = (X_1, \dots, X_{n-1}, 1 - \sum_{i < n} X_i)$ that minimizes:

$$\sum_i X_i G_i + G_{\text{mix}}(X) - \sum_i X_i (G_{i,\text{gas}} + RT \ln P_{i,\text{gas}})$$

If $\sum_i X_i G_i + G_{\text{mix}}(X) - \sum_i X_i (G_{i,\text{gas}} + RT \ln P_{i,\text{gas}}) \leq 0$,

Then, set $P_{\text{ss}} = 10^{-5} P_{\text{ref}}$,

Add constraint:

$$\sum_i X_i G_i + G_{\text{mix}}(X) = \sum_i X_i (G_{i,\text{gas}} + RT \ln P_{i,\text{gas}})$$

$$G_i + \frac{\partial G_{\text{mix}}(X)}{\partial X_i} - G_n = G_{i,\text{gas}} - G_{n,\text{gas}} + RT(\ln P_{i,\text{gas}} - \ln P_{n,\text{gas}})$$

The method is to minimize the total Gibbs free energy is to calculate the $\frac{\partial G(X)}{\partial X_i} = 0$. It can be solved by applying Newton method to the derivatives. To confirm whether a solid solution will disappear, it is similar as the pure mineral. After solving the equation set, if the nominal pressure of the solid solution is negative, then the solution should be removed:

If $P_{\text{ss}} < 0$,

Then, set $P_{\text{ss}} = 0$,

Delete constraint

$$\sum_i X_i G_i + G_{\text{mix}}(X) = \sum_i X_i (G_{i,\text{gas}} + RT \ln P_{i,\text{gas}})$$

$$G_i + \frac{\partial G_{\text{mix}}(X)}{\partial X_i} - G_n = G_{i,\text{gas}} - G_{n,\text{gas}} + RT(\ln P_{i,\text{gas}} - \ln P_{n,\text{gas}})$$

There is another feature of this model – isolation. The isolation was firstly proposed by Petaev & Wood (1998), but the isolation in their model would be operated at every step, which is not a free function that can be called as users want. In my model, the isolation is designed to be a function that can be called when and where the users want to. The basic principle of isolation is to determine how much condensate would be isolated, decrease their nominal pressure, and subtract the nominal total pressure of the corresponding elements as well.

Similar as the isolation, this model can also add new material into the system. This method is useful when modeling the formation of chondrules, which requires adding dust into the system. This method has similar mechanism as the isolation, so I won't go into details.

[A short example of using the UNICON](#)

One advantage of this model is that users can customize their thermodynamic database. The first step to use this model is to check the thermodynamic data. The default value is mainly taken from Robie & Hemingway (1995). For gaseous species and pure mineral, the model only requires the chemical formula and the formation Gibbs energy at 1 atm at various temperature.

In principle, the temperature should cover the temperature range of condensation. However, the model can extrapolate the formation Gibbs energy to uncovered temperature range. For the solid solution, the additional requested information is the endmember species, Gibbs energy change of mixing.

Provided the thermodynamic data described above, the next step is to set up the nominal total pressure of each element. The total pressure can be modified later during the modeling if users want to. After setting up the model, the users can start to solve for the mineral assemblage. To solve it, the model will start from a known solution x_0 , for instance, the phase assemblage at 1800 K and 10^{-3} atm, modify the P-T condition gradually toward the condition the user inputs, and calculate the phase assemblage after each small change of the P-T condition. The model would converge to the solution $x_{P,T}$ of the input P-T condition at the final step.

When the temperature is low enough, the solid phase will start to condense. Each time changing the P-T condition, the model will iterate all the potential solid phase and test whether the solid/solution would condense. Also, the model would iterate the existing solid/solution phases to test whether they would disappear. The appearance/ disappearance criterions have been illustrated in the above section.

During each step, the users can also call the isolation or adding method to remove/add material into the system. The isolation of solid would not change the chemical equilibrium, but adding material into the system likely change the phase assemblage. It is because the added material may not be stable at this P-T condition. Therefore, the adding method would add the new material in many tiny steps and solve the equation set after each step. After modifying the total pressure by isolation/addition, the system would solve the equation set again to find out the new solution.

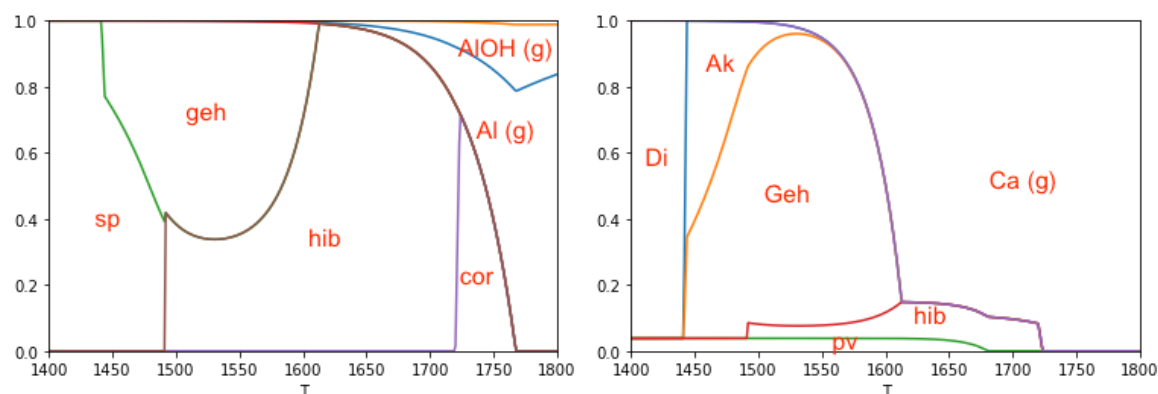


Fig.1. An example of the Al (left) and Ca (right) distribution of solid and vapor assemblage at 10^{-3} atm.

Comparison with the previous model

In principle, all the condensation model based on equilibrium calculation should have the same mathematic principle. That is to say, if all the models use the same thermodynamic data and same P-T condition, the phase assemblage should also be the same.

Instead of optimizing the thermodynamic data, the purpose of this model is to provide an open-source program, which is easy to use and customizable. Users can just use one command and acquire the result they are interested in. It is also easy for users to design the P-T evolution and calculate the phase assemblage along the process. In addition, they can update the thermodynamic data easily and the model still works well. Another main improvement compared with the previous model is adding robust engines to detect the transition of the solid and solution phases and guarantee the users can acquire the solution by just simply inputting the P-T condition and bulk composition. In addition, the UNICON is written in Python, which is more popular than Fortran nowadays.

Besides the common state change regarding the P-T, the model can also isolate the existing phase or adding new material during the modeling. Since the nature is complex, it wouldn't be surprising that the actual reaction would happen in an open system. For instance, the formation of fine-grained Ca, Al-rich inclusion may involve the isolation of hibonite; the chondrule formation would involve adding dust; and the chemistry evolution of protoplanetary disk may involve the removal of the refractory phase (the X-wind model, Shu et al., 1998).

Application

The formation sequence of fine-grained Ca, Al-rich inclusion (FGI)

The Ca, Al-rich inclusion (CAI) is common in the carbonaceous chondrites and the fine-grained inclusions (FGI) occupy a large portion of the CAI. In the famous Allende meteorite, a large portion of the CAI is FGI (Kornacki & Wood, 1976). The FGI get its name from its small size and it can also be easily distinguished by its unique mineralogy assemblage. The FGI contains melilite, spinel, perovskite, fassaite, and anorthite, with rare occurrence of hibonite and olivine. The equilibrium calculation (e.g. Ebel, 2006 and this work) shows the condensation sequence should be melilite > spinel > pyroxene > olivine > anorthite. This result is calculated from a close system with the composition of solar nebulae (Lodders, 2003). However, the actual observation shows that spinel is enclosed by melilite and anorthite by olivine. According to the stratigraphic relationship, the condensation sequence seems to be spinel > melilite > pyroxene > anorthite > olivine. The controversy between anorthite and olivine can be solved if the system has low enough pressure (Pataev & Wood, 1998, Ebel, 2006). However, the condensation sequence of spinel and melilite remains a puzzle. Using the TEM to study the FGI shows the spinel tends to grow on the surface of hibonite and the spinel condensation may be a kinetic effect (Han et al., 2015). The kinetic effect would be obvious only when the gaseous pressure is much larger than the saturation pressure of the solid (Davis & Richter, 2014). The thermodynamic equilibrium calculation shows that when the melilite starts to condense, the saturation pressure of spinel is higher than its gaseous pressure. It means that the kinetic effect should not be prominent at this stage. Even in the views of chemical kinetics, some high-energy gaseous atoms collided with hibonite and formed a spinel with high Gibbs free energy. In the high temperature environment, the spinel would quickly become unstable and decompose. Also, this mechanism may form tiny spinel grains, but it is unlikely to form a large grain, which may occupy a large

portion of a nodule of a FGI (Krot et al., 2004). Therefore, I run the model to explore the potential condensation condition of spinel.

The first trial is to change the total pressure of the system. The total pressure is set ranged from 10^{-8} to 10^{-3} atm. The bulk is the solar composition. The system gradually decreases the temperature given a fixed total pressure. The mineral assemblage is calculated at each temperature step. At lower total pressure, the condensation temperature of all the phases would decrease. It is because the chemical potential of gaseous phase is controlled by pressure and hard to reach the Gibbs free energy of the solid mineral in low pressure. Among these pressure range, the melilite always condenses earlier than the spinel. The gap between their condensation temperature becomes smaller when the pressure is lower. However, even at the pressure of 10^{-8} atm, the gap of condensation temperature between these two phases is still 105 K. This pressure is already low compared with the pressure of the protoplanetary disk that people suggest. Therefore, if the spinel formed prior to the melilite, it would require unrealistically low pressure.

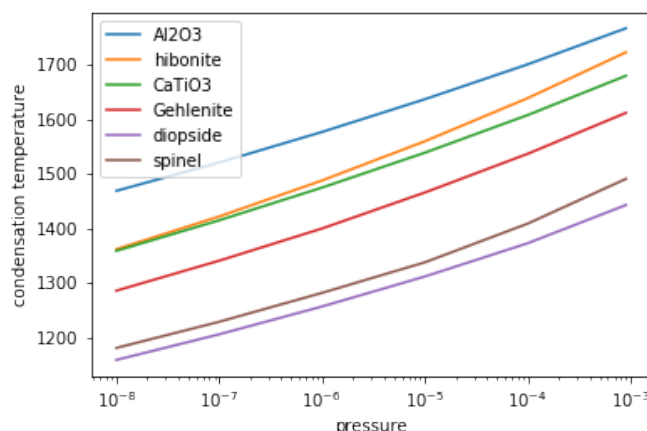


Fig.2. The plot of condensation temperature vs. pressure. Different curves represent different minerals. When temperature decreases, the condensation temperature of all solids decreases. The gap between melilite and spinel gradually decreases when temperature drops.

Another trial is to isolate the refractory phase prior to melilite. The motivation is that the REE of the FGI indicates that some of the refractory phase (e.g. hibonite) was removed prior the condensation of FGI (Davis & Grossman, 1979). Davis et al., (2017) refines the thermodynamic calculation and shows the group-II REE pattern can be generated at various temperature range after formation of hibonite. The removal of hibonite and perovskite results in the removal of the total amount of Ca and Al. In terms of mass balance, the subtraction of Al delays the formation of both spinel and melilite, while the subtraction of Ca only affects the melilite. Therefore, it is possible that the isolation might make spinel condense earlier than melilite. I run the mode under the pressure of 10^{-6} atm, because the FGI may form in such low pressure (Krot et al., 2004). The starting bulk is solar composition. The isolation will occur at 1420K, when the hibonite and perovskite are full condensed. Different isolation factors are applied in the modelling. The isolation factor is defined as the fraction of isolated material over the whole condensate. The

results are shown in Fig.3. The Quotient at Fig.3(1) shows the ability of spinel condensation vs. temperature. The spinel Quotient is defined as:

$$Q = \frac{G_{\text{Mg (g)}} + 2G_{\text{Al (g)}} + 4G_{\text{O (g)}} - G_{\text{sp}}}{RT} (\ln P_{\text{Mg (g)}} + 2 \ln P_{\text{Al (g)}} + 4 \ln P_{\text{O (g)}}) \quad (10)$$

The isolation would harm the condensation Quotient, when all the hibonite is consumed by melilite. When the isolation factor is larger or equal to 5, all the hibonite transforms to melilite, and later transforms to spinel. The competition between melilite and spinel on Al would postpone the condensation of spinel. Therefore, isolation of hibonite would not promote the prior condensation of spinel.

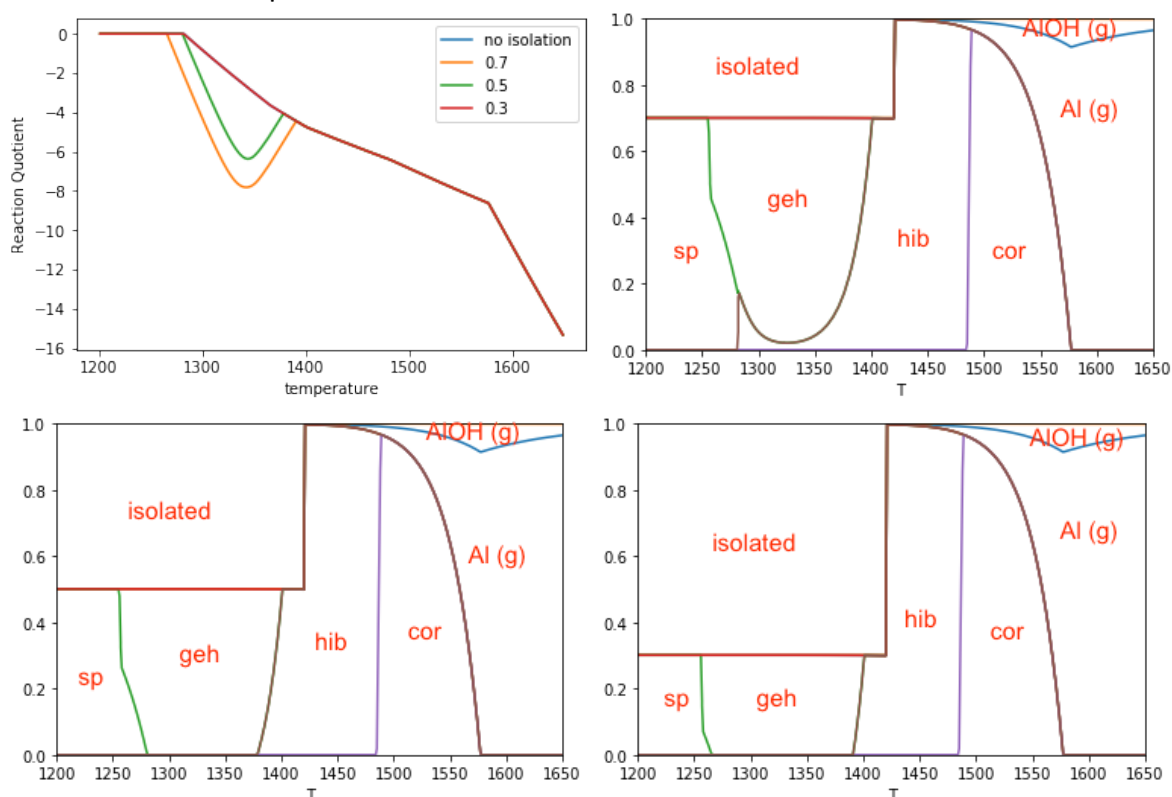


Fig. 3. (upper left) the reaction quotient of spinel vs. temperature. Different curves represent different isolation degree. (upper right, bottom left, and bottom right) The distribution of Al among solid and vapor. They represent different isolation factors (degree): 0.3, 0.5, and 0.7, respectively. When the isolation factor is larger or equal to 5, all the hibonite would transform into melilite (gehlenite).

The formation of fluffy type A inclusion – decreasing pressure or adding more dust?

The CAIs are divided into two groups: type A and B inclusions (Grossman, 1975). The division mainly depends on the existence and abundance of the clinopyroxene, which now is identified as fassaite or Al-diopside. Type A inclusions principally contain little fassaite except in the Wark-Lovering rims, while type B inclusions contain 35 – 60 percent fassaite. In reality, some fassaite are also observed in the mantle of the type A inclusion, but it is made up of far less than 35

percent. In addition, the Type A inclusion can be subdivided into two groups – Fluffy and Compact Type inclusions. The division is based on the morphology of the Type A inclusions. The fluffy type A (FTA) inclusions have irregular shape usually with winding Wark-Lovering rims (Macpherson and Grossman, 1984). They also included the CAIs with nodulous shape in the FTA inclusions, but later Krot et al., (2004) classified them into fine-grained inclusions. Besides the irregular shape, the FTA inclusions also contain the reverse-zoning melilite and V-rich spinel (Macpherson and Grossman, 1984). The melilite in the FTA inclusion can be as coarse-grained as up to 1.5 mm, where the melilite has 20 – 30 mol% of Åkermanite (also defined as Åk number) at the core and 0 – 10 mol% in the rim. Contrast to the FTA, the Compact Type A (CTA) inclusions have a relatively elliptical shape and are considered to crystallize from liquid droplet. On the other hand, given the irregular shape, the FTA inclusion is thought to condense from the solar nebulae gas directly. However, the reverse-zoning melilite needs explanation under the framework of condensation, so Macpherson and Grossman (1984) suggested that reverse-zoning melilite could be formed by decreasing pressure.

Firstly, the model is run with decreasing pressure to reproduce the reverse zoning of melilite. Here I only model the simplest situation: the FTA formed its reverse-zoning melilite in isothermal condition with decreasing pressure. The bulk composition is set as solar composition (Lodders, 2003). I start the model at 10^{-3} atm and 1485 K: the melilite has Åk number of 25, which is closed to the composition of the Mg-rich melilite in the FTA. Then I decrease the pressure of the system from 10^{-3} to 10^{-6} atm, and record the mineral assemblage. The result shows that the Gehlenite is formed when the pressure decreases, so the Åk number decreases accordingly. This process produces the reverse zoning quite well, and the reverse zoning would form from 10^{-3} to 10^{-5} atm.

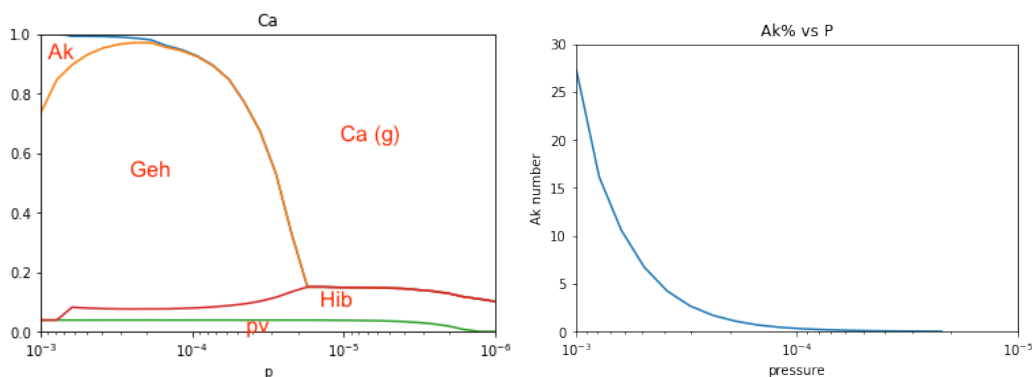


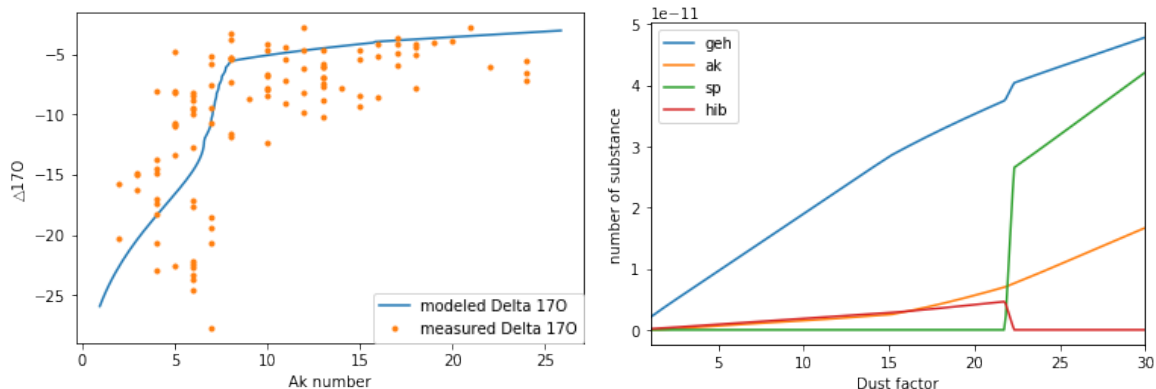
Fig. 4. (left) the distribution of Ca between condensates and vapor. The x axis is pressure of the system. (right) the plot of Åk number vs. pressure. Åk number is the mole fraction of Åkermanite.

An alternative explanation of the reverse zoning is inspired by the oxygen isotopes of FTA. The O isotopes of FTA inclusions have been measured systematically (Harazono and Yurimoto, 2003, Katayama et al., 2012, Kawasaki et al., 2012, 2015, 2017). In terms of reverse-zoning melilite, a correlation between Åk number and $\Delta^{17}\text{O}$ has been found that Mg-poor melilite is ^{16}O -rich and Mg-rich melilite is ^{16}O -poor. Melilite closed to the gehlenitic end (Åk \sim 5) has $\Delta^{17}\text{O}$ of -25‰ , and the $\Delta^{17}\text{O}$ rapidly increases to -5‰ as the Åk increases to 10 - 20. Then the $\Delta^{17}\text{O}$ stays constant at

-5 ‰ when the melilite increases the Åk over 20. The W-L rim has uniform ^{16}O -rich isotopes among its spinel and fassaite.

There are many theories regarding the oxygen isotopic variation in the solar system: for instance, proto-solar UV radiation and the self-shielding (Clayton, 2003), oxygen diffusion (Fagan et al., 2004) and/or the mixing between solar gas and dust (Krot et al., 2010). Krot et al., (2010) have proposed that the solar system might comprise of ^{16}O -poor dust and ^{16}O -rich gas. In term of oxygen abundance, 26% oxygen stayed in the dust and 74% stayed in the gas. The dust here means the material that would eventually incorporated into the solid phase, for instance, all the Al, Ca, Mg, Si, Ti elements. In order to make a ^{16}O -poor environment, dusty region is required, i.e. high dust/gas ratio. Moreover, higher dust/gas ratio results in higher concentration of the elements condensing into solid phase. It is equivalent to higher pressure. Therefore, given isothermal condition, in the dusty region the Mg-rich melilite would condense with ^{16}O -poor isotopes. When they were transported into the region with less dust, where the concentration of the elements forming solid would decrease, the Al-rich melilite would form. This new model intrinsically connects the oxygen reservoir and the pressure together, while the previous model treated the correlation of them as incidence.

In my model, the dust/gas ratio varies from 1 to 15 in order to achieve $\Delta^{17}\text{O}$ ranging from -26‰ to -5 ‰. The initial temperature is set as the condensation temperature of melilite with Åk number of 1 in the normal dust/gas ratio (i.e. the ratio is one). The calculation is run under different pressure from 10^{-3} to 10^{-6} atm. However, in all situation the fassaite would condense before the $\Delta^{17}\text{O}$ reaches -5‰. Therefore, heating effect is also considered. When the temperature increases, the Åk number of melilite would decrease to balance the increment made by adding dust. By increasing temperature and dust/gas ratio simultaneously, the observation of reverse-zoning melilite composition and oxygen isotopes can be reproduced (Fig. 5). Here I will discuss one example of the modeling. The modeling is conducted under 10^{-6} atm. The temperature decreases slowly in the dusty region (dust/gas ratio between 15 and 30) and faster in the dust-less region (dust/gas ratio less than 15). From the Fig.5 – top left, we can see the modeling reproduces the measured value quite well. This can also meet the physical world: the dusty region required heating to form Mg-rich melilite. When the melilite grain moved out of the dusty region, the ambient temperature also decreased accordingly, which suggests that a mechanism gathered the dust and also produced heating.



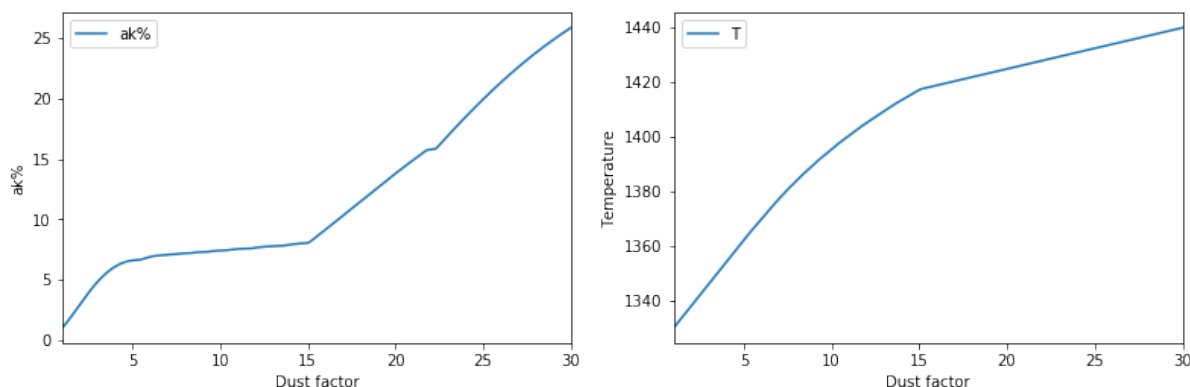


Fig. 5. Result of the model by mixing dust and gas. Dust factor means the dust/gas ratio. (top left) the modeled and measured $\Delta^{17}\text{O}$. (top right) the absolute abundance of different minerals with the evolution of the dust/gas ratio. The unit of y axis is the unit mole of minerals times gas constant times temperature (nRT). (bottom left) the plot of Δk of melilite and dust/gas ratio. (bottom right) the plot of the ambient temperature changing with dust/gas ratio. It implies in order to form reverse-zoning melilite the dusty region should be heated up.

Chemistry of protoplanetary disk and stellar environment

The chemistry of exoplanets has attracted large interest in the planetary science (e.g. Carter-Bond et al., 2012). Since planets were basically accumulated from planetesimals, it is necessary to acquire the chemistry of the planetesimals. An intuitive method is to use the P-T condition and bulk composition of the planetary disk and calculate the chemistry of condensates at different positions. Carter-Bond et al., (2012) used this method and derived the planets with low Mg/Si ratio, which was consistent with the observation (Delgado Mena et al. 2010). However, the derived planets also contain a large portion of Ca and Al, but this may be contradicted to our knowledge at least regarding the solar system. There is no evidence that the Mercury is enriched in either Ca and Al. Also, the enstatite chondrites, which might be the building blocks of the planets of the inner solar system, are depleted in these two elements (Wasson & Kallemeyn, 1988). Shu et al., (1998) proposed that the protosun may eject the material of the inner solar system and transport them into the outer part, and it was confirmed by the observation (Shu et al., 2000). Therefore, it is possible that the refractory phase that condensed in the early stage was transported away. Despite the detail of transportation, the removal of the condensate should be considered when modelling the chemistry of the planetesimals. Given the isolation function of my model, users can set up the isolation at any P-T condition, which can be used to model the ejection of the X-wind model. Also, my model can store the amount of isolated materials, and these materials can be added into the bulk of the outer solar system where the ejecta might settle.

Another potential application is to model the condensation occurring in the stellar interior. Ebel and Grossman (2001) modelled in the condensation of different supernova layers. In their model, they have added various constraints like only monoatomic gas in the vapor and no CO

gas. These constraints can also be added in my model – simply remove the thermodynamic data of the unwanted species. In terms of modeling AGB star, my model can simulate the dredging process by adding new material in the bulk composition during condensation.

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