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Dmitryivanovite: A new high-pressure calcium aluminum oxide from the Northwest Africa 470 CH3 chondrite characterized using electron backscatter diffraction analysis

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ABSTRACT

Dmitryivanovite ($CaAl_2O_4$) is a newly described, calcium aluminum oxide from the Northwest Africa 470 (NWA470) CH3 chondrite (Ivanova et al. 2002). NWA470 contains abundant small Ca,Al-rich inclusions (CAIs), and dmitryivanovite, whose composition is close to stoichiometric $CaAl_2O_4$ [$Ca_{1.000}(Al_{1.993}Si_{0.003}Ti_{0.002})_{1.998}O_4$], was found in one of these CAIs. It occurs as ~10 μm subhedral grains intergrown with grossite ($CaAl_4O_7$), perovskite, and melilite. Electron backscatter diffraction (EBSD) analysis revealed that dmitryivanovite is a high-pressure polymorph of $CaAl_2O_4$ ($a = 7.95$, $b = 8.62$, $c = 10.25$ Å, $\beta = 93.1°$, space group $P2_1/c$, and $Z = 12$). Dmitryivanovite is the third phase to be described from nature in the binary system of $CaO–Al_2O_3$, the other two being hibonite ($CaAl_{12}O_{19}$) and grossite ($CaAl_4O_7$)—all are found in CAIs. The presence of $CaAl_2O_4$ in NWA470 suggests a local elevated dust/gas ratio in the solar nebula. The phase diagram of $CaAl_2O_4$ shows that ~2 GPa is required to stabilize the high-pressure $CaAl_2O_4$ polymorph at 1327 °C, above which $CaAl_2O_4$ condenses from the solar nebula. Because it is unlikely that the solar nebula ever had such a high total gas pressure, it appears more probable that condensation of the low-pressure polymorph occurred in the solar nebula with an enhanced dust-to-gas ratio and that subsequently the high-pressure polymorph was produced by shock metamorphism, most likely after the $CaAl_2O_4$-bearing CAI was incorporated into the NWA470 parent asteroid.

Keywords: Dmitryivanovite, CAI, electron backscatter diffraction, new minerals, CH chondrite

INTRODUCTION

Ca,Al-rich inclusions (CAIs) are important components of chondritic meteorites, inasmuch as thermodynamic calculations show that the high-temperature minerals (melilite, aluminum oxides, spinel) abundant in them would be the first condensates in a cooling solar nebula (e.g., Grossman 1972; Grossman and Larimer 1974), and cosmochronology studies verify that they are among the oldest native materials known from our solar system (e.g., Wadhwa and Russell 2000). Because CAIs greatly differ in composition, oxidation state, thermal history, and isotopic characteristics from the other components of chondrites, some special setting or circumstance should have been required for their formation (e.g., Guan et al. 2000). CAIs were originally viewed as simply the first condensates from a hot, monotonically cooling gas of solar nebula composition and were considered to have formed by some process different from the one that formed chondrules. Eventually it was realized that the majority of CAIs had been, in fact, repeatedly thermally processed in the nebula, and that few are truly condensates (Stolper and Paque 1986; MacPherson and Davis 1993), although the heating mechanism remains mysterious. Thus, the detailed mineralogical analysis of CAIs is critical to a full understanding of their unique formation conditions in the solar nebula.

In this paper, we report the first crystallographic characterization of a new calcium dialuminum oxide ($CaAl_2O_4$) in a CAI from the CH3 chondrite Northwest Africa 470 (NWA470) (Ivanova et al. 2002). Although CAIs constitute only ~1 vol% of the CH chondrites, they are critical components (e.g., Weisberg et al. 1995; Grossman et al. 1988). The CAIs in CH chondrites are extremely small (usually ~100 μm) and show different mineralogical characteristics from CAIs in other carbonaceous chondrites. The distinctive aspect of CAIs in CH chondrites is the absence of secondary alteration, unlike for those in CM2 and CV3 chondrites, and thus they have probably retained their original, primitive chemical and mineralogical characteristics (e.g., Brearley and Jones 1998). Due to the small size of CAIs in CH chondrites, it is sometimes difficult to fully characterize their constituent phases, especially those that are new to science. In this respect, electron backscatter diffraction (EBSD) analysis combined with field-emission gun, scanning electron microscopy (FEG-SEM) is a useful non-destructive technique that permits the necessary elucidation of crystallography of thin sections at the submicrometer scale (e.g., Harland et al. 1981; Goehner and Michael 1996; Kogure 2002).

The new mineral was found in a CAI from the NWA470

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CH3, as first reported by Ivanova et al. (2002), who detailed its composition and occurrence, and proposed a possible formation scenario based on condensation calculations, but who were unable to obtain any crystallographic data due to its small size. Since then, we have verified the composition of the phase and succeeded in determining its crystal structure using EBSD analysis, which permitted us to successfully propose the phase as a new mineral. We have named this mineral dmitryivanovite, after Dmitriy A. Ivanov (1962–1986), a geologist, mineralogist, and petrologist who died tragically on a field expedition to study igneous rocks in the Caucasus Mountains, where he was investigating the petrogenesis of magmas of the recent volcanic centers of the Caucasus Ridge. The new mineral and the name have been approved by the Commission on New Minerals and Mineral Names of the IMA. The type polished section containing dmitryivanovite is deposited in the meteorite collection of the Russian Academy of Sciences at the Vernadsky Institute, Moscow, Russia.

Sample and analytical techniques

The NWA470 meteorite is a metal-rich carbonaceous chondrite found in 1999 and classified as a CH3 chondrite (Ivanova et al. 2002). NWA470 is slightly weathered (chondrite weathering grade W1), has weakly developed shock features (chondrite shock stage S2), and shows no evidence of post-accretional aqueous alteration or thermal metamorphism. NWA470 resembles other CH chondrites in having abundant (24 vol%) Fe-Ni metal grains with similar chemical zoning characteristics and solar Ni/Co ratios (Meibom et al. 1999; Weisberg and Prinz 1999; Petaev et al. 2001, 2003), numerous small CAIs of different types, and fine-grained dark inclusions. NWA470 may be paired with Acfer 182, 207, and 214 because of similar petrography (Bischoff et al. 1993). The new mineral, dmitryivanovite, was found in one of the grossite-rich CAIs (labeled E1-005) in a single polished section of NWA470 (section No. 1). Grossite was first reported by Weber and Bischoff (1994a, 1994b) in CAIs from some CH chondrites (Acfer 182, Acfer 059, El Djouf 001, and ALH85085), and is a major component of CAIs in these meteorites (Weber et al. 1995), but dmitryivanovite has never been reported previous to Ivanova et al. (2002).

We employed a ThermoNoran PhaseID EBSD system installed in a Hitachi S-4500 FEG-SEM equipped with a Kevex EDS (Department of Earth and Planetary Science, University of Tokyo) for crystallographic characterization of dmitryivanovite. The accelerating voltage of the incident beam was 20 kV with a beam current of 2–3 nA. The collection semi-angle of the EBSD detector was ~37.5°. In the EBSD technique, the analyzed section is tilted by ~70° from the horizontal toward a phosphor screen (detector) upon which backscattered electrons form an EBSD (Kikuchi) pattern. The incident electrons are scattered mainly by phonons at the surface of the specimen with a large scattering angle and a small energy loss. These divergent electrons in the specimen are scattered again to form Kikuchi bands at certain angles. Calculations of Kikuchi patterns and analyses of observed EBSD patterns were performed using a computer program developed by Kogure (2003). We referenced the crystal structures of several calcium aluminate structures [synthetic $CaAl_2O_4$ (both low-pressure and high-pressure phases), grossite ($CaAl_4O_7$), and hibonite ($CaAl_{12}O_{19}$)] to compare with the obtained EBSD patterns. Finally, we calculated the powder X-ray diffraction pattern for dmitryivanovite using the XPOW Program (Downs et al. 1993).

Occurrence

NWA470 contains numerous CAIs of various mineralogical assemblages and is typical of the CH meteorite group (e.g., Weisberg et al. 1995; Brearley and Jones 1998). The dominant CAI type in NWA470 is usually concentrically zoned with grossite + hibonite + perovskite + spinel cores surrounded by melilite mantles and anorthite and Al-diopside rims (Ivanova et al. 2002). One of these particular CAIs (CAI E1-005) contains

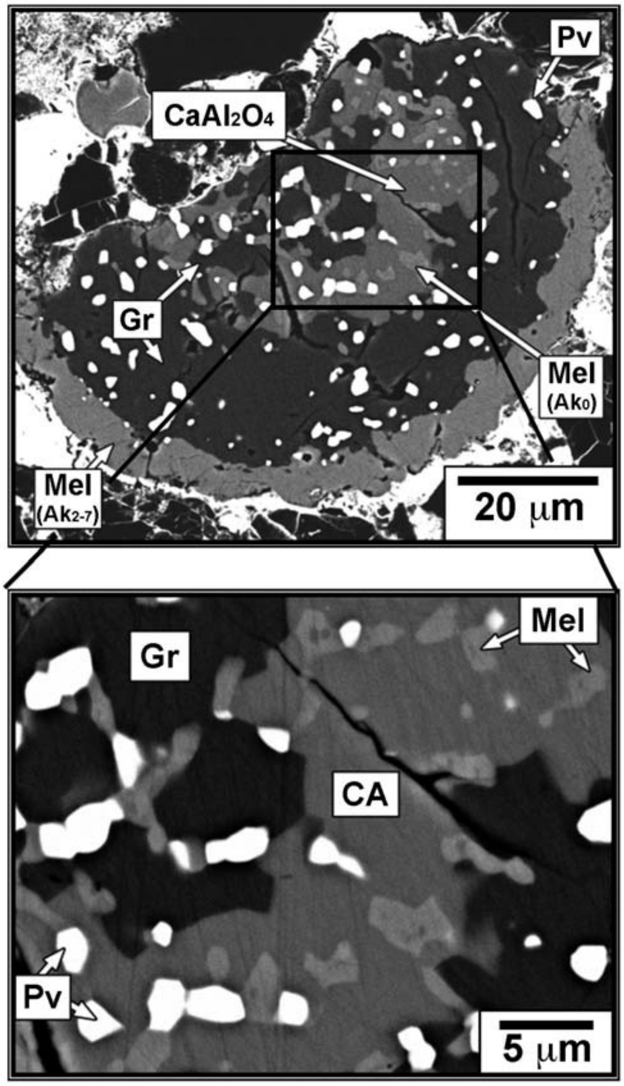

FIGURE 1. Backscattered electron image of E1-005 (adapted from Ivanova et al. 2002). The innermost core of the inclusion consists mainly of grossite (Gr) intergrown with perovskite (Pv) and melilite (Mel) that is pure gehlenite (Ak_0); external to this is a zone rich in dmitryivanovite (CA) that encloses small grains of melilite, perovskite, and grossite (Gr); surrounding this is a massive zone consisting mainly of grossite that encloses perovskite, and finally the outermost zone consists exclusively of melilite that is different in composition ($Ak_{2–7}$) from the small interior gehlenite grains.

the dmitryivanovite, whose composition is close to stoichiometric $CaAl_2O_4$. The innermost core of the CAI consists mostly of grossite, with interstitial perovskite (≤3 μm) and melilite (≤5 μm) grains. Surrounding this is a zone consisting mainly of dmitryivanovite, which encloses melilite, perovskite, and minor grossite (Fig. 1). The dmitryivanovite-bearing zone is up to 20 μm thick. The dmitryivanovite-rich zone is in turn enclosed in a region consisting mostly of grossite with abundant included perovskite grains. Finally, the entire CAI is mantled by a nearly monomineralic, ~8 μm thick rind of melilite (Fig. 1). Tiny grains of perovskite decorate the contact between the melilite rind and the interior zone, but the melilite rind itself is notably lacking in perovskite, which is abundant everywhere else in the CAI.

The entire CAI is so small and fine grained that optical properties of the $CaAl_2O_4$ cannot be determined beyond the observations that it is birefringent and, when viewed in plane-polarized light, colorless. Dmitryivanovite has blue cathodoluminescence. The largest clot of dmitryivanovite crystals is no larger than ~10 μm, but the sizes of individual crystals could not be determined by SEM.

COMPOSITION AND CRYSTAL STRUCTURE

The composition of dmitryivanovite as reported by Ivanova et al. (2002) is close to the theoretical value for pure $CaAl_2O_4$. The empirical formula for dmitryivanovite (based on 4 O atoms) is: $Ca_{1.000}(Al_{1.993}Si_{0.003}Ti_{0.002})_{1.998}O_4$.

We analyzed several different areas of $CaAl_2O_4$ in the E1-005 CAI from NWA470 by EBSD. Because the spatial resolution of EBSD analysis is <100 nm laterally and on the order of 10 nm for depth (Harland et al. 1981), it is obvious that the EBSD patterns we obtained are from single phases without overlap by surrounding phases. All the analyzed areas gave sharp EBSD patterns (Fig. 2). The obtained EBSD patterns were compared to those of several calcium aluminate oxide phases including $CaAl_2O_4$ (Table 1). We found that the EBSD patterns of the new mineral matched only the high-pressure polymorph of $CaAl_2O_4$, and was quite distinct from low-pressure $CaAl_2O_4$ (Ito et al. 1980). Thus, dmitryivanovite has the following crystallographic parameters: a = 7.95, b = 8.62, c = 10.25 Å, space group $P2_1/c$, and Z = 12 (all taken from Ito et al. 1980). The crystal structure of dmitryivanovite is shown in Figure 3. Aluminum is in tetrahedral coordination, which is consistent with the result suggested by microprobe analysis. The powder X-ray diffraction pattern calculated for dmitryivanovite using the XPow program is given in Table 2. It was, of course, impossible to obtain an X-ray powder pattern directly from the available natural material.

DISCUSSION

The binary system of CaO-Al_2O_3 includes two important minerals found in CAIs: hibonite ($CaAl_{12}O_{19}$) and grossite ($CaAl_4O_7$). However, neither of two other intermediate compounds in this system, $CaAl_2O_4$ and $Ca_3Al_2O_6$ had not been conclusively found to occur in nature until $CaAl_2O_4$ was reported in the NWA470 CH3 chondrite (Ivanova et al. 2002). An additional phase,

TABLE 1. Cell parameters of calcium aluminum oxides

Phase	System	Space group	Lattice constants			
			a	*b*	*c*	β
Grossite (natural)*	Monoclinic	$C2/c$	12.94(1)	8.910(8)	5.446(4)	107.0(1)
Grossite (synthetic)†	Monoclinic	$C2/c$	12.89	8.88	5.45	107.05
Hibonite‡	Hexagonal	$P6_3/mmc$	5.564	5.564	21.892	
$CaAl_2O_4$ (low-*P*)§	Monoclinic	$P2_1/c$	8.07	8.09	15.21	90.14
$CaAl_2O_4$ (high-*P*)§	Monoclinic	$P2_1/c$	7.95	8.62	10.25	93.10

* Weber and Bischoff (1994a, 1994b).
† Boyko and Wisnyi (1958).
‡ Kato and Saalfeld (1968).
§ Ito et al. (1980).

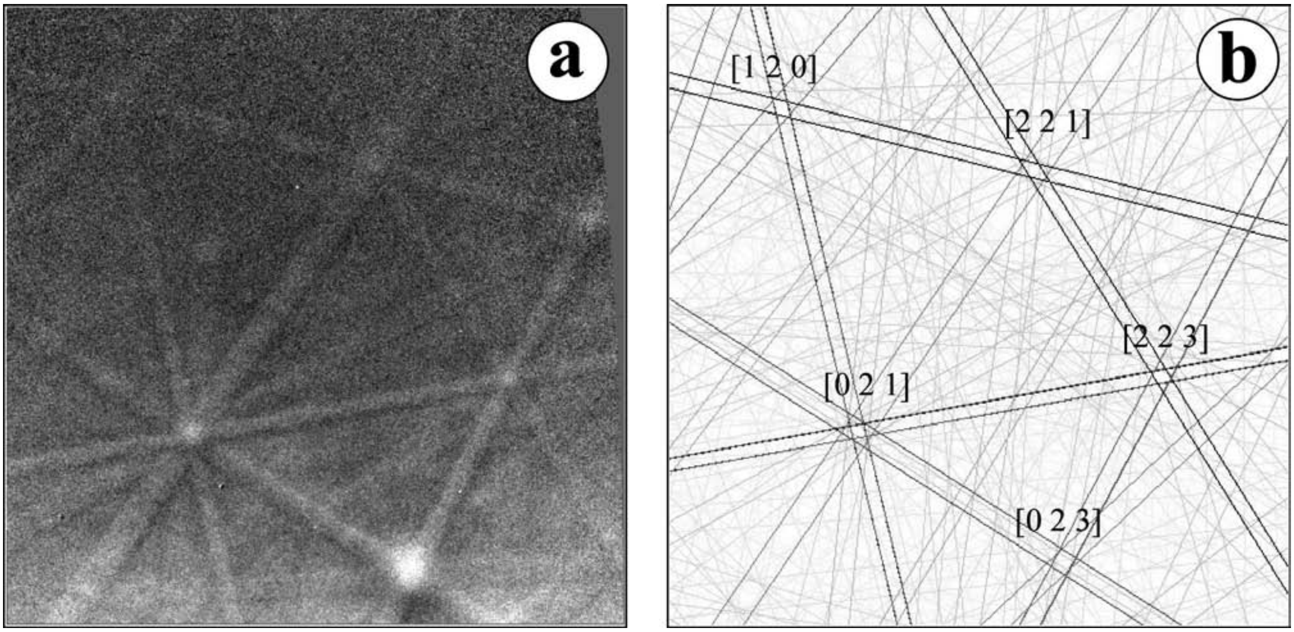

FIGURE 2. (**a**) The observed EBSD pattern of dmitryivanovite. (**b**) The matching calculated pattern of the high-pressure polymorph of synthetic $CaAl_2O_4$ (Ito et al. 1980).

$Ca_{12}Al_{14}O_{33}$ (mayenite), has been experimentally produced in the presence of water and hence may not be a true binary compound (Nurse et al. 1965; Jerebtsov and Mikhailov 2001). Thus, the discovery of dmitryivanovite presents us with the third calcium aluminum oxide to be described from nature.

Ivanova et al. (2002) proposed that the initial composition of the CAI precursor materials in NWA470 must have been highly CaO-Al_2O_3 rich and MgO-SiO_2 poor to form both grossite and dmitryivanovite. The most likely scenario for producing such a bulk composition involves either nebular condensation or evaporation, or a combination of both. Calculations by several investigators suggest that the condensation of $CaAl_2O_4$ from a nebular system requires considerable enrichment in dust relative to gas (e.g., Ebel and Grossman 2000; Ebel 2006). Therefore, the presence of the $CaAl_2O_4$-bearing CAI in this CH3 chondrite suggests that there was just such a region in the solar nebula where dust was suitably abundant. The apparent rarity of dmitryivanovite suggests that such dust regions may have been correspondingly rare in the nebula or that these regions were poorly sampled by available meteorites.

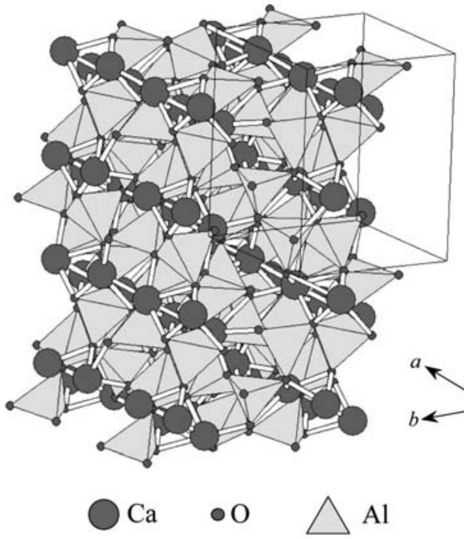

FIGURE 3. Crystal structure of dmitryivanovite drawn by the ATOMS software using the structural data by Dougill (1957).

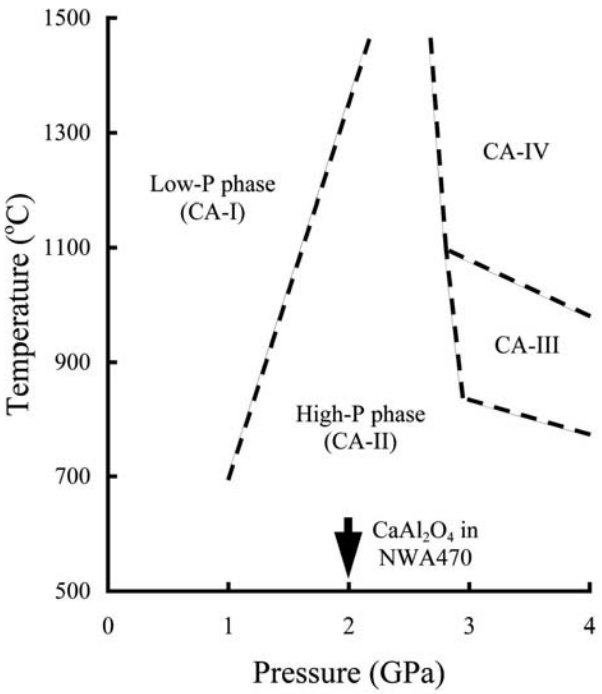

FIGURE 4. Stability field of $CaAl_2O_4$ under high pressures and high temperatures after Ito et al. (1980). The presence of the high-pressure polymorph of $CaAl_2O_4$ (CA-II) is consistent with the degree of shock experienced by NWA470 (~50 °C, ~2 GPa).

Our EBSD analysis reveals that $CaAl_2O_4$ is present within NWA470 only in its high-pressure polymorph. The phase boundary between low-pressure $CaAl_2O_4$ and the high-pressure polymorph has been experimentally determined (Ito et al. 1980) to lie between 1 GPa at 700 °C and 2.5 GPa at 1500 °C (Fig. 4). Approximately, 2 GPa is required to stabilize the high-pressure $CaAl_2O_4$ polymorph at 1327 °C, above which $CaAl_2O_4$ condenses from the solar nebula (Ebel 2006). It is unlikely that the solar nebula ever had such a high total gas pressure, and thus dmitryivanovite probably originally formed as the low-pressure polymorph. NWA470 shows weakly developed, but distinct, shock features corresponding to the shock stage S2, and the estimated shock pressure and peak temperature are ~2 GPa and <50 °C, respectively (Stöffler et al. 1991). Under these pressure-temperature conditions, the high-pressure polymorph of $CaAl_2O_4$ should be stable because of the large stability field of this phase at low temperatures (Fig. 4). Therefore, we propose that the high-pressure polymorph of $CaAl_2O_4$ (dmitryivanovite) was produced during shock metamorphism of the NWA470 parent asteroid. Alternatively, the CAI could have been shocked before final incorporation into NWA470 since CH chondrite components might have been formed or modified in impact related processes (e.g., Krot et al. 2006). This conclusion suggests that the low-pressure polymorph of $CaAl_2O_4$ (as yet unknown as a

TABLE 2. Partial calculated X-ray powder-diffraction pattern for dmitryivanovite

d_{calc}	I_{calc}	*hkl*	d_{calc}	I_{calc}	*hkl*
6.593	5	011	2.224	11	$\bar{3}21$
5.839	7	110	2.198	14	033
5.161	12	$\bar{1}11$	2.137	15	$\bar{2}14$
4.987	18	111	2.1	5	$\bar{3}22$
4.4	7	012	2.084	12	$\bar{3}13$
3.969	13	200	1.985	19	400
3.605	7	210	1.963	9	$\bar{2}24$
3.583	17	$\bar{1}21$	1.894	15	240
3.523	7	121	1.888	29	224
3.172	5	013	1.871	8	$\bar{2}41$
3.083	6	$\bar{1}22$	1.843	6	323
3.018	100	$\bar{2}12$	1.778	7	412
2.92	83	220	1.658	8	$\bar{3}41$
2.882	52	212	1.58	7	$\bar{3}34$
2.766	26	031	1.572	10	$\bar{2}16$
2.559	42	004	1.553	5	315
2.505	46	032	1.536	6	432
2.503	9	123	1.52	9	$\bar{2}52$
2.494	9	222	1.514	10	216
2.486	5	$\bar{3}11$	1.504	10	244
2.453	9	014	1.485	5	$\bar{5}21$
2.408	13	$\bar{1}32$	1.467	29	036
2.398	14	104	1.449	7	$\bar{5}13$
2.371	31	132	1.437	8	060
2.3	7	302	1.341	6	523

Note: Only diffraction spacings with $I/I_0 \geq 5$ are given down to d_{calc} of 1.34.

natural phase) may be present in some meteoritic CAIs from less highly shocked asteroids.

The discovery of dmitryivanovite from the NWA470 CH3 chondrite is among the first new mineral identifications bringing EBSD analysis to bear on astromaterials. EBSD analysis has a great potential to characterize new phases in astromaterials and to constrain their unique formation conditions.

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