

Doctoral Dissertation (Censored)

博士論文 (要約)

**Hafnium-tungsten chronology and geochemistry of pallasites**

(パラサイト隕石のハフニウム-タングステン年代学および地球化学的研究)

A Dissertation Submitted for the Degree of Doctor of Philosophy

December 2019

令和元年 12 月博士 (理学) 申請

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## Abstract

Planetesimals play a role as seeds for planetary embryos and planets. Constraining the formation process of planetesimals is crucial for understanding the origin and diversity of planets in our solar system. Since meteorites are the products of a variable process during the early solar system evolution, the planetesimal formation process can potentially be investigated through meteorite chronology and geochemistry. Moreover, meteorites exhibit vast ranges of physical, petrological, and chemical properties, providing a powerful means to investigate the nature and evolution of planetesimals in the early solar system. Undifferentiated meteorites were derived from, at least partly, undifferentiated planetesimals and thus represent materials condensed from the protoplanetary disk. On the other hand, differentiated meteorites represent more evolved materials from differentiated planetesimals. The constraint on the parent body of differentiated meteorites will provide the conditions for planetesimals to differentiate.

Pallasite meteorites constitute one group of differentiated meteorites. These meteorites are composed of roughly equivalent amounts of olivine and FeNi metal. Since olivine and FeNi metal are thought to be the major components of mantle and core of a fully differentiated planetary body, pallasites are expected to possess the information of both planetary mantle and core.

Despite their high importance in exploring the evolution of planetesimal interior, how pallasites formed are unresolved. Based on a slow cooling rate of 2.5 - 18 K/Myr determined for pallasite metal, the size of the pallasite parent body was estimated as 200 – 400 km. However, the predicted size cannot account for the co-existence of metal and olivine because these phases should be efficiently separated by the vast density difference on such a large planetesimal. To explain the co-existence of metal and olivine, several models were proposed, such as convection during the fractional crystallization and impact mixing of core and mantle material. However, a model that comprehensively explains the observed pallasite features remains to be established.

The size estimate of the pallasite parent body is based on a simple planetary cooling model, in which heat production from radioactive decay of extinct nuclides is not taken into account. As inferred from the Al-Mg dating on pallasite olivine, the pallasite parent body could have accreted within 2 Myr after the CAI formation. In such a case, heat production from  $^{26}\text{Al}$  and  $^{60}\text{Fe}$  may play a critical role in the thermal evolution of the parent body. In addition, if olivine and FeNi were mixed significantly later than the core-mantle segregation, these two phases could have different thermal and age records. Therefore, extracting the formation age and thermal history from both metal and olivine of pallasites is necessary to understand the pallasite formation mechanism.

The purposes of this study are to complement the insufficient chronological and thermal records of pallasites and, in turn, to reassess the thermal history and differentiation conditions of the pallasite parent body.

In Chapter 2, we applied the  $^{182}\text{Hf}$ - $^{182}\text{W}$  dating to FeNi metal in pallasites. Tungsten isotopes would vary not only by the decay of  $^{182}\text{Hf}$  but also by the nucleosynthetic anomaly in the solar nebula and the neutron capture effect due to the irradiation of cosmic rays. Therefore, we evaluated the nucleosynthetic anomaly and the neutron capture effect by measuring  $^{183}\text{W}/^{184}\text{W}$  and  $^{196}\text{Pt}/^{195}\text{Pt}$  values. From the measured  $^{183}\text{W}/^{184}\text{W}$  ratios, we found the lack of the nucleosynthetic anomaly in pallasites, indicating that the pallasite parent body accreted in the inner solar system. From the Pt isotopic measurement, we detected the neutron capture effect in some pallasites. The  $^{182}\text{W}/^{184}\text{W}$  values corrected for the neutron capture effect yield a Hf-W age of  $0.08 \pm 1.08$  Myr after the CAI formation, suggesting that the pallasite parent body accreted and differentiated within 1.2 Myr after the CAI formation.

In Chapter 3, we determined the cooling rate of the chromite and olivine in the pallasite Sericho. From the mineralogical observation of the Sericho pallasite, we confirmed that neighboring chromite and olivine were reacting under subsolidus conditions and exchanging  $\text{Fe}^{2+}$  and Mg. We applied the olivine-spinel geospeedometer to this chromite and obtained a prolonged cooling rate of 10 – 30 K/Myr. This estimate is coincident with the metallographic cooling rate of FeNi metal in pallasites.

In Chapter 4, a new thermal evolution model of the pallasite parent body was calculated using the data obtained in this study. By considering the heat production from the decay of  $^{26}\text{Al}$  and  $^{60}\text{Fe}$ , we found that the radionuclides comfort the cooling rate of a planet with 20 km in radius. On the other hand, the cooling rate of a planet larger than 50 km in radius is little affected by the radionuclides due to prolonged cooling time related to its size. The cooling rate obtained by the olivine-spinel geospeedometer could be accomplished at the core-mantle boundary of a 75 km sized planet. The estimated parent body size implies that planetesimals smaller than 100 km in radius would accrete and fully differentiate to make the core and mantle within 1.2 Myr after the CAI formation.

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# **1. Introduction**

## **1.1. The solar system evolution and planetesimal formation**

A fundamental and long-standing problem in planetary science is how our solar system was made and has evolved into its present form. Many scientists tried to explain how the Sun and planets came into existence, for example, Pieere Laplace's "a monistic theory", James Jeans' " a dualistic theory", Bondi and Hoyle's "an accretion theory", and so on (Woolfson, 2000). Now on, "the solar nebular disk model" (Woolfson, 1993) became a favorable model to explain the solar system formation scenario.

At the beginning of our solar system formation, contraction of the molecular cloud led to make a protoplanetary disk. Radiation from the Sun and release of gravitational energy as the molecular cloud collapse heat the material within 3 AU hot enough to vaporize (Woolum and Cassen, 1999). As the protoplanetary disk cools down, the dust particles are aggregated into cm-sized dust agglomerates by Brownian motion, differential vertical settling, and turbulence (Weidenschilling, 1980; Dullemond and Dominik, 2004; Blum and Wurm, 2008; Dauphas and Chaussidon, 2011). As the coagulation of dust agglomerates follows, km-sized planetesimals have formed on the scale of  $10^4$ - $10^5$  years

at 1 AU (Chambers, 2006; Nichols Jr, 2006). If the accretion of planetesimals occurred when short-lived radioactive nuclides, such as  $^{26}\text{Al}$  and  $^{60}\text{Fe}$ , were still extant, radiogenic heating should cause significant melting and planetary differentiation (Hevey and Sanders, 2006). Subsequent growth of planetesimals led to the formation of planetary embryos, finally forming the current solar system (Chambers and Wetherill, 1998; Goldreich et al., 2004).

Planetesimal-driven migration is thought to have an essential role in creating the terrestrial planets. Yet, there are many theoretical difficulties to coagulate dust agglomerates into planetesimals, such as the radial drift barrier and the fragmentation barrier (Weidenschilling, 1977; Blum and Münch, 1993), and the formation mechanism of a planetesimal is unclear. To understand how planetesimals formed, we need to constrain when and how long it took to accrete, how large they grew, and where they accreted. Constraining these parameters from a material science perspective is therefore significant to unravel the solar system evolution.

Nowadays, a vast range of extraterrestrial materials is available, such as meteorites, micrometeorites, interplanetary dust particles, Apollo samples, and Luna samples (Grady and Wright, 2006). Meteorites are most abundant among these samples (more than seven million meteorites in the Meteoritical Bulletin Database) and exhibit a wide variety of

petrological and chemical properties, providing a powerful means to investigate the nature and evolution of their parent bodies and our solar system.

Meteorites are roughly divided into two types of groups, which are undifferentiated and differentiated meteorites. Some undifferentiated meteorites like chondrites include Ca-Al-rich inclusions (CAIs) and chondrules. CAIs record the oldest formation age ( $4567.3 \pm 0.16$  Myr; Connelly et al., 2012) and are thought to have formed as free-floating objects within the solar nebulae. Chondrules are igneous spheres with  $\sim 1$  mm in diameter and they are the major components of most chondrites (e.g. Lauretta et al., 2006). Chondrules were formed through flash heating of high temperature and recondensation, indicating these are formed by thermal processing of particles in nebular shock waves (e.g. McSween et al., 2006). Along with their similar compositions to the Sun (Anders & Grevesse, 1989), chondrites are considered to represent the aggregates of condensed materials within the protoplanetary disk and undifferentiated planetesimals (e.g. Weisberg et al., 2006). In contrast, differentiated meteorites represent differentiated planetesimals or planets. These meteorites are generally thought to represent various layers of the parent bodies. For example, the HED meteorites (howardite-eucrite-diogenite) and iron meteorites are regarded as the representative of the crust and the core of the parent bodies (e.g. Wadhwa et al., 2006). Therefore, by clarifying the age and

formation process of differentiated meteorites, we can expect to constrain the timing and conditions to differentiate planetesimals.

## **1.2. The pallasite meteorites**

Pallasite meteorites are stony-iron meteorites mainly composed of coarse olivine and FeNi metal with minor amounts of troilite, schreibersite, chromite, pyroxene, phosphates, and phosphoran olivine (olivine with high phosphorous content; Buseck, 1977) (e.g. Rayleigh, 1942; Buseck, 1977). Since the significant melting and subsequent differentiation of a planetary body should produce iron core and olivine-dominated mantle (e.g. Righter and Drake, 1997), pallasites are expected to possess the information of both planetary mantle and core. Additionally, from the co-existing texture of olivine and FeNi metal, pallasites are classically interpreted as the core-mantle boundary of the parent body (Buseck, 1977). Most of the pallasites are classified into two major groups, which are the Main Group and the Eagle Station group. Along with some ungrouped pallasites, such as Milton, Zinder, and Northwest Africa 1911, at least five different parent bodies are implied (Boesenberg et al., 2012).

The Main Group is the major group of pallasites, consisting of 59 meteorites out of 125



known pallasites (the Meteoritical Bulletin Database). Hereafter, without special mention, we just express the Main Group pallasites as pallasites. Pallasite olivine shows a restricted range of major element composition. Except for the Springwater pallasite, reported forsterite contents of pallasite olivine range from ~86 to ~89, and the major element composition was homogeneous among individual meteorites (e.g. Boesenberg et al., 2012; McKibbin et al., 2013; DellaGiustina et al., 2019). The restricted variation in fayalite content within pallasites indicates fractional crystallization of olivine during its formation (Scott, 1977) or significant subsolidus homogenization by prolonged annealing. From oxygen isotopic measurement of pallasites, the genetic relationships with IIIAB iron meteorites and HED meteorites were initially pointed out because these meteorites form parallel distribution to the terrestrial fractionation line on the three oxygen isotope diagram (Clayton and Mayeda, 1996; Wasson and Choi, 2003). However, recent high-precision oxygen isotope analysis of HED meteorites and pallasites shows their distinct isotopic compositions (Greenwood et al., 2015). A recent report shows that even pallasites are divided into two groups with a high  $\Delta^{17}\text{O}$  and a low  $\Delta^{17}\text{O}$ , which are  $-0.166 \pm 0.003$  and  $-0.220 \pm 0.003$ , suggesting the multiple origins for pallasites (Ali et al., 2018).

While Mg/Fe composition and oxygen isotopic composition of pallasite olivines are quite homogeneous, there are intra-grain variations in many trace element abundances.

Specifically, Ti, Ca, Cr, V, and Ni decrease from the core to the rim in olivine grains (Miyamoto, 1997; Hsu, 2003; Tomiyama and Huss, 2006). These trends are thought to be the result of a diffusional exchange reaction with FeNi metal during slow cooling (Hsu, 2003; Boesenberg et al., 2012) (see section 3.1 for more details).

FeNi metal, another major phase composing pallasite, shows the variations in trace element concentrations. Though Scott (1977) proposed that pallasite metal could be produced after ~82 % fractional crystallization of IIIAB iron melt based on Au-Ni compositions, other elements such as Ir, Ni, Co, Pt, Ga, Ge, Cu, and Sb do not show a IIIAB-pallasite continuum (Scott, 1977; Wasson and Choi, 2003; Boesenberg et al., 2012).

Pallasite metal often exposes the Widmanstätten pattern like iron meteorites, indicating that the FeNi metal was cooled slowly. Yang et al. (2010) used the Ni-profile across the taenite core of pallasite metal to estimate the cooling rate to be 2.5 – 18 K/Myr, supporting the view that pallasites represent a deep interior of the parent body. However, this cooling rate is slower than those of IIIAB irons, which are 50 – 350 K/Myr (Yang et al., 2010). This discrepancy accompanied by the distinct trace element trends between pallasites and IIIAB irons calls into question the genetic link between these meteorites (Wasson and Choi, 2003; Yang and Goldstein, 2006; Yang et al., 2010; Boesenberg et al., 2012).

The unique petrologic feature of pallasites has rendered it a significant target of various

isotopic dating studies, among which olivine  $^{26}\text{Al}$ - $^{26}\text{Mg}$  chronology yielded the most precise age. The reported  $^{26}\text{Al}$ - $^{26}\text{Mg}$  model age was 1.24 (+0.40/-0.28) Myr after the CAI formation, suggesting the early accretion of the pallasite parent body (Baker et al., 2012). While the Al-Mg chronology gives the constraint on the formation age of olivine, the precise age of the FeNi metal remains to be determined. Moreover, the olivine Al-Mg model age needs to be re-evaluated, considering the recent finding that  $^{26}\text{Al}$  was heterogeneously distributed among the protoplanetary disk (Bollard et al., 2019).

The size of the parent body was estimated from the cooling rate of pallasites, assuming that the parent body was cooled by heat conduction. The estimated parent body radius, which can achieve the cooling rate of pallasite, is 200 – 400 km (Yang et al., 2010; Tarduno et al., 2012). However, such a large size of the pallasite parent body has difficulty in explaining the co-existence of olivine and FeNi metal. When the parent body size is larger than 10 km, the buoyant force should separate olivine and metal into layers in their parent body (Rayleigh, 1942; Wood, 1981), due to the large density difference between these minerals (olivine:  $\sim 3.3 \text{ g/cm}^3$ , FeNi metal:  $\sim 7.9 \text{ g/cm}^3$ ; Mineralogy Database). How to mix olivine and FeNi metal remains the most difficult problem in the pallasite forming mechanism. Rayleigh (1942) imposed that the metal intruded into an olivine layer. The impact mixing model is another popular pallasite formation model. Scott (2007)

suggested that a catastrophic impact caused the mixing of a metal core with an olivine mantle and chained differentiated bodies together. While the mixing mechanisms are favorable to produce dramatically mixed texture and minor phases of pallasites, they are insufficient to describe the chemical characteristics suggesting the fractional crystallization origin of olivine (Boesenberg et al., 2012) and the result of annealing experiment, which suggest that rounding of pallasite olivine was likely to be caused as igneous cumulate (Saiki et al., 2003). Boesenberg et al. (2012) revisited the petrology and geochemistry of major and minor phases of pallasites and proposed a more basic process with the basis of inward metal crystallization (Yang et al., 2010), in which liquid metal crystallizes downward from the base of the overlying dunite layer and intruded into the dunite layer.

While hypotheses noted above were made on the assumption that pallasite represents the core-mantle boundary of the parent body, Tarduno et al. (2012) questioned the assumption. They found the paleomagnetic record in olivine inclusion, implying the existence of core dynamo in the pallasite parent body until olivine was cooled under the blocking temperature. By combining the paleomagnetic data with the cooling rate of pallasites, they implied that pallasite burial depth was ~40 km in the case of a 200 km sized parent body. The scenario was also supported by Bryson et al. (2015), reporting

shallower depth compared to Tarduno et al. (2012) for the pallasite origin. From the paleomagnetic studies, the injection of the core material of the impactor into the parent body mantle was proposed as another mixing mechanism (Tarduno et al., 2012).

In summary, the formation mechanism of pallasites remains enigmatic. In particular, there is a major difficulty in reconciling the slow cooling rate required by the metallographic feature of pallasite metal and the co-existence of olivine and metal with the absence of shock texture (DellaGiustina et al., 2019).

### **1.3. Composition of this thesis**

One possible cause that the discussion does not converge is that the estimation of pallasite parent body size depends on only a simple cooling model. The current celestial thermal models of the pallasite parent body do not take heat source into account. If the parent body accreted very early in solar system history, as implied from the  $^{26}\text{Al}$ - $^{26}\text{Mg}$  dating of pallasite olivine (Baker et al., 2012), we have to consider the heat generated by the decay of short-lived radionuclides, such as  $^{26}\text{Al}$  and  $^{60}\text{Fe}$ . As demonstrated by Hevey and Sanders (2006), the parent body which accreted within 1 Myr would be melted and differentiated by the heat source from  $^{26}\text{Al}$  decay, even if its size was as small as 20 km

in radius. Therefore, determining the pallasite formation age precisely and accurately is essential to discuss the thermal history and the physical condition of the pallasite parent body. In the present study, the age of pallasite metal has been determined by  $^{182}\text{Hf}$ - $^{182}\text{W}$  dating. Moreover, by conducting geochemistry of pallasite chromite and olivine, the cooling rate of pallasite has been further constrained. These new results allow us to reevaluate the accretion time and size of the parent body and discuss the pallasite formation mechanism.

This thesis is organized as follows. In Chapter 2, we would report the new  $^{182}\text{Hf}$ - $^{182}\text{W}$  age determination of pallasite FeNi metal. In addition to the determination of the  $^{182}\text{Hf}$ - $^{182}\text{W}$  age, we also discuss the accretion site of the pallasite parent body based on the nucleosynthetic tungsten isotope composition and re-visit the  $^{26}\text{Al}$ - $^{26}\text{Mg}$  age of pallasite olivine. In Chapter 3, we would give the new estimation of the cooling rate on pallasite olivine and chromite. Based on these results, we discuss the thermal history of the pallasite parent body by incorporating the heat generated by the extinct radionuclides in Chapter 4 and, in turn, the formation mechanism of pallasites.

## **2. Hafnium-tungsten chronology of pallasites**

本章については、5年以内に論文雑誌に投稿予定のため非公開。

### **3. Estimation of the cooling rate on pallasite olivine and chromite**

本章については、5年以内に論文雑誌に投稿予定のため非公開。



## **4. On the origin of pallasites**

本章については、5年以内に論文雑誌に投稿予定のため非公開。

## 5. Conclusion and future aspect

In this study, we determined on the cooling rate of the chromite and olivine, and the Hf-W ages of FeNi metal of pallasites. From the results obtained in these chapters, we estimated the accretion time and accreted size of the pallasite parent body to constrain the early solar system evolution in chapter 4. The key findings of this thesis are as follows.

1. We applied the Hf-W dating to metals of the Brahin, Esquel, Imilac, and Seymchan pallasites. To decide the Hf-W age, the nucleosynthetic anomaly and the neutron capture effect on the pallasites were evaluated by measuring  $^{183}\text{W}/^{184}\text{W}$  and  $^{196}\text{Pt}/^{195}\text{Pt}$ , respectively. The studied pallasites did not exhibit the nucleosynthetic anomaly, while some meteorites show slightly elevated  $\epsilon^{196}\text{Pt}$  values, reflecting that the neutron capture effect does exist. The neutron capture effect may account for the previously reported pallasite metal  $\epsilon^{182}\text{W}$  values lower than the CAI initial value. The  $\epsilon^{182}\text{W}$  values corrected for the neutron capture effect were identical within the error among the four pallasites. The obtained  $\epsilon^{182}\text{W}$  values correspond to the model Hf-W age of  $0.08 \pm 1.08$  Myr after the CAI formation.
2. The lack of nucleosynthetic anomaly on the pallasite meteorites suggests that the

pallasite parent body accreted in the inner solar system, thereby requiring re-evaluation of the previously reported model Al-Mg age of pallasite olivine. The revised Al-Mg age corresponds to  $-0.17 \pm 0.65$  Myr after the CAI formation, which coincides with the model Hf-W age obtained in this study. The age coincidence of FeNi metal and olivine suggests that the crystallization of olivine from the parent body mantle and the segregation of the metallic melt caused at the almost same time.

3. We estimated the cooling rate of chromite and olivine in the pallasite Sericho, by observing iron and magnesium profile across neighboring the olivine-chromite pair. The gained profile was compared with the model profile calculated by modified olivine-spinel geospeedometry. The estimated cooling rate for the pallasite chromite and olivine is 10 – 30 K/Myr around 969 K. The estimated cooling rate of the chromite and olivine is consistent with the metallographic cooling rate of pallasite metal (2.5 – 18 K/Myr; Yang et al., 2010).
4. Considering the closure temperature of the Hf-W chronology, which is 1403 K in olivine, the age difference between the Hf-W age and the Mn-Cr age of pallasite reflect the cooling rate of 12 – 18 K/Myr from ~1400 K down to ~1260 K. The cooling rate estimated from the two chronometry is consistent with the cooling rates of

chromite, olivine, and FeNi metal.

5. The  $\epsilon^{182}\text{W}$  of pallasites are identical or smaller than those of metal in chondrites.

Especially, the  $\epsilon^{182}\text{W}$  of pallasites is lower than that of CR chondrules, suggesting that the pallasite parent body accreted along with forming of chondrules.

6. The obtained Hf-W age of pallasite metal is remarkably older than those of the impact generated iron meteorites, which is hard to reconcile with the impact-induced mixing scenario for the pallasite formation. On the other hand, the Hf-W age of pallasite coincides with those of the oldest magmatic irons, suggesting that the pallasite parent body is one of the oldest differentiated parent bodies in the solar system.

7. Given the early formation of the pallasite parent body, the heat generation by  $^{26}\text{Al}$  as well as  $^{60}\text{Fe}$  should be taken into account for discussing its thermal history. The planetary body with 20 km in radius would be affected by the radioactive heat generation, while the effect on the body with 50 km and larger in radius would be minor. The cooling rate of pallasite oxides, 10 – 30 K/Myr can be accomplished at the core-mantle boundary of a 75 km sized body. By considering the presence of a regolith layer on the surface of the pallasite parent body, the estimated size would be

even smaller as 50 km.

8. For better size estimation, several terms that disregarded in the model should be considered. These terms include the effect of convection, various physical parameters of the regolith layer, and the initial temperature distribution.
  
9. The obtained results and discussion suggest that a planetesimal with a radius smaller than 100 km accreted within 1.2 Myr after the CAI formation and underwent global melting and differentiation, which led to the core-mantle layered structure.

## Acknowledgements

First of all, I would like to express the utmost gratitude to my supervisor, Prof. Tsuyoshi Iizuka (Univ. Tokyo). His continuous guidance and support helped me to complete this Ph.D. thesis.

I thank Profs. T. Hirata, S. Nakai, H. Hiyagon, and T. Mikouchi (Univ. Tokyo) for their constructive comments, which improved this Ph.D. thesis. I greatly appreciate my co-worker Prof. A. Ishikawa (Tokyo Tech) for giving me useful advice on the chemical separation of platinum group isotopes, and kindly supporting the chemical procedures. I am also grateful to Drs. H. Yoshida, K. Ichimura, and A. Kobayashi (Univ. Tokyo) for their technical support on handling SEM/EDS, FE-EPMA, and ICP-MS. Prof. A. Yamaguchi and Dr. A. Takenouchi (NIPR) helped me to handle SEM-EBSD, and I want to show gratitude for their cooperation. I want to thank Profs. T. Yokoyama (Tokyo Tech) and T. Hirata (Univ. Tokyo) for providing isotopic standards used in this thesis.

The discussions with and advice from Prof. K. Ozawa (Univ. Tokyo) helped improve my investigation, and I would like to appreciate for his great help. I am grateful to Drs. K. Itano (Kanazawa Univ.) and Y. Hibiya (JAMSTEC) for their innumerable assistance during years spent as the members of the same laboratory and even after their graduation. I will thank Mr. Y. Sato (Univ. Tokyo) and Mr. S. Shimizu (Chiba Univ.) for helping me to implement the numerical calculation. I appreciate Prof. Y. Takahashi and Ms. M. Kurisu (Univ. Tokyo) for many kind favors during some experimental troubles. Continuous support from the administrative staff in the Department of Earth and Planetary Science of the University of Tokyo is also appreciated.

My research project was supported by the JSPS Grant-in-Aid for JSPS Research Fellow Grant Number 17J04987. This study was also partly supported by the JSPS Grant Number 15H05833.

Yoshitaka Homma  
Tokyo, December 2019

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