Research Article

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The Mg/Fe ratio of silicate minerals in the meteoritic materials and in the circumstellar environment: A case study for the chondritic-like composition

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Abstract: Kaba meteorite as a reference material (one of a least metamorphosed and most primitive carbonaceous chondrites fell on Earth) was chosen for this study providing an adequate background for study of the protoplanetary disk or even the crystallization processes of the Early Solar System. Its olivine minerals (forsterite and fayalite) and their Mg/Fe ratio can help us to understand more about the planet formation mechanism and whether or not the metallic constitutes of the disk could be precursors for the type of planets in the Solar System. A multiple methodological approach such as a combination of the scanning electron microscope, optical microscope, Raman spectroscopy and electron microprobe of the olivine grains give the Fe/Mg ratio database. The analyses above confirmed that planet formation in the protoplanetary disk is driven by the mineralogical precursors of the crystallization process. On the other hand, four nebulae mentioned in this study provide the astronomical data confirming that the planet formation in the protoplanetary disk is dominated or even driven by the metallic constituents.

Keywords: meteorites, circumstellar environment, planet, mineral, astrophysics

1 Introduction

Mg, Si and Fe are the most important mineral-forming elements in the Solar System, which play a dominant role in the buildup of the mantle, as well as, the crust of silicate-rich planets. The two major types of the cosmic silicate mineral grains are the groups of the olivine (Ol) with a general formula of Mg$_{2x}$Fe$_{2x}$SiO$_4$ (x is between 0 and 1, end-members are forsterite-Mg$_2$SiO$_4$, and fayalite-Fe$_2$SiO$_4$) and the pyroxenes (Px) with a general formula of Mg$_{x}$Fe$_{1-x}$SiO$_3$ (x is between 0 and 1) (enstatite-MgSiO$_3$, ferrosilite-FeSiO$_3$). In protoplanetary disk (PPDs) and the interstellar medium (ISM), the presence of silicate minerals can be expected in amorphous and/or in crystalline forms. The later ones can be classified into two crystallographic systems based on their internal structures: orthorhombic such as Ol, and a certain type Px; and monoclinic (another part of Px group) (Henning 2010).

The compositions and origin of crystalline or amorphous ferromagnesiolsicates are poorly understood, however, they are thought to be derived from supernovae (SN) and the atmosphere of a low-mass red giant or asymp-
totic giant branch stars (AGB) with close-to-solar metallicity (Floss and Haenecour 2016). Trigo and Rodriguez et al. (2009) noted that an intermediate-mass AGB star is favoured to explain SLN initial abundances in chondritic meteorites. A laboratory experiment showed that a mixture of magnesiosilica and Fe-rich metallic grains may yield ferromagnesiosilica minerals (Rietmeijer 2013). The composition of Earth’s lower crust and mantle is dominated by Mg-rich silicates. Astronomical observations also support that the interstellar silicates are Mg-rich (Min et al. 2007), due to the higher abundance of Mg as opposed to that of Fe. Note that, the Mg/Fe ratio for the case of a highly forsteritic olivine (Fo=0.99) with a small amount of Fe can be described by the formulation of $Mg_{2(2-2x)}Fe_{2x}SiO_4$ where $x=0.01$. The Mg/Fe ratio in silicates may be an important factor for examining the material properties in a terrestrial mantle, therefore, we attempt to discuss shortly the observed compositions up to date with their implications on silicate chemistry in circumstellar environments. The major components of a typical carbonaceous chondride (CC) are the chondrules, the interstitial fine-grained matrix consisting of hydrous and anhydrous silicates, oxides, sulfides, metals and organic matter, the Ca-Al-rich inclusions (CAIs), the amoeboid olivine aggregates (AOAs), and isolated grains with forsteritic- and fayalitic olivine composition embedded in the matrix, respectively. The CCs may contain a few percent carbons up to 5% by weight (Gilmour 2003). The main part of the carbon can be found in the organic matter and carbonates; however, very important carbon-based minerals are graphite and diamond. Moreover, CCs also may contain a small amount of silicon carbide, which is a very characteristic carbon-based compound in meteorites containing presolar grains from the AGB stars, supernovae, and novae (Gilmour 2003).

In CCs, highly forsteritic ($Mg_2SiO_4$) olivine can be found in the greater part of the petrological components (e.g., chondrules, AOAs, matrix minerals) (Gucsik et al. 2013), however, enstatite ($MgSiO_3$), ferrosilite ($FeSiO_3$) and fayalitic olivine ($Fe_2SiO_4$) also may occur in the texture. The abundance of fayalitic olivine is much smaller than that of the forsterite.

Based on the Solar System abundances of the elements on meteorites (CI chondrites), the dominance of Mg over Fe was inferred by Anders and Grevesse (1989) and Lodders et al. (2009). In addition, analytical examinations show that in cometary materials the forsterite is a major constituent of the olivine grains having an overabundance of Mg over Fe (Jessberger 1999; Hanner 2003; Wooden and Lindsay 2011; Foster et al. 2013; Lindsay et al. 2013; Rietmeijer 2013).

Chondrites exhibit low Al concentrations ranging from 0.2 to 7.0 wt.% (Nittler et al. 2004). In comparison, according to the results of Hezel et al. (2008), the bulk Al concentration in the examined CV chondrites is 1.68 wt.%, while the Mg concentration is obtained to be 14.3 wt.%. Interestingly, the Fe concentration in CV chondrites is measured to be 23.5 wt.%. Namely, the abundance of Mg is higher than that of Fe in the CV chondrites considering the molar masses of these elements. The abundance of Al and Ca in silicates is much smaller than that of Mg and is also smaller than that of Fe (Nittler et al. 2004). Table 1 summarizes the FeO and MgO abundance, as well as the Mg/Fe ratio of various meteorites and groups including the CV, falls, too.

### Table 1. The FeO and MgO abundance, and Mg/Fe ratio of various meteorites and groups.

| Meteorite or meteorite group | MgO (wt%) | FeO (wt%) | Mg/Fe |
|-----------------------------|-----------|-----------|-------|
| Renazzo (olivine chondrule core) | 56.7 | 1.29 | Fe0.972 |
| Renazzo (olivine chondrule rim) | 55.9 | 1.71 | Mg0.962 |
| Orgueil (bulk) | 15.91 | 23.6 | Fe0.657 |
| Kaba (bulk) | 23.9 | 28.64 | Mg0.393 |
| CI | 16.1 | 23.4 | Mg0.348 |
| CM | 19.4 | 27.0 | Fe0.652 |
| CO | 24.05 | 31.9 | Mg0.358 |
| CV | 24.05 | 30.2 | Mg0.393 |

Silicates are the most abundant grain species around young main-sequence stars (Waelkens et al. 1996). According to spectroscopic examinations, silicates in the interstellar medium (ISM) and in the protoplanetary disks (PPDs), are mostly Mg-rich. Most of the terrestrial-like exoplanets are thought to have been accreted from undifferentiated PPD materials, which are compositionally similar to the chondritic meteorites. In this study, we assumed the chondritic-like composition in most of the circumstellar environments in the Galaxy. We have made a simple model for the Mg/Fe ratio based on the mineralogical investigation of silicates in the carbonaceous matrix of the Kaba (CV3). In this paper, we used the most relevant data provided by Futó and Gucsik (2017) based on the previous results of Gucsik et al. (2013).
2 Experimental Procedure and Samples

2.1 Chemical composition of the Kaba sample

For examining the Mg/Fe ratios in given constituent minerals, the results of previous studies were used (Gucsik et al. 2013; Futó and Gucsik 2017). Moreover, the analyses focused on the investigation of the major element composition of the constituent minerals to estimate the Mg/Fe ratio. The quantitative analyses of olivine for major and minor element composition were performed by using a wavelength-dispersive spectrometer (WDS) coupled to a JEOL JXA-8900R SEM (scanning electron microscope) at Okayama University of Science, Okayama, Japan. Further details of the above-mentioned technique, as well as sample preparation, can be found in Gucsik et al. (2013).

2.2 Micro-Raman spectroscopy and 3D microscopy of the Kaba sample

Renishaw InVia Raman microscope (Renishaw, Wotton-under-Edge, United Kingdom) was used for the mineral characterization of the Kaba meteorite in the range between 100-3200 Raman shift/cm$^{-1}$. Raman beam size is around 2 microns. The spectra were collected using the 100x Leica objective. The laser used for the mapping was 532 nm excitation with 2400 l/mm grating at 10% of 50 mW laser power with 50 µm beam size. The recorded rectangle map contains 500 acquisitions (10 s exposure time at each point, 2 accumulations) with 30 µm step size. Beam centring and Raman spectra calibration were performed daily before spectral acquisition using a built-in Si standard.

Photos of the Kaba meteorite were recorded by a digital 3D microscope (Keyence VHX-6000, Keyence, Osaka, Japan). 100x-1000x zoom lens were used at 200x and 400x magnification with polarized trans- and reflective illumination.

The instruments above were used at Institute for Nuclear Research (ATOMKI), Debrecen, Hungary.

2.3 Mineralogy of the Kaba sample

The Kaba meteorite fell on April 15, 1857, around 22h. Its actual weight is 2,601 kilograms. The extraterrestrial origin was recognized by dr. József Török, a teacher of natural history at the Reformed College of Debrecen. It is among the first meteorites in which organic material was found. One of the rarest types of meteorites (CV3), it contains relatively large chondrules (Nagy 2007).

The Kaba belongs to the oxidized subgroup of the CV3 carbonaceous chondrite class (McSween 1977; Peck 1984). The lithological diversity of Kaba is obvious. Though the ratio of the main petrographic components may vary by various authors indicating the variety in the texture and mineralogy, we can conclude that the total volume of chondrules consisting of olivine and pyroxenes in various sizes and types, is ~30%. The amount of olivine-rich inclusions is ~2% (in volume), while the amount of Ca-Al-rich inclusions is 3% (in volume). In addition, small (below 20 µm) mineral fragments (olivine (Fa10-90), feldspar, enstatite, hedenbergite, diopside, andradite) in a hydrocarbon-impregnated, phyllosilicate-bearing fine-grained opaque matrix (~55-69% of total volume) can be also found (Sztrókay et al. 1961; Gucsik et al. 2013). Despite the effects of high-temperature hydrothermal and aqueous alterations, the Kaba can be regarded as one of the least metamorphosed ones within the CV3 carbonaceous chondrite group.

According to the currently available data, the constituent parts of the meteorite formed at a temperature of 2000-5000 K, with selective condensation in different areas of the Solar System. Melt droplets (chondrules) of varied composition were cooled and have formed together with the debris of previous collisions. During the process, forsterite, pyroxene and anorthitic minerals were formed. The characteristic feature of the Kaba meteorite is that it underwent the lowest degree of thermal metamorphosis in CV3 carbonaceous chondrites.

A thin section was made from the Kaba meteorite sample, which was mounted in a non-radiative epoxy material. It was polished by silicon colloidal liquid to avoid any pollution material (Figures 1a and 1b). The petrological investigation of the chondrule region and its vicinity is demonstrating that there are neither coexisting secondary crystallization effects nor dislocation patterns in the olivine grain and its matrix material. This indicates that the selected grain is a pristine material, which was not affected by other processes (Figures 1a and 1b).

3 Results and Discussion

3.1 Raman spectra of Kaba carbonaceous meteorite

Raman spectral properties of the carbonaceous matrix of the Kaba (Figure 2a) meteorite samples are characteris-
**Figure 1.** A transmitted optical microscope image shows the Kaba meteorite thin section (A). The selected olivine grain is distinguished by a red square. Cross-polarized picture of an olivine grain exhibits homogeneous and well-crystalline structure (B).
Figure 2. (a) Raman spectrum of the Kaba meteorite shows well-pronounced peaks of olivine; (b) 2D Raman mapping image of an olivine grain showing the forsteritic olivine (pale blue), sulphides (green), graphite (blue) and the metals (red) set into the carbonaceous matrix.

The Raman spectral features of olivine also confirm that the forsteritic content of Kaba carbonaceous meteorite is affected by the amorphization, which is in good agreement with Gucsik et al. (2013).

The 2D Raman mapping of the Kaba meteorite shows that the olivine grain is dominantly coexisting with the carbonaceous phases and exhibits the relatively low iron but high magnesium content (Figure 2b). The forsteritic content of olivine may refer to the crystallization processes of the Early Solar System.
3.2 Aqueous alteration, metamorphism and metasomatism of the Kaba meteorite

Most of the chondrites are considered chemically primitive objects as their chemical composition is very close to the Sun’s elemental composition. Since various components in chondrites are derived from diverse environments (or parts of the protoplanetary disc) the composition may be heterogeneous even within a single meteorite. Thermal and shock metamorphism, as well as aqueous alteration, may affect the primary composition of the chondrites resulting in a chemical and isotopic equilibration. The degree of thermal metamorphism and aqueous alteration may vary among the chondrite classes, however, there is no sharp boundary between the thermal metamorphism and aqueous alteration (Huss et al. 2006). Members of the carbonaceous chondrite class can be regarded as the most primitive or pristine materials of the Solar System, however significant differences among the carbonaceous chondrite groups and subgroups occur regarding the degree of alteration and metamorphism. CV3 carbonaceous chondrites also suffered thermal and shock metamorphism, and aqueous alteration in various degrees resulting in secondary processes and mineralization in the texture (Krot et al. 1997; Huss et al. 2006).

The presence of water- and OH-bearing phyllosilicates are thought to be a primary signature of aqueous alteration in carbonaceous chondrites (Trigo-Rodríguez et al. 2015, 2019), as well as other secondary minerals, such as fayalite, Ca-Fe pyroxene, magnetite, and andradite also indicate fluid-rock interaction (Krot et al. 1997). Keller and Buseck (1990) found Fe-rich saponite in the matrix, in the CAIs, as well as in the altered chondrules (in glass and enstatite) by using the EPMA method, indicating low temperature (<100°C) hydrothermal alteration. Gucsik et al. (2013) discussed the presence and significance of zoned olivine and concluded that the forsterite was controlled by hydrothermal alteration at about 250°C. The abundance of iron oxide-rich fractures in chondrules and AOAs suggests intense metasomatism of iron-rich fluids at low temperature, as a parent body process (Gyollai et al. 2018), while Krot et al. (1997) showed Ca, Si, Mg, and Fe mobilization and redistribution due to progressive metasomatism. Also, Krot et al. (2018) based on oxygen-isotopic analysis of certain Fe-bearing components showed that the secondary fayalite and magnetite formed during aqueous alteration of rock between 200-300°C. C and N elemental abundances and isotope ratios in the bulk also reflect the degree of thermal alteration of indigenous organic matter. An alteration scheme for CV3 carbonaceous chondrites proposed by Pearson et al. (2006) based on C and N isotopes ratios and abundances shows that the Kaba is the least metamorphosed member of the CV3 carbonaceous chondrites.

3.3 Mg/Fe ratio of Solar System

The data and the calculated values listed in Table 2 also show that the silicates in the Solar environment are Mg-rich minerals. The smallest Mg/Fe ratio is found in the case of the sample of Fa 8-2, the highest value has been found for the case of the sample Fo8-2, in which the silicate compound is composed almost entirely of olivine.

Table 2. The FeO and the MgO abundances in different mineral structures of the Kaba meteorite (CV3) and in the bulk chemical composition of the matrix of the Allende (CV3) chondrite. The data have been taken from Futó and Gucsik (2017). The Mg/Fe ratio is being calculated from the concerning data that are taken from Gucsik et al. (2013).

| Kaba (CV3) mineral structure | Mineral sample | MgO (wt%) | FeO (wt%) | Mg/Fe |
|-----------------------------|----------------|-----------|-----------|-------|
| Porphyritic chondrule        | Forsterite Fo1-1 | 0.55       | 0.30      | Mg0.993 Fe0.007 |
| Granular ol-px chondrule    | Forsterite Fo3-1 | 0.5499     | 0.89      | Mg0.98 Fe0.02 |
| Isolated olivine grain       | Forsterite Fo3-2 | 0.5470     | 1.02      | Mg0.976 Fe0.024 |
| Complicated aggregate       | Forsterite Fo8-2 | 0.5691     | 0.21      | Mg0.995 Fe0.005 |
|                            | Fayalite Fa9-2   | 0.26       | 0.68      | Mg0.003 Fe0.997 |

Fe is one of the abundant cations of the silicates due to its high cosmic abundance. At the same time, Fe has been frequently appeared in the oxides (FeO, Fe2O3, Fe3O4), too. Note that Mg-rich minerals form by equilibrium condensation in stellar environments. However, non-equilibrium condensation can result in higher Fe contents and such processes may account for the Fe-rich grains (Bose et al. 2010). Moreover, such processes may be responsible for the Fe-enrichments in the carbonaceous chondrites.

The Mg-rich silicates are more frequent than that of other types of silicates owing to the higher abundance of Mg as opposed to that of Ca or Al. Accordingly, the Mg-rich silicate constituents are characteristic features of the composition of the minerals in the atmospheres of stars, the ISM and thus the Mg-rich silicates are thought to have been dominated in most of the circumstellar environment and PPD’s, respectively.
The relatively high ratio of Fe in silicates in our Galaxy might be derived from cases as follows: (1) Fe-rich grains were ejected by supernovae that polluted the interstellar medium from which a host star and its planetary system have formed. (2) Silicates, with a relatively high Fe/Mg ratio, that can be found in circumstellar environments in the outer galactic region may have formed in the inner or central region of the Galaxy (in metal-rich zones) and later migrated outward (Simonia 2017a and Simonia 2017b-and references therein).

3.4 Spectral peculiarities of cosmic olivine and pyroxenes

The dust of the circumstellar environment (disks, shells) is constantly subjected to illumination by short wavelength electromagnetic radiation from the central star. Circumstellar dust grains of micron and nanometer size continuously absorb and reprocess the infrared radiation. Dust grains of different mineral compositions and shapes are also subjected to constant bombardment by fluxes of fast electrons and protons. As a result of these interactions, the physical and chemical properties of the circumstellar dust grains may evolve. Different forms of olivine and pyroxene dust were detected in the circumstellar disks, shells, and planetary nebula dust matter. In the infrared spectra of these astrophysical objects, spectral features of olivine and pyroxenes were detected at 11.2, 20, 24, 28, 34 and 43 µm (Waelkens et al. 1996; Waters et al. 1996; Nuth et al. 2005).

The method for revealing and identification of cometary crystalline silicates was suggested by Simonia (2017a,b). The presence of forsterite and enstatite dust particles in comet 122P/deVico dust halo was proved on basis of revealing luminescence of these mineral particles (in the spectrum of this comet) (Simonia and Gucsik 2019). For the case of the cometary forsterite dust – luminescence emissions are centred at 4320 (432 nm), 4520 (452 nm), and 7150 Å (715 nm), but for enstatite grains – luminescence emissions at 4000 (400 nm) and 6740 Å (674 nm).

In our opinion, it will be reasonable to investigate the evolution of physical and chemical features of olivine and pyroxenes from the stage of circumstellar dust environment to cometary dust halo. For the implementation of such research, it will be necessary to obtain: 1) infrared spectra of the circumstellar dust (stars of various spectral types); 2) optical spectra of hyperbolic comets; 3) optical spectra of the periodic comets with P > 400 years. Identification of cometary olivine and pyroxenes will be possible using its luminescence detection. Such kind of research will be effective by using of orbital instruments. Archived data of astrophysical observations and laboratory experiments may play an important role in the investigation of cosmic olivine and pyroxenes.

Such kind of investigations may be extended to the special class of planetary nebula – O-rich nebula. The main steps of such investigations may be the following: 1) obtaining infrared spectra of O-rich planetary nebula on bases of orbital instruments; 2) obtaining laboratory infrared spectra of olivine and pyroxenes at temperature T ≤ 54 K; 3) the comparative analysis of obtained lab spectra with astrophysical spectroscopic data. It will be important to identify the state of olivine and pyroxene minerals in planetary nebula dust (amorphous and crystalline states). Lab data will be unique “tool” for such identification. We selected objects with: 1) relatively intense infrared radiation; 2) magnitude < 13; 3) presence of O atoms or ions. The infrared radiation of nebula in J, H, K bands may be considered as an indirect sign of silicate dust presence in nebula matter. Several planetary nebulae are listed in Table 3, as follows.

The NGC 6153 planetary nebula is a metal-rich planetary nebula at a distance of 1.36 kpc (Gaia Collaboration 2018). It has been well modelled by a 3D bi-abundance model by (Yuan 2011). Gómez-Llanos and Morisset (2020) estimated the abundances of the heavier elements in the nebula. In the metal-rich dust – in logarithmic value – the results are to be Mg = Si = Fe = −1.0. These are higher values than of ones in the gaseous phase. The C and O are the same or lower in this phase (Table 2, Figure 3a). The Cat’s Eye Nebula (NGC 6543) has a well-known bipolar structure with a pair of jets (Table 2, Figure 3b). Its spectra show strong

Table 3. Some nebulae and their right ascension (α) and declination data (δ).

| Object name | Magnitude | Right Ascension (α) | Declination (δ) |
|-------------|-----------|---------------------|-----------------|
| IC 5117     | 11.297    | 10h 17m 30.96s      | +44d 35m 47.64s |
| NGC 6886    | 12.219    | 20h 12m 2.90s       | +19d 59m 23.0s  |
| NGC 6884    | 11.159    | 20h 10m 23.70s      | +46d 27m 39.0s  |
| NGC 6572    | 8.585     | 18h 12m 06.21s      | +06d 51m 13.40s |
| PN SwSt1    | 10.517    | 18h 16m 12.27s      | −30d 52m 07.79s |
Figure 3. (a) The NGC 6153 is a metal-rich planetary nebula (the image is credited from ESA/Hubble); (b) Cat’ Eye Nebula (NGC 6543) containing relatively high abundance of metallic composition such Mg and Fe (the image is credited from NASA)
molecular lines. The OC-rich nebula’s dust feature is hard to study because of the low-quality spectrum available, only. Delgado-Inglada et al. (2016) found the nebula’s Fe++ average abundance to be 4.93+/-0.08 (12 + log (Fe++/H+)). Based on ISO (Infrared Space Observatory) measurements, this nebula includes both amorphous and crystalline silicates, respectively (Bernard-Salas and Tielens 2005; Cohen 2005). A detailed study of the SwSt1 nebula and its star can be read from Sterling et al. (2005). SwSt 1 is an O-rich planetary nebula around a C-rich central star (De Marco 2001) (Table 2). This young and compact O-rich nebula exhibits lines emitted by warm silicate dust. The mass ratio for C/O is close to the one of the Sun, while the abundance of N is very low, (N/O = 0.08). Forbidden lines of Fe+ and Fe3+ could be also found. The Cn 1-5 planetary nebula is an ionized carbon-rich nebula with NiII and FeII lines. Its double halo (Delgado-Inglada and Rodríguez 2014) is a so-called AGB halo (according to the signs of thermal pulse on the asymptotic giant branch) (Table 2). On the Spitzer spectra, one can find strong PAH (polycyclic aromatic hydrocarbons) bands and weak crystalline silicate features. The C/O ratio of 1.29 is low, however, the enrichment of He and N is high.

The study of the above-mentioned planetary nebula can help us to understand more about the formation mechanism of the solar-type protostars. Therefore, the chemical composition of planetary nebulae, especially the metallic abundance, would be related to the precursors of the protoplanetary disk similar to the Early Solar System (ESS). However, it is important to note that the diffusion of the nebulae in the Interstellar Medium (ISM) can be efficient, so the final contribution for a condensing star is relatively minor (see e.g., the dilution values considered to explain the Short-Lived Nuclides (SLN) in the ESS by Trigo and Rodríguez et al. 2009).

4 Conclusion

In this study, a combination of optical microscopy, electron probe microanalysis, scanning electron microscopy and Raman spectroscopy gives detailed information about the chemical and structural properties of the minerals. The results would be summarized as follows. The mineral composition of the Kaba meteorite is rich in highly forsteritic (Fo > 0.99) olivine. In addition, the data also shows that the silicates in the Solar environment are mostly Mg-rich minerals, too. The high ratio of Mg, Si, and O of the elemental abundances in chondritic meteorites refer to the dominance of magnesium silicates, which have an abundance higher than that of the abundances of Fe-, Ca-, Al-silicates. The composition of the planet-building materials is assumed to be Mg-rich in the most circumstellar environments. It is a key factor in relation to the chemical evolution of the Galaxy. Consequently, it would be useful to compile a spectral atlas of the optical and infrared emissions of cosmic olivine and pyroxenes from different objects including asteroids, comets, and nebula dust.

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