ZONED Ca–Al-RICH CHONDRULE IN BARI: NEW EVIDENCE AGAINST THE PRIMORDIAL CONDENSATION MODEL

GERO KURAT, GEORG HOINKES
Naturhistorisches Museum Wien, Vienna (Austria)

and

KURT FREDRIKSSON
Max-Planck-Institut für Chemie, Abteilung Kosmochemie, Mainz (Federal Republic of Germany)

Revised version received April 8, 1975

One large (8.5 mm) chondrule in the Barili carbonaceous chondrite is strongly enriched in refractory elements similarly to the white objects in Allanda which are widely believed to be "primitive" condensates. However, detailed investigations show the Barili chondrule to have an element distribution opposite to that predicted by the "primitive condensation" models. Refractory elements like Ti and Al are enriched at or near the surface of the chondrule. The contents of some volatile elements, e.g. Na, Si, and Cr, are higher in the central portion. It is suggested that this chondrule – and similar objects in other C-chondrites – originated in one, or more likely several, impact events and that the element distribution is the result of volatilization–condensation.

1. Introduction

Ca–Al-rich inclusions and chondrules have recently been described from various Type-III carbonaceous chondrites [1–10]. Many of these inclusions are highly enriched in Ca, Al, Ti, Zr, Y, REE, Ir, Sc, and other elements as compared to chondritic composition [10–15]. Enrichments of both the major and trace elements are clearly related to the vapor pressures of the respective element, oxide or compound [10,11]. Also, the large enrichment factors of some elements as well as the apparent simultaneous enrichment of siderophile, e.g. Ir [13–15], and lithophile elements, e.g. Ca, Al, Ti, Sc, Zr, Y, REE, seem to exclude an origin of these inclusions by magmatic differentiation processes. Since the chemistry is obviously governed by gas–liquid and/or gas–solid differentiation processes only two models for the genesis of these materials have been suggested so far: enrichment of refractory elements by partial evaporation or by partial condensation. Theoretical calculations assuming thermal equilibrium show that the early condensates from a gas of solar composition should be free of Fe and depleted in Si [16,17], with perovskite, spinel and anorthite as early phases. Since these are some of the phases observed in Ca–Al-rich inclusions, it has been widely believed that the Ca–Al-rich material represents early condensates from the primitive solar nebula (i.e., [9,11,17]). As pointed out before [10] this requires a rather complicated sequence; the Ca–Al-rich material has to condense, be removed from the system (to prevent further condensation) and be brought back and mixed with the products of the condensation of the main constituents, i.e. the bulk of the meteorites in which they occur. Furthermore, the products of a

1 Present address: Institut für Mineralogie und Petrographie, Universität Innsbruck, A-6020 Innsbruck, Austria.
2 Permanent address: Division of Meteorites, Smithsonian Institution, Washington, D.C. 20560, U.S.A.
condensation from a gas of solar composition can only be solids [16,17] because the condensation temperatures of all the phases involved are far below the solidi of these phases and the respective systems. Petrological investigations of Ca–Al-rich materials, however, unambiguously point to a liquid origin (i.e., [8,10,18]). This is in contradistinction with the condensation models and supports the partial evaporation–condensation hypotheses [10,19,20] for the origin of the Ca–Al-rich inclusions as they occur now.

In the course of a study of the Bali carbonaceous chondrite we found a large rounded Ca–Al-rich particle, apparently a chondrule. We present here the results of a detailed petrologic investigation of this particular chondrule because it bears some new evidence for a non-condensation genesis.

The study is based on microscopic investigations and electron-microprobe analyses which were performed with an ARL EMX-SM and an ARL SEMQ at an acceleration potential of 15 kV with a sample current of 1–2 \( \times 10^{-9} \) A. Wet chemically analyzed minerals were used as standards. The measured X-ray intensities were corrected according to the procedure of Bence and Albee [21].

2. Description

The originally egg-shaped chondrule had a large diameter of 8.5 mm (Fig. 1). It is rather coarse-grained (0.5–2 mm) and consists of approximately equal amounts of melilitite and augite with some interstitial anorthite. All phases present incorporate varying amounts of small spinel crystals which range in size from about 1 \( \mu \)m to 50 \( \mu \)m. All together this chondrule is very similar to a chondrule described by Clarke et al. [22] from the Allende chondrite. There is, however, a difference in the distribution of minerals within the chondrule. The Bali chondrule has a rim of on average 1.5 mm width which consists predominantly of melilitite and scattered augite. The latter being present either as individual prismatic

![Fig. 1. Ca–Al-rich chondrule from the Bali carbonaceous chondrite. Melilitite and augite form large crystals (up to 2 mm) and are indistinguishable in this photograph. Anorthite occurs as irregularly shaped interstitial grains within the main central portion. The mottled appearance of most of the crystals is due to tiny spinel crystals. Largest visible diameter of the chondrule is 7.5 mm. Plain transmitted light.](image-url)
crystals or as interstitial grains in between the melilitic grains. Both phases bear highly variable amounts of small spinel crystals. This surface zone is readily visible under crossed polarizers because the melilitic shows anomalous (blue) interference colors. This behavior of the melilitic changes gradually towards the inner portion of the chondrule. There also anorthite appears occasionally and fills some of the interstices between the melilitic and/or augites. The augites too have a different optical appearance within the surface zone and the central portion. Near the surface they are pleochroic from light green to colorless whereas in the central portion they are non-pleochroic. These optical changes are due to changes in chemical composition as shown in Table 1 and discussed below.

The melilitic at the surface is rich in the gehlenite (Ca$_2$Al$_2$SiO$_7$) component. The feldspathic component (Ca$_2$MgSi$_2$O$_7$, i.e. Mg content) of the melilitic strongly increases towards the central portion of the chondrule. Positively correlated with this MgO increase are the alkali contents whereas the Al content decreases rapidly from the surface to the center. The compositional variation of melilitic contents found here lies within the range of melilitic compositions reported from Allende inclusions [1-7,9,22]. It is noteworthy, however, that the feldspathic contents of both the Bali and Allende melilitic are clearly higher than in melilitic from Ca-Al-rich objects in Lancé [10,18]. The augites are typical aluminous titanagulites similar to those already described from Ca-Al-rich inclusions in other chondrites (i.e., [1-7,9,22]).

They are essentially Fe-free and also very low in alkalies. Again, the composition of these titanagulites changes with the distance from the chondrule surface: TiO$_2$ and Al$_2$O$_3$ (and perhaps FeO and the alkalies) decrease, whereas SiO$_2$ and MgO (and Cr$_2$O$_3$?) increase towards the central portion. The spinel composition changes similarly, with TiO$_2$ and FeO decreasing, and Cr$_2$O$_3$ increasing towards the center. Anorthite is not present within the outermost zone. However, although subtle, some compositional change was observed with MgO (and Na$_2$O?) increasing and CaO decreasing towards the center.

3. Discussion

Some petrologic arguments have already been presented [10,18] against the primordial condensation model for Ca-Al-rich objects with the shape (chondrules) and textures (igneous) of the majority of them. This study further enhances these arguments.

---

**TABLE 1**

Averaged electron-microprobe analyses of different phases in a Ca-Al-rich chondrule from the Bali carbonaceous chondrite. Analyses are arranged in the order of increasing distance from the chondrule surface (surface-intermediate zone-central portion).

<table>
<thead>
<tr>
<th>Melilitic</th>
<th>Aluminous Titanagulate</th>
<th>Spinel</th>
<th>Anorthite</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>surface</td>
<td>intermediate</td>
<td>center</td>
</tr>
<tr>
<td>SiO$_2$</td>
<td>31.1</td>
<td>34.5</td>
<td>38.9</td>
</tr>
<tr>
<td>TiO$_2$</td>
<td>0.03</td>
<td>0.02</td>
<td>0.05</td>
</tr>
<tr>
<td>Al$_2$O$_3$</td>
<td>23.7</td>
<td>18.5</td>
<td>8.9</td>
</tr>
<tr>
<td>Cr$_2$O$_3$</td>
<td>0.03</td>
<td>0.03</td>
<td>0.04</td>
</tr>
<tr>
<td>FeO</td>
<td>0.10</td>
<td>0.08</td>
<td>0.03</td>
</tr>
<tr>
<td>MgO</td>
<td>4.6</td>
<td>7.5</td>
<td>9.6</td>
</tr>
<tr>
<td>CaO</td>
<td>40.5</td>
<td>40.1</td>
<td>40.2</td>
</tr>
<tr>
<td>Na$_2$O</td>
<td>0.06</td>
<td>0.15</td>
<td>0.25</td>
</tr>
<tr>
<td>K$_2$O</td>
<td>0.06</td>
<td>0.06</td>
<td>0.08</td>
</tr>
<tr>
<td>Total</td>
<td>100.18</td>
<td>100.94</td>
<td>98.05</td>
</tr>
<tr>
<td>Number of analyses</td>
<td>19</td>
<td>7</td>
<td>4</td>
</tr>
</tbody>
</table>

---

The Ca-Al-rich is zoned — minerals of this zoning is expected from the i.e. all refractories of the surface whereas the Na$_2$O are concentrated in this distribution, typically different elements show distinctly different elements. The apparent core and vapor pressure relationships indicate an element between liquid and solid chondrule and to be conclusive evidence from a liquid state might have occurred on the solar nebula where this chondrule and the carbon of carbonaceous objects are well preserved. "Primordial" contain either old or "pristine high Mg [24]. The element and relationships explained by rapid cooling of an overheated melt. Thus, i.e. heated subtemperature (which had [8,18]) is needed to be the most plausible.

Such events change the target and produce gas clouds from the elements could contain indistinguishable observed enrichment. The condensation of compounds from the vaporization of high vapor pressures in overheated [10,19,20]. Gas is found to be the same distributions in the carbonaceous counterparts, glass.

Both models of condensation and evaporation can...
from Ca–Al-rich are typical of the major exclusion zone of other chondrites. The Ca–Al-rich chondrule from the Bali chondrite is zoned — mineralogically and chemically. The nature of this zoned is exactly opposite to what could be expected from the primordial condensation model; i.e., all refractories (e.g., Ti, Al) are enriched near the surface whereas the more volatile elements (Mg, Cr, Si, Na) are concentrated in the center of the chondrule. This distribution, the equal enrichments of geochemically different elements, e.g., Ir and Sc [11,13–15] and the apparent correlation between enrichment factors and vapor pressures [10] (elements and/or compounds) indicate an elemental fractionation via an interaction between liquid and/or solid and gas. The shape of the Bali chondrule and its mineralogy and texture seem to be conclusive evidence for an original crystallization from a liquid although some mild recrystallization might have occurred. Since condensation from the solar nebula would not produce a liquid [16,17] this chondrule and many similar objects from a variety of carbonaceous chondrites cannot be unaltered "primordial" condensates although they may be relatively old or "primitive" (e.g., low 87Sr/86Sr [23] and high 26Mg [24]). Rather, the overall composition and the element and mineral distribution seem easiest explained by rapid loss of volatiles from the surface of an overheated melt which contains sufficient heat to preserve a diffusion gradient (and/or equilibrium mineral assemblages). If overheating, i.e., heated substantially beyond the liquidus temperature (which also may be very high ~2,000 °K [8,18]) is necessary, high-velocity impacts seem to be the most plausible heat source.

Such events could cause substantial volatilization of target and projectile material, generating dense gas clouds from which a variety of compounds and elements could condense. Thus, two geochemically indistinguishable mechanisms could account for the observed enrichment of refractory elements: fractional condensation of low-vapor-pressure elements or compounds from an impact gas cloud and fractional evaporation of high-vapor-pressure elements or compounds from overheated target and/or projectile material [10,19,20]. Gas–liquid interactions have already been found to be the most plausible cause of zonal element distributions in meteoritic chondrules and their lunar counterparts, glass spheres [18,25].

Both models, fractional condensation and fractional evaporation, can also account for the large variations in composition between different "white" fragments. Although only some fragments from the Allende chondrite have been studied so far, the measured Ir contents vary by a factor of ~70 [11] or ~400 [27], if Ir depleted Na-rich fragments are included. This is not easily explained by the equilibrium condensation model. Furthermore, inclusions in Lanzé seem to be much more highly enriched (at least ten times) in refractory elements than those from Allende. Unfortunately, their small sizes prevent thorough bulk chemical studies but for some refractories enrichments of up to ~400 times the chondritic level can be calculated from mineral analysis [10]. This, as well as the different enrichment level of refractories in white inclusions from different chondrites, also seem easier explained by non-equilibrium processes rather than equilibrium condensation in the solar nebula.

Enrichment factors of 20 [11,12,13–15] to 400 [10] of refractories over the chondritic abundances require fractions of 5% to less than 1% being taken out of a bulk of chondritic composition. Probably none of the processes proposed here can produce these enrichments in just one step. Rather, repeated impacts causing partial evaporation and partial recondensation can account for the variety of white inclusions we observe. The early history of the parent bodies of chondrites as well as other planetesimals and planets is believed to be characterized by high impact rates. This way the observed "primitive" isotopic nature of Ca–Al-rich inclusions could be brought into accord with all the observations pointing to a secondary origin, like the igneous origin [10,19,20,22] and the apparent mixed chemistry [12].

The parent material for the white inclusions can either be chondritic material or a material already enriched in real primitive condensates. What we observe now are certainly not "primitive" condensates but rather secondary objects which could have partly originated from such condensates but could also have been formed from a chondritic parent by either partial evaporation and/or condensation of matter in the course of repeated impact events.

Acknowledgements

This work has been supported by the Jubiläumsfonds der Österreichischen Nationalbank (Projects
No. 210 and 735) and the Fonds zur Förderung der Wissenschaftlichen Forschung (Projects No. 1059 and 2272).

References

27. H. Wänke, et al., unpublished data.