

Research Article

István Viczián* and Ferenc Kristály

Clay minerals and other hydrous alteration products in the Kaba meteorite: Review of the literature and new XRD investigations

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Abstract: The present study is a review of publications regarding special aspects of the mineralogy of the Kaba meteorite, clay minerals, hydrous phases, and organic matter. In Section 1, the history of fall and finding of the meteorite and the first classical mineralogical description in the Year 1961 are mentioned. The most frequent alteration product of primary silicates and the glassy matrix is Fe-bearing saponite. By modelling of hydrothermal alteration of Allende-type meteorites, artificial saponite could be produced. The thermodynamic study proved the possibility of hydrous neof ormation of silicates and magnetite at low temperatures, mostly below 100°C. The formation of Fe-rich external rims in forsterite grains by hydrothermal action was demonstrated by electron probe microanalysis and scanning electron microscopy-cathodo-luminescence methods. Foliation in CV3 carbonaceous chondrites indicates that the Kaba meteorite is derived from the external, shallow zone of the parent body. A combination of X-ray power diffraction (XRD) and Mössbauer spectroscopy made the exact modal analysis of CV3 chondrites possible, including Kaba. The study of alteration by *in situ* determination of stable oxygen isotope contents may provide constraints regarding the oxygen isotopic composition of the aqueous fluid on the CV parent asteroid. The degree of thermal metamorphism of the organic matter was determined by using Raman spectroscopy. The presence of aromatic and aliphatic, saturated and unsaturated hydrocarbons, and various amino acids was demonstrated. The non-biogenic origin is more probable; however, there are textural and mineralogical observations that may indicate also the biogenic origin of organic matter. In addition

to the review of the literature above, as an appendix, shortly new XRD investigations of the authors are reported that indicated smectite-like phases in a thin section made of the meteorite.

Keywords: Kaba meteorite, carbonaceous chondrites, saponite, hydrous alteration, organic matter.

1 Introduction

The conditions of the fall of the Kaba meteorite in the Year 1857 were described by Nagy (2008). He also reviewed the contemporary cultural background and the history of further scientific investigations. The meteorite fell on relatively dry ground, and it was found already on next day near the surface; after a few days, it was transferred to the Debrecen Reformed College, where it is preserved since that time in a protected museum environment. Therefore, a secondary, terrestrial alteration is not probable and was not considered by most of the authors.

The mineralogical studies were reviewed by Bérczi *et al.* (1998) in the introduction of their study, dealing mainly with signs of foliation in the meteorite. Solt *et al.* (2000) compared chemical analyses of chondrites with special respect to water contents, correlation of the components H and Na, and various forms of iron, such as metallic, sulphide, reduced, and oxidized.

An excellent review of alteration processes can be found in the study by Kimura and Ikeda (1998). A detailed discussion is devoted to the Kaba meteorite in the textbook of Kubovics (2008) “Általános kőzettan” (General Petrology), in the chapter on meteorites. This book is in the Hungarian language; however, it may be interesting also at the international level because it is the only publication available relating to several new data obtained by Kubovics and his co-workers. Both publications will be discussed later in the present study.

Kaba belongs to the CV3 group of carbonaceous chondrites. The designation means that the Kaba carbonaceous

* **Corresponding author: István Viczián**, Department of Mineralogy and Geology, Debrecen University, Debrecen, H-4032, Hungary, e-mail: viczianif@gmail.com

Ferenc Kristály: Department of Mineralogy and Petrology, Miskolc University, Miskolc, H-3515, Hungary

chondrite (C) belongs to the textural type 3 and the chemical type called Vigorano, according to the classification of Van Schmus and Hayes (1974). According to a further subdivision based on petrographic modal analysis, Kaba belongs to the oxidized group, its closest relatives being the meteorites Bali and Grosnaja (Table 5 in McSween 1977). CV chondrites “*are amongst the most primitive meteorites known, and they contain components formed in both the solar nebula and on the parent body*” (Howard *et al.* 2010).

The general importance of carbonaceous chondrites is that they “*are the legacy of the first accretionary stages of our protoplanetary disk*” (Trigo-Rodríguez *et al.* 2019). They have been formed in the outer protoplanetary disk, where highly porous and fragile carbon and water-rich transitional asteroids formed. The knowledge of the formation of carbonaceous chondrites can also contribute to the understanding of the delivery of volatiles to terrestrial planets.

The goal of the present study is to give a review of the studies related to hydrous alteration of the primary material, which is considered by most authors as a process that proceeded in the interior of the parent body. Góbi and Kereszturi (2017) published an improved thermodynamic model of the hydration of primary silicates, taking into account the role of the principal parameters of the process, such as the water:rock ratio, the initial temperature of the reaction, the exothermic rise of temperature, and porosity. The model discusses the simple and presumably very frequent reaction of olivine (forsterite) with water, by which serpentine is produced. The importance of these hydration reactions is that they are exothermic reactions and may contribute to the heating of the parent bodies, even when the initial temperature is extremely low (*e.g.* 200 K).

The present study is a substantially enlarged version of the review already published in the Hungarian language in the proceedings of the Kaba meteorite Symposium held in Debrecen, 2017 (Viczián 2018). In addition, the results of some recent X-ray investigations, performed on an uncovered thin section of the meteorite material, are presented.

2 Indications of hydrous alteration in the first detailed mineralogical description by Sztrókay *et al.* (1961a,b)

The first detailed mineralogical and chemical analysis was given by Sztrókay *et al.* (1961a,b) in Hungarian and English

languages. They determined 0.53% H_2O^- contents (“moisture”). Considering that later other authors identified the hydrous mineral as saponite that contains water in the interlayer space, this value may indicate several per cent saponite contents in the material.

The study of the authors, however, did not find hydrous or clay minerals by microscopic or X-ray investigations available at those times. However, they make the assumption in the discussion that probably chlorite (or other hydrated silicates) may have formed:

[...]the water vapour formed by the decomposition of hydrocarbons at medium pressure and temperature may lead to the hydration of some of the silicates. Now, the presence of chlorite (and possibly other hydrated silicates) is probable in carbonaceous chondrites[...] (Sztrókay *et al.* (1961b, p. 101)).

There is a figure only in the English version (Plate II, Figure 4) that shows a fibrous mineral described in the text in the following way: “*a granular fibrous silicate accompaniment grouped around the large porphyritic olivine grain*” (p. 63). The caption of Figure 4 says that “*The granular and fibrous accompaniment consists mostly of pyroxenes.*” In a later study, Keller and Buseck (1990) supposed that fibrous material determined by Sztrókay *et al.* (1961b) as pyroxene might be in fact saponite. The same may be the case for the glass in the matrix that is described as being altered: “*Its colour is brown, turbid [= muddy, disordered], and it contains fine crystallites in great abundance[...].*” However, “*crystallites... could not be accurately specified*” (p. 68).

3 Identification of Fe-rich saponite

Keller and Buseck (1990) identified the hydrated clay mineral saponite in the Kaba meteorite using electron microscopy. In a single Ca-Al-rich inclusion (CAI), high-Al phyllosilicates were found that resemble Na-phlogopite. The authors also refer to an earlier study by Fegley and Post (1985) who first found layer silicates in CAI (see citation in their article).

Keller and Buseck (1990) identified the principal clay mineral as Fe-bearing saponite and observed that it is relatively abundant both in the chondrules and in the matrix. It occurs most frequently in the chondrules where it derives from glass and enstatite. Between unaltered enstatite grains, saponite forms ribbons. There is a crystallographic orientation relationship between saponite and enstatite. In the matrix, saponite exhibits coarse- and fine-grained clusters, isolated packets, and sparse,

lamellar replacements of Fe-rich olivine. Most saponite derives from the fine-grained olivine of the matrix.

Considering the source of elements necessary for the formation of saponite, when the precursor phases were enstatite and glass in the outer parts of porphyritic olivine chondrules, few Na, Al, and Si might have derived from glass. In the transformation from the matrix olivine, alkali elements and Al came from glass and sparse feldspar; presumably, the process was dissolution by aqueous fluids. In this case, mass transfer was necessary from a longer distance than in the case of the chondrules. Iron liberated by the dissolution precipitated in the form of magnetite. Presumably, the formation of saponite by aqueous alteration occurred on the parent body, following accretion. The temperature did not exceed 100°C considering terrestrial saponites form in the 0 to 100°C temperature range.

4 Comparison of alteration with other CV3 chondrites

In the discussion, the Kaba meteorite was compared to other carbonaceous chondritic meteorites by Keller and Buseck (1990).

In the CV3 group, the matrix of the Mokoia meteorite similarly contains saponite. The conditions here were more oxidizing, as shown by more frequent iddingsitic rims in the fayalitic olivine of the matrix. On the other hand, in the chondrules of Mokoia, the chief alteration product is Na-phlogopite, which is supposed to be formed before accretion.

In the CO3 group, the Lancé meteorite was compared to Kaba. It contains little serpentine in the matrix, formed in more oxidizing conditions, at higher temperatures, and in the presence of less water than in Kaba.

The most intensely altered group is CI, which is a product of the further transformation of the CV3 and CO3 groups. CI meteorites contain frequent saponite, serpentine, ferrihydrite, and carbonates formed in a broader temperature range. Examples are Orgueil and Yamato 82162 meteorites.

A high number of microprobe analyses of the matrix and chondrule rims of carbonaceous chondrites were collected and evaluated by Zolensky *et al.* (1993). In the group of CV chondrites, also data from the Kaba meteorite were included. Microprobe analyses of the matrix and chondrule rim were shown in a Fe–Mg–Si triangular diagram. Matrix has much higher average Fe contents than rims

(Figure 3 in the study cited); however, the compositional variation of both varieties, the matrix and rim, is almost the same, while Mg/Si ratio always remains almost constant (Figure 2 in the study cited). Based on previous investigations by Keller and Buseck (1990), they state that the typical phyllosilicate in the matrix of Kaba is saponite. The chemical composition of saponites is also shown in the Fe–Mg–Si triangular diagram (Figure 9 in their study). In general, saponites contain much less Fe than the average composition of the matrix. Saponites in Kaba have the lowest Fe contents among carbonaceous chondrites considered in this study. Concerning stability, they observe that saponite has an extremely wide temperature stability field and forms at high water/rock ratios (mass ratio >5), as compared to serpentine. In general, the temperature range of the formation of CV chondrites is set between 50 and 150°C.

The study of Keller and Buseck (1990) on the Kaba meteorite was among the first that determined phyllosilicates in chondrites in detail. The method applied in this period of investigations was always transmission electron microscope (TEM) and microprobe analysis. The application of X-ray diffraction for the study of chondrite composition started only in the years after 2000. Assisted by the position-sensitive detector (PSD), XRD modal analyses were carried out by Howard *et al.* (2010) on six CV3-type carbonaceous chondrites, including Kaba. This way, we know the bulk modal composition of the meteorite. According to their determination, phyllosilicates are present only in oxidized chondrites, but contents are relatively low in Kaba: 3.5%, a number very similar to other oxidized CV3 chondrites; Grosnaja: 4.2%; Mokoia: 3.7%; and Allende: 1.9%. In reduced CV3 chondrites, no phyllosilicate was found. Howard *et al.* (2010) came to the following conclusion: “*That the reduced CVs have escaped aqueous alteration, and the generally low abundance of phyllosilicate in oxidized CV samples, indicates a short-lived availability/low abundance of fluid and restricted flow on the CV parent body where only samples at the ice:rock interface may have experienced partial hydration before the supply of fluid was exhausted.*”

Gyollai *et al.* (2011) studied the alteration of chondrules in the Kaba meteorite by optical microscopy in transmitted and reflected light. They distinguished five steps of increasing transformation of primary silicates to phyllosilicates. The same alteration can also be observed in the phenocrysts of the groundmass. Kaba was compared to the Yamato-86751 meteorite, which is equally an oxidized CV chondrite. In the latter one, chondrules are much less altered and the matrix is much more altered than those in Kaba.

5 Experimental alteration

Experimentally produced alteration of Allende-type meteorites, including the transformation of forsterite to fayalite and the formation of saponite, was observed by Tomeoka and Kojima (1995). A small piece of Allende meteorite was exposed to hydrothermal conditions. The most important alteration product obtained in both the matrix and chondrules was saponite. Its texture was similar to that observed in other carbonaceous chondrites, including Kaba, as it was described previously by Keller and Buseck (1990). In addition to saponite, an Al-rich phyllosilicate was formed of glass, preferentially in the mesostasis of chondrules. The formation of saponite, however, was preceded by the alteration of forsterite (Mg-olivine), mostly in the outer side of chondrules and aggregates. Here, the outer rim of individual crystals became enriched in Fe, added from the matrix. Surprisingly, small forsterite crystals occurring in the matrix remained unchanged. Fe was transported from the matrix to the chondrules, and reversely, chondrules gave over the elements Ca, Mg, Al, Na, and S to the matrix. As a whole, the meteorite became more homogenized upon the hydrothermal alteration.

Experimental conditions were 450°C and 800 bar. Tomeoka and Kojima (1995) conclude that the alteration in other meteorites like Kaba and Mokoia may have proceeded in a similar hydrothermal way; however, in the natural conditions, the temperature and pressure of the fluid were much lower than in the experiments.

Contrary to the Allende meteorite altered experimentally, the real composition of the Allende meteorite is essentially completely anhydrous, except for rare hydrous phases, like clintonite, $\text{CaAlMg}_2(\text{SiAl}_3\text{O}_{10})(\text{OH})_2$, and margarite, $\text{CaAl}_2(\text{Al}_2\text{Si}_2\text{O}_{10})(\text{OH})_2$, both members of the brittle mica group (Keller and Buseck 1991), that are restricted to chondrules and CAIs. The important alteration processes are the formation of fayalitic outer rims around forsterite and strong Na mobilization resulting in nepheline and sodalite. Krot *et al.* (1998) supposed that both Allende- and Bali-type oxidized chondrites were derived from the same asteroid, of similar original composition, only their degree of thermal alteration is different. Allende represents the higher degree of alteration; consequently, Allende type is derived from Bali type. Kaba belongs to the Bali type and represents a lower alteration temperature range than Allende.

6 Other hydrothermal effects

Kimura and Ikeda (1998) distinguished two types of alteration in CV3 chondrites, hydrous and anhydrous.

They identified saponite and Na-phlogopite as magnesian phyllosilicates in Kaba using electron probe microanalysis and backscattered electron images. Moreover, they observed bronzite in chondrules from Kaba. They considered all these minerals to be products of secondary hydrous alteration. Their systematic work found that the chondrules in the Kaba meteorite went through an extensive hydrous alteration, while anhydrous alteration was much weaker and was limited to the formation of zoning in olivine crystals. They concluded that anhydrous alteration in Kaba predated hydrous alteration and took place before the accretion of chondrules into the CV parent asteroid.

Krot *et al.* (1998) carried out a thermodynamic analysis of hydrous alteration processes. They divided the group of oxidized CV3 chondrites into two subgroups, Bali-type and Allende-type oxidized chondrites. The Kaba meteorite belongs to the Bali-type subgroup, characterized by the following hydrothermal alterations:

- the formation of saponite and Na-phlogopite,
- the formation of magnetite by the reaction of metallic iron and water, and
- the formation of fayalite and Ca-Fe-pyroxenes from the reaction of magnetite and silica.

It could be proved that all these minerals may form from aqueous solutions below 300°C. Temperature of the formation of fayalite and Ca-Fe-pyroxenes may be lower than 100 or 200°C, depending on the activity of SiO_2 . The activity of SiO_2 is supposed to be in the range of 10^{-5} – 10^{-4} . According to Keller and Buseck (1990), also saponite forms below 100°C. In the case of saponite, the activity of SiO_2 is even higher, around 10^{-3} . The formation of all three silicates requires the high activity of Fe.

Among primary silicate phases, especially glass of anorthitic composition in the mesostasis of chondrules and Fe-Mg pyroxenes underwent alteration. When comparing the primary composition with the composition of layer silicates formed, the strongest dissolution affected Ca but also some of Si, Al, Mg, Fe, Mn, and Na were dissolved. Products of alteration are saponite, fayalite, and Ca-Fe-pyroxenes. They seem to be coexistent, and saponite did not crystallize at the expense of the other two silicates. In Bali-type meteorites, similarly as in Kaba, fayalite forms in the matrix and in the magnetite nodules of chondrules. Porphyritic forsterite crystals in the chondrules, however, did not transform completely to fayalite or do not have fayalitic rims, in contrast to the Allende-type meteorites.

The opinion on the secondary formation of fayalite in chondrules of Kaba and Mokoia meteorites was later modified by *in situ* oxygen isotopic measurements (Choi

et al. 2000). It seems that fayalite formation from magnetite does not necessarily involve water. They found that $\delta^{18}\text{O}$ values in these two phases in the chondrules of Kaba are not different. It indicates that in Kaba, fayalite is formed by the replacement of magnetite in a dry environment, with the O coming entirely from the precursor magnetite and silica. In contrast, in chondrules of Mokoia, magnetite, and fayalite seem to have formed simultaneously, in isotopic equilibrium in a water-rich environment at low temperatures, ~ 300 K.

Further *in situ* oxygen isotopic analysis contributed to the detailed characterization of fluid–mineral interaction in chondrules (Krot *et al.* 2018), CAIs, and amoeboid olivine aggregates (AOAs) (Krot *et al.* 2019). It was concluded that plagioclase and plagioclase mesostasis in Kaba chondrules experienced nearly complete oxygen isotope exchange with the fluid. Anorthite and possibly melilite in CAIs and AOAs from Kaba experienced oxygen isotope exchange with the same fluid that affected Kaba chondrules. The hydrous alteration occurred on the CV parent asteroid. These data are useful to constrain oxygen isotopic relations of an aqueous fluid on the CV parent asteroid.

Gyollai *et al.* (2018) studied hydrous alteration in chondrules and AOAs by optical, electron optical, and fourier transform infrared spectroscopy (FTIR) methods. They agreed with Krot *et al.* (2018, 2019) that water was necessary for these alterations. They distinguished two steps, first high-temperature hydrothermal alteration, and second aqueous alteration. Both alteration processes affected the interstitial area and fractures of olivine. High-temperature hydrothermal alteration produced Ca-Al-bearing minerals: hedenbergite, melilite, and dmisteinbergite – a high-temperature polymorph of anorthite, mainly in the interstitial area of AOAs. Low-temperature aqueous alteration first produced iron metasomatism on the rims of forsteritic olivine and later veins across AOAs and chondrules containing troilite or iron oxides (including ferrihydrite, goethite, and hematite). Phyllosilicates were not found in this study. Temperature ranges are estimated for low-temperature alteration, for the metasomatic iron alteration: 200–250°C, for ferrihydrite formation: <25°C, and for goethite and hematite: 50–75°C. All processes are supposed to have taken place in the parent body; however, the secondary terrestrial formation of goethite is not excluded. As for the formation of ferrihydrite in the parent body, however, one has to consider that the persistence of such an unstable mineral in the interior of a meteorite, over billions of years, is highly improbable. If there is this mineral indeed, its secondary, terrestrial formation would be much more probable, similarly as in the case of goethite.

7 Cathodoluminescent studies of alteration of forsterite

According to the electron probe microanalysis and cathodoluminescent analysis of Gucsik *et al.* (2013), redistribution of elements, primarily Fe, occurs in the forsterite crystals of chondrules in the Kaba meteorite. The core of Mg-rich forsterite is enriched in Ca, Al, and Ti, whereas the rim of Mg-rich forsterite is enriched in Mn, Cr, and Fe. However, the Fe-enriched rim cannot be yet called fayalite. The zoning of forsterite crystals can be well demonstrated in the cathodoluminescence pictures scanning electron microscopy-cathodoluminescence made by the same authors. The core of the crystals emits strong blue and weak red-infrared (IR) cathodoluminescence emission, resulting in a lightly violet colour. The rims are bright red. The emission of blue CL radiation is due to Ca, Ti, and Al substitution, and red-IR emission is due to Cr and red emission mostly to Mn substitution. The blue emission is due to Al–O–Al defect centres and indicates relatively low-temperature (at around 250°C) hydrothermal processes in the Kaba meteorite because these centres would disappear at higher temperatures.

Contrary to the opinion of Kimura and Ikeda (1998), the observed zoning in forsterite is explained by Gucsik *et al.* (2013) also by hydrous processes, using the hydrothermal alteration theory developed by Krot *et al.* (1998). Hydrothermal solutions may have partly dissolved the elements Ca, Al, and Ti from the rims, and simultaneously deposited elements Fe, Mn, and Cr in their place. Presumably, no real chemical reaction, only diffusion of elements in the rims of forsterite crystals, has taken place, upon the effect of hydrothermal solutions. The structure of the forsterite crystals remained ordered. The formation of other reaction products, *e.g.* saponite, was not observed.

There are opinions that doubt that the formation of fayalite, ferrous olivine, and magnetite is related to hydrous thermal metamorphic processes. Based on their XRD modal analyses of CV3 chondrites, Howard *et al.* (2010) observed that there is no significant connection between contents of these minerals and the presence of phyllosilicates that are equally products of hydrous alteration.

8 Oriented texture

One of the properties attributed to hydrous processes is oriented texture.

Bérczi *et al.* (1998) made backscattered electron photographs of small portions of a 4 cm² thin section and

composed of these photographs a single picture of the whole thin section. In this composite picture, foliated structure could be observed both in the matrix and in the arrangement of elongated, larger, inclusion-like objects. Similar foliation has been observed earlier also in other carbonaceous chondrites, Leoville and Allende. At higher resolution, layered texture can be observed also in the inner structure of chondrules, spinel-rich inclusions, and opaque assemblages. This is a common feature of the CV chondrites, Allende and Mokoia. Foliation is called in the study as “a kind of layered or laminated textural map.” In the summary, the authors mean that foliated textures in CV chondrites indicate that these meteorites may have derived from the outer shell of the parent body. It seems that sedimentary origin is supposed but is not expressively formulated.

Kubovics *et al.* (1998) reported on the same subject. In both publications, the unequilibrated character of the foliated texture is stressed. The expression “stratification” also occurs in the text that also implies some kind of sedimentary process. Based on these observations, Lukács and Bérczi (1999) concluded that possibly “*sedimentary processes touched the surface layers of the CV3 parent body.*” A different kind of stratification is described by Grossman *et al.* (2000) when chondrules “sedimented” from dust in the interplanetary space: “*Chondrites are sedimentary rocks formed from primitive, nebular materials that accumulated into small planetesimals and asteroids in the earliest stages of the solar system’s formation.*”

Chondrules with zonal internal structure were already observed by Sztrókay *et al.* (1961b): “*There are characteristic chondrules consisting of olivine and bronzite, the two ingredients occurring in a zonal arrangement, the centre consisting of olivine, the surrounding zone of bronzite*” (p. 65). However, it is not sure whether this type of zonal arrangement can be attributed to hydrous effects.

9 Organic matter

Sztrókay *et al.* (1961a,b) considered that organic matter in Kaba is hydrocarbon. Its amount was specially determined along with the normal silicate analysis and expressed in the form CO₂ and H₂O. Elemental carbon content C is 1.99%. The computed total hydrocarbon content is 4.03%. By microscopic observations, they stated that “*the ground mass is impregnated in a cloud-like pattern with dark brown aliphatic hydrocarbons..., which are more translucent here and darker there. Hydrocarbons are especially dense around the chondrules, condensed into an opaque cover. However,*

there are impregnations in the form of spots also in the interior of looser or vitreous-crystalline chondrules” (p. 63).

Sztrókay *et al.* (1961a,b) made X-ray and IR analysis on the organic matter using powdered bulk samples. X-Ray diffraction analysis was carried out by the Debye–Scherrer method. Organic matter was enriched in the residue of dissolution in hydrochloric acid. In the X-ray pattern of this fraction, around the direct beam, a wide ring could be observed, and also, a halo covered the entire pattern. IR spectroscopic analysis of the same insoluble residue has shown aliphatic hydrocarbons of higher molecular weight. Another result of IR analysis was that S bound to higher homologues of hydrocarbons was detected. On the other hand, no trace of halogen was found in the Kaba meteorite.

In a broader context, Bonal *et al.* (2006) studied the transformation of organic matter in CV3 chondrites, including Kaba, by Raman spectroscopy. In each sample, a so-called G band can be detected (G for graphite). The presence of G band indicates polyaromatic carbonaceous material. It can be considered together with the so-called D band (D for disorder). D band is not present in a perfectly stacked graphite and is induced by structural defects in the polyaromatic material. The exact location, intensity, and intensity ratio of these bands are parameters that indicate the degree of thermal metamorphism. All parameters show that Kaba belongs to the less metamorphosed group of CV3 chondrites, along with the Vigorano and Efremovka meteorites. Even in this group, Kaba is the least metamorphosed.

Bonal *et al.* (2006) compared the parameters obtained for meteorites to the values determined in terrestrial coals for which vitrinite reflection data were known. The results are comparable only with limitations because the original organic matter has been different. However, metamorphic degree of meteorites is comparable to high-rank terrestrial coals (anthracite and meta-anthracite).

In CV3 meteorites, thermal metamorphism and subsequent aqueous alteration proceeded within the asteroid; however, they are independent processes. Metamorphism affected also the (Fe and Mg) chemical zonal structure of outer rims of olivine crystals, but this zoning depends also on the intensity of later hydrous alteration.

A high number of special organic compounds were identified in Hungary, on the initiative of Kubovics. We know about conference lectures on the subject (listed in the Hungarian version of the present review, Viczián 2018); however, no written publication is available. Results are only summarized in the chapter on meteorites of the textbook “General Petrology” by Kubovics (2008, p. 103, in Hungarian): In Kaba meteorite... “*matrix contains... hydrocarbons of various composition, aliphatic and aromatic, saturated and unsaturated organic compounds, including amino*

acids like glycine, alanine, glutamic acid, valine, proline, similarly to the Murchison and Mokoia (etc.) meteorites, as well as elemental sulphur.” All these compounds are considered by Kubovics to be formed by abiogenic processes.

Valuable analytical data concerning organic matter in the Kaba meteorite were published by Polgári *et al.* (2018). The study was shortly referred to by Polgári *et al.* (2021). A 3 mm-sized chondrule was analysed. The chondrule centre contains olivine, pyroxene, troilite, and ferrihydrite with aliphatic hydrocarbons. The area near the rim of the chondrule contains more dark, organic material. Organic compounds were identified in microscope combined by attenuated total reflectance fourier transform infrared spectroscopy. Aliphatic hydrocarbons, polycyclic aromatic hydrocarbons, and vibrations indicating (C–N/C–H), COOH, and CO bonds were identified. C_{tot} content is 1.36 ± 0.08 wt%. Stable C isotope is light, $\delta^{13}\text{C}$: -15.7 ± 0.69 ‰. The study presents a complex verifying interpretation, which include high-resolution, high-magnification (1,000×) textural description of samples by optical rock microscopy, on different types of chondrites among them, those of Kaba. These textural features show strong similarities to terrestrial mineralized microbially mediated biosignatures. Furthermore, the *in situ* FTIR and Raman spectroscopy resulted in the determination of so-called bioindicator minerals like ferrihydrite and magnetite. The authors assume that these organic materials and inorganic minerals are indicating that biologic activity proceeded already in the parent body. One has to remark, however, that the observations and argumentation of this work go beyond the current knowledge and further work would be necessary to establish the possibility of biogenic activity in meteorite parent bodies. In particular, like in the case of the results of Gyollai *et al.* (2018), also the question of the long-term persistence of the unstable phases like ferrihydrite remains to be solved.

10 XRD analysis

10.1 Previous X-ray studies

Systematic application of X-ray diffraction for the study of the modal composition of meteorites started only in the years after 2000. Bland *et al.* (2004) introduced a new method based on the combination of XRD and Mössbauer spectroscopy. The X-ray equipment used had a curved position-sensitive detector (XRD-PSD). It is able to analyse very little amounts of material, which is an important aspect of the analysis of valuable meteorite materials.

This first publication on the new method dealt exactly with carbonaceous chondrites. Unfortunately, Kaba was not among the studied samples. However, these data are interesting for comparison with the Kaba results. In three of the studied four meteorites, phyllosilicates were found by XRD; however, their more specific identification was taken over from previous HRTEM and microprobe studies (cited in the study). The following data were presented:

Allende (CV type): no phyllosilicate was detected in the present study, but minor amounts of saponite were found earlier by electron microscopic methods (Zolensky *et al.* 1993).

Murchison (CM type): serpentine (22.8%) and cronstedtite/tochilinite (58.5%). The latter phase is a regular intergrowth of an iron serpentine layer and a Fe^{2+} Mg–sulphide–hydroxide layer, resulting in a basal layer distance of 17.8 Å.

Tagish Lake (C2 ungrouped): interstratified saponite/serpentine clay (60.3%). By TEM, various basal layer distances were found, and the most frequent one was that of saponite. Saponite was identified by 10–12 Å layer lattice fringes, and serpentine was identified by 7 Å layer lattice fringes. In addition, regular intergrowths with 14 Å basal spacing were observed, interpreted as chlorite/serpentine interstratifications.

Orgueil (CI type): serpentine (7.3%) and saponite/serpentine (64.2%). In the matrix, serpentine (7 Å layers) and saponite (10–11 Å layers) are coherently intergrown in disordered sequences.

Another work applied the same integrated use of Mössbauer spectroscopy and X-ray diffraction to quantify the modal mineralogy of a high number of *ordinary chondrites* (Menzies *et al.* 2005). They found the most primitive unequilibrated ordinary chondrites to contain a significant amount of phyllosilicate material (up to 20%) that was converted during metamorphism to form ferromagnesian silicates. Unfortunately, the disordered and fine-grained nature of the material and the method applied did not permit specific determination of the phyllosilicates. Quantitative determination was made only using non-basal reflections. They found weak and diffuse basal reflections at 4° and 8.8° $2\theta\text{CuK}\alpha_1$ (22 and 10.0 Å, respectively) only in the most equilibrated chondrites studied. They think that it is “almost certainly a mixed layer clay containing smectite and illite.”

Howard *et al.* (2015) reviewed CR and CM meteorites from the point of view of phyllosilicate contents determined by the XRD analysis. Previously, X-ray properties of phyllosilicates in CM2 meteorites were specially studied by Howard *et al.* (2009). Two forms of serpentine were distinguished: well-crystalline cronstedtite with sharp peaks at 12° and 25° 2θ and disordered fine-grained “FeMg–serpentine” with

diffuse hk peaks at 19° and $61^\circ 2\theta$, and no appreciable basal reflections in the low-angle range. There is no cronstedtite, only a little disordered “FeMg–serpentine” and an amorphous Fe-rich phase in the CR type, while in the CM type both cronstedtite and “FeMg–serpentine” are abundant, and amorphous phase is absent. The characteristic peaks of the two serpentine varieties are well illustrated on the annotated XRD patterns in Figure 1 of Howard *et al.* (2009).

CI-type meteorites contain the highest amounts (70–80%) of phyllosilicates among carbonaceous meteorites (King *et al.* 2015). On the XRD-PSD pattern, relatively broad diffraction peaks at $\sim 19^\circ$, $\sim 34^\circ$, and $\sim 60^\circ 2\theta$ (more precisely at $\sim 20^\circ$, $\sim 35^\circ$, and $\sim 60^\circ 2\theta$, see Figure 1 of the study cited) are attributed to fine-grained, poorly crystalline phyllosilicates, in which CI chondrites largely consist of intimately mixed saponite and Fe-bearing, Mg-rich serpentine. In fact, the basal reflections of serpentine at $\sim 12^\circ$ and $\sim 25^\circ 2\theta$ are well visible; however, there is no reflection of saponite at lower angles. Due to the intergrown nature of the serpentine and saponite, the authors did not attempt to deconvolute the two phases and instead reported only the total phyllosilicate abundance.

Finally, the Kaba meteorite was studied in the group of the CV3-type chondrites (Howard *et al.* 2010). Because of the low amounts of phyllosilicates in this group (<5%), their disordered structure and very fine particle size, and also because the XRD method applied does not permit exact determination of basal reflections in the low-angle range, they did not attempt to determine the type of phyllosilicate. In the quantitative determination, they considered only the broad hk reflection at $\sim 19^\circ 2\theta$. These platy crystals consist only of just a few structural layers along [001]. These layers are too thin to diffract X-rays coherently enough to produce 00l reflections.

Considering these data, carbonaceous chondrites display a wide range of possible basal reflections of varying position and sharpness. In the group of CV3-type carbonaceous chondrites to which Kaba belongs, however, no serpentine-like 7 \AA minerals, only saponite and its interstratifications with similar three-layer phyllosilicates were found so far. According to the chemical analyses, the interlayer cation of saponite is most likely Na.

10.2 Material and methods of the present study

In 2017, in the Department of Mineralogy and Petrology, Miskolc University, XRD investigations were carried out on a relatively large uncovered thin section provided to us by Arnold Gucsik. The thin section was prepared

from a chip of the Kaba meteorite weighing approximately 1 g, allocated from the Reformed College of Debrecen, where the Kaba meteorite is kept. The chip was mounted in a three-component non-radiative epoxy material at Okayama University of Science (Okayama, Japan). Silicon colloidal liquid was used for the polishing of that sample to avoid any metallic or even carbonaceous contaminations. Some details of the sample preparation as well as data acquisition can be found in Gucsik *et al.* (2013).

The diffractometer was of type Bruker D8 Advance, with Cu-K α radiation and parallel beam geometry. It was attempted to identify the principal components and to demonstrate the presence of clay minerals. First, a general view of the thin section was taken by irradiating various quadrangles of 12×8 and 12×4 mm size. In the quadrangles, both chondrules and matrix are present. In a portion of the thin section, dominated by the matrix (Figure 1), clinoenstatite, diopside, and also pyrite were found, in addition to magnetite and forsterite (olivine). Some peaks in the range $5\text{--}7 \text{ \AA}$ could not be identified.

Another X-ray pattern is shown in Figure 2. Here, the background is subtracted. Portions of the whole diagram are enlarged in order to identify the components in detail. Members of olivine series, a nearly pure forsterite, fayalite, and a transitional olivine composition, can be distinguished using the enlarged 111 and 131 reflections. Other phases are enstatite and members of the magnetite–magnesioferrite series.

In both diagrams, a weak but clearly discernible reflection appears at about $4.5\text{--}5.0^\circ 2\theta$, corresponding to $18\text{--}19 \text{ \AA}$. This reflection may be considered as basal reflection of some smectite-like clay mineral, and its possible identification will be discussed later.

In order to study clay minerals in the matrix in more detail, enlarged portions of the matrix were studied. Figure 3 shows the micrograph of an altered grain of about $200 \mu\text{m}$ longitudinal size in the matrix, which is composed of yellowish material, most probably a clay mineral coloured by infiltrated organic matter. In the X-ray pattern (Figure 4), four very broad reflections appear in the low-angle range.

10.3 Possible interpretations

The exact interpretation of eventual mixed-layering is not clear and would require various additional treatments. Here, only some considerations can be made.

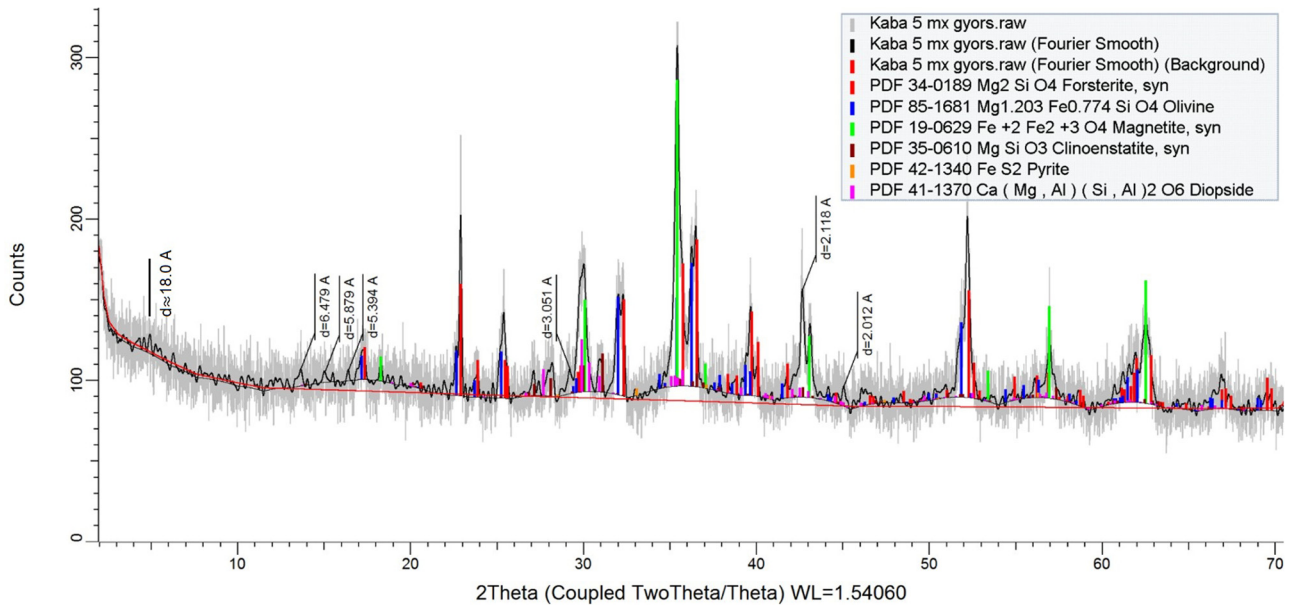


Figure 1: XRD pattern of a portion of an uncovered thin section including chiefly matrix, Kaba meteorite. Broad reflection in the 23–18 Å range is probably a basal reflection of a smectite-like clay mineral.

The reflections on Figure 4 may be attributed to basal reflections of discrete and mixed-layer clay minerals composed of the components Na-smectite (~ 12 Å) and “illite” (=dehydrated, 10 Å-smectite), and probably also extremely hydrated smectite or smectite–organic complex (~ 18 Å). Tentatively, the reflection at 22.0 Å may correspond to the 001 basal reflection of a disordered, partly regular, rectorite-like structure. The

presence of serpentine-like phases is not probable because no 7 Å layer distances were observed on the HRTEM images.

One may suppose that the 10 Å component is dehydrated smectite. Keller and Buseck (1990, their Fig. 5) observed by HRTEM clearly 1 nm = 10 Å distances between the basal layers of Fe-rich saponite in the Kaba meteorite. Small size of coherently scattering units, frequent phase

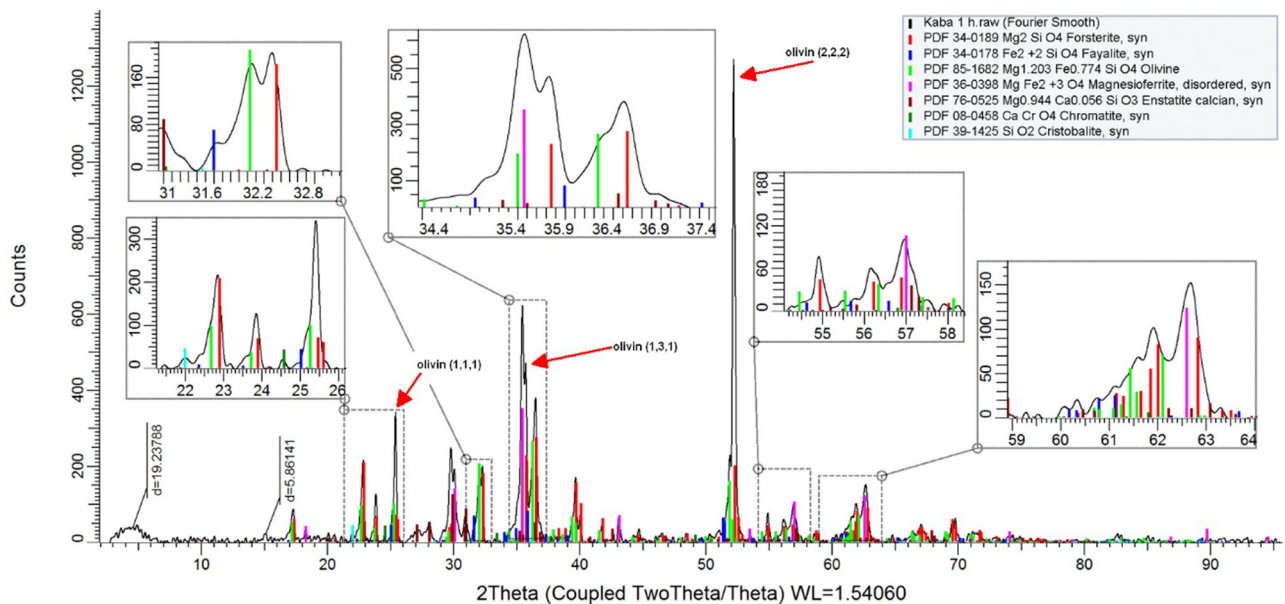


Figure 2: XRD pattern of a portion of an uncovered thin section including matrix and chondrules, Kaba meteorite. Enlarged parts of the diagram show detailed identification of olivine composition. Reflection at 19.2 Å is probably a basal reflection of a smectite-like clay mineral.

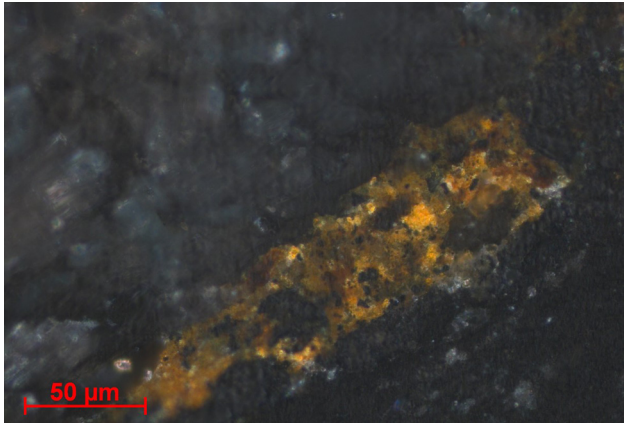


Figure 3: Micrograph of a particle in the matrix containing devitrified glass and organic matter.

boundaries between the units that may be understood as fundamental particles, bending of individual sheets, and a generally disordered stacking sequence of the smectite layers are also visible on the HRTEM pictures. In this case, in electron microscopic analysis, collapse of basal distances to 10 Å is supposed to be due to the vacuum conditions. Moreover, the assumption is not excluded that part of the silicate sheets had the distance of 10 Å already in the natural state.

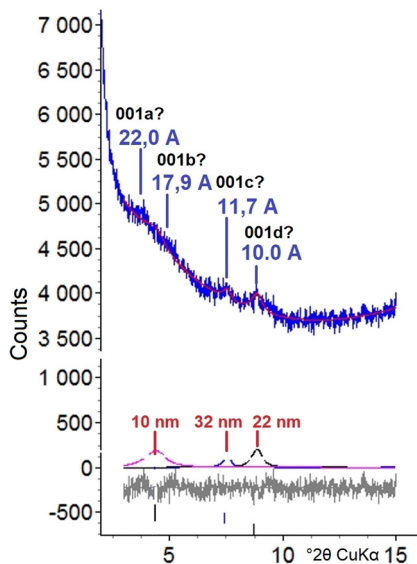


Figure 4: XRD pattern of the particle shown in Figure 3. Numerical data: tentative indexes of basal reflections. d values in Å units. “a” phase: regularly mixed-layer Na-saponite/10 Å-saponite (?), “b” phase: extremely hydrated saponite or saponite–organic complex, or irregular mixed-layering of “a” and “c” phases (?), “c” phase: Na-saponite (?), “d” phase: dehydrated, 10 Å-saponite (?). Below: particle size computed from line broadening.

High interlayer distances at about 18 Å or more imply the explanation assuming regular interstratification of smectite-like layers with another component layer. In terrestrial conditions such regular mixed-layering, e.g. rectorite, corrensite, is known to form at elevated temperatures, while the corresponding irregularly mixed-layer phases are normally formed at lower, Earth surface temperatures. In the Kaba meteorite, which suffered some moderate thermal effects, similar ordering is not excluded.

Another explanation of the high distances would be a complex of smectite with organic matter. Considering terrestrial analogues, natural interlayer organic complexes of montmorillonite minerals are relatively rare in soils, it seems, however, that they are more common in muddy sediments and mudstones (Lu *et al.* 1999). In naturally occurring montmorillonitic soils, similar basal reflections at 24 Å (001) and 12 Å (002) were reported by Theng *et al.* (1986). They interpreted it as regular interstratification of illite and montmorillonite layers where organic matter is stored in the interlayer space of the expandable component. In most cases, however, organic matter is adsorbed preferably on the external surface or on the layer edges of clay minerals (Perez Rodriguez *et al.* 1977) and similar high basal spacings can be achieved only by special additional treatments. In general, in terrestrial natural clay–organic complexes, size of the organic molecules in the interlayer space does not exceed the size of the interlayer distance of normal montmorillonites with two layers of water and interlayer cations. When the measured distance between the silicate layers is about 4 Å, the organic or inorganic interlayer phases cannot be distinguished in the XRD patterns of untreated, natural samples. Special thermal or chemical treatments would be necessary to demonstrate the existence of organic matter in the interlayer space (Du *et al.* 2019). However, in view of the common occurrence of smectite-like phyllosilicates and organic matter, this way of interpretation is worth of further considerations.

11 Conclusions

Reviewing a number of relevant studies on Kaba CV3 chondrite, the following conclusions can be drawn.

Low-temperature secondary crystalline phases and some sort of foliated texture indicate that the Kaba meteorite is derived from a *near-surface portion* of the parent body.

Organic matter consists mostly of aliphatic and aromatic hydrocarbons. The Kaba meteorite also contains a number of relatively complex amino acids. However, they

are considered by most authors to be of abiogenic origin. On the contrary, based on the complex evaluation of textural features and of some inorganic minerals, other authors supposed biologic activity already in the parent body.

Thermal metamorphism at relatively low temperatures gave rise to the transformation of the organic matter to a degree comparable with terrestrial anthracites and meta-anthracites. The degree of transformation in the Kaba meteorite is the lowest among CV3 chondrites studied.

The subsequent *hydrothermal alteration* of primary crystalline phases and glass produced magnetite in the temperature range 100–300°C and newly formed coexistent Fe-rich saponite, fayalite and Ca–Fe-pyroxenes at low temperatures, possibly below 100°C. Hydrous solutions also caused Mg–Fe zonation in olivine. As determined by quantitative XRD, modal contents of saponite in oxidized CV3 chondrites are moderate, in Kaba: 3.5%.

The analytical observations seem to be in accordance with the general tendencies predicted by the thermodynamic model of serpentinization (Góbi and Kereszturi 2017), although the main hydrated mineral in Kaba is saponite, which is a smectite variety and not serpentine. One of the main parameters of the process is the water:olivine ratio, which is in the CV group, to which Kaba belongs, the highest among carbonaceous chondrites. As shown in Figure 1 of the study cited, the initial temperature needed for hydration (T_{init} in K) falls into the range of liquid water (273–373 K), although the model extends also to the realm of water ice. As compared with other meteorite types, the effects of hydration on the initial composition and conditions are relatively slight: the rise in temperature due to reaction (ΔT in K) is probably less than 50 K, and the resulting change in porosity ($\Delta\eta$) is less than 0.5%.

In relation to the organic matter, the authors of the serpentinization model point out, that by hydration, “*new minerals are formed including phyllosilicates (e.g. serpentine) that are able to catalyse several chemical processes on their surface including the polymerization of prebiotic molecules.*” This statement is important considering the presence of complex organic molecules and the possibility of smectite–organic complexes in the Kaba meteorite, considering that the catalytic effect of smectites is even higher than that of serpentines. On the other hand, one has to consider that the chief transformation of the organic matter proceeded during a thermal event that happened earlier than the formation of smectite.

By *new XRD analysis* of an uncovered thin section, possibly mixed-layer phases of different smectite-like layers could be identified by the present authors. In the future, more exact determination of the layer sequence may indicate details of the conditions of hydrous alteration

and thermal effects. Another promising direction of study would be to prove the existence of natural clay–organic complexes in carbonaceous chondrites, where the chief alteration product is smectite.

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