



Cosmic history and a candidate parent asteroid for the quasicrystal-bearing meteorite Khatyrka

Matthias M.M. Meier^{a,*}, Luca Bindi^{b,c}, Philipp R. Heck^d, April I. Neander^e,
Nicole H. Spring^{f,g}, My E.I. Riebe^{a,1}, Colin Maden^a, Heinrich Baur^a, Paul J. Steinhardt^h,
Rainer Wieler^a, Henner Busemann^a

^a Institute of Geochemistry and Petrology, ETH Zurich, Zurich, Switzerland

^b Dipartimento di Scienze della Terra, Università di Firenze, Florence, Italy

^c CNR-Istituto di Geoscienze e Georisorse, Sezione di Firenze, Florence, Italy

^d Robert A. Pritzker Center for Meteoritics and Polar Studies, Field Museum of Natural History, Chicago, USA

^e Department of Organismal Biology and Anatomy, University of Chicago, Chicago, USA

^f School of Earth and Environmental Sciences, University of Manchester, Manchester, UK

^g Department of Earth and Atmospheric Sciences, University of Alberta, Edmonton, Canada

^h Department of Physics, and Princeton Center for Theoretical Science, Princeton University, Princeton, USA

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ABSTRACT

The unique CV-type meteorite Khatyrka is the only natural sample in which “quasicrystals” and associated crystalline Cu, Al-alloys, including khatyrkite and cupalite, have been found. They are suspected to have formed in the early Solar System. To better understand the origin of these exotic phases, and the relationship of Khatyrka to other CV chondrites, we have measured He and Ne in six individual, ~40-μm-sized olivine grains from Khatyrka. We find a cosmic-ray exposure age of about 2–4 Ma (if the meteoroid was <3 m in diameter, more if it was larger). The U, Th–He ages of the olivine grains suggest that Khatyrka experienced a relatively recent (<600 Ma) shock event, which created pressure and temperature conditions sufficient to form both the quasicrystals and the high-pressure phases found in the meteorite. We propose that the parent body of Khatyrka is the large K-type asteroid 89 Julia, based on its peculiar, but matching reflectance spectrum, evidence for an impact/shock event within the last few 100 Ma (which formed the Julia family), and its location close to strong orbital resonances, so that the Khatyrka meteoroid could plausibly have reached Earth within its rather short cosmic-ray exposure age.

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1. Introduction

The motivation for this noble gas study is the curiosity and fascination with the origin of an exotic type of material: quasicrystals. Short for quasi-periodic crystals, they are materials showing a quasi-periodic arrangement of atoms, and rotational symmetries forbidden to ordinary crystals (e.g., five-fold). First proposed by Levine and Steinhardt (1984), they were first synthesized and identified in the laboratory by Shechtman et al. (1984). The ensuing multi-decade search for natural quasicrystals was eventually successful when a powder from a millimeter-sized

* Corresponding author.

E-mail address: matthias.meier@erdw.ethz.ch (M.M.M. Meier).

¹ Current address: Department of Terrestrial Magnetism, Carnegie Institution of Washington, Washington, USA.

rock sample (later called the “Florence Sample”) from the collection of the Museum of Natural History in Florence, Italy, displayed a diffraction pattern with a five-fold symmetry (Bindi et al., 2009). This first natural quasicrystal, with a composition Al₆₃Cu₂₄Fe₁₃, was named icosahedrite. In the host rock of the Florence sample, silicates and oxides are partially inter-grown with exotic copper–aluminum-alloys (khatyrkite, CuAl₂, and cupalite, CuAl) which contain the quasicrystals. The oxygen isotopic compositions of silicates and oxides in the Florence Sample suggest an extraterrestrial origin (Bindi et al., 2012), as they match the ones found in some carbonaceous chondrites. The provenance of the Florence Sample was eventually traced back to a Soviet prospecting expedition to the Far East of Russia in 1979 (Bindi and Steinhardt, 2014). To find more of the exotic material, an expedition was launched to the Koryak mountain range in the Chukotka Autonomous Region in 2011. Eight millimeter-sized fragments of extraterrestrial origin were found by panning

Holocene (>7 ka old) river deposits (MacPherson et al., 2013). Again, Cu, Al-alloys were found attached to, or intergrown with, extraterrestrial silicates and oxides. The oxygen isotopes, chemistry and petrology of the eight new fragments all suggest a CV_{ox} (carbonaceous chondrite, Vigarano-type, oxidized subtype) classification for the meteorite, now named Khatyrka (Ruzicka et al., 2014). The achondritic, diopside–hedenbergite-rich and Cu, Al-alloy-encrusted Florence Sample suggests that Khatyrka is an unusual chondritic breccia which accreted both achondritic and exotic materials (MacPherson et al., 2013).

So far, no other meteorite is known to contain Cu, Al-alloys or quasicrystals. The lack of $^{26}\text{Mg}/^{24}\text{Mg}$ anomalies suggests that the Cu, Al-alloys formed after the primordial ^{26}Al ($t_{1/2} = 0.7$ million years, Ma) had decayed away, i.e., at least ~ 3 Ma after formation of the oldest condensates in the Solar System 4.567 billion years (Ga) ago (MacPherson et al., 2013). Hollister et al. (2014) found high-pressure mineral phases (including stishovite and ahrensite) indicating Khatyrka was at one point exposed to pressures >5 GPa and temperatures >1200 °C, followed by rapid cooling. Synthetic icosahedrite remains stable under these conditions (Stagno et al., 2015). These conditions are also typical for asteroid collisions (Stoeffler et al., 1991). In dynamic shock experiments, Asimow et al. (2016) successfully synthesized icosahedral phases with a composition close to that of icosahedrite, and Hamann et al. (2016) reported the shock-induced formation of khatyrkite (CuAl_2), mixing of target and projectile material, and localized melting along grain boundaries, resembling the assemblages found in Khatyrka. Lin et al. (2017) argued that a two-stage formation model is needed to explain the mineralogical and petrographic features observed in Khatyrka: first, an event as early as 4.564 Ga in which quasicrystals with icosahedrite composition formed; and, second, a more recent impact-induced shock that led to the formation of a second generation of quasicrystals, with a different composition. Ivanova et al. (2017) suggested the possibility of an anthropogenic origin of the Cu, Al-alloys from mining operations, but this explanation is incompatible with the chemistry and the thermodynamic conditions needed to explain all observations listed above (see also MacPherson et al., 2016).

In this work, we aim to better understand the origin and history of the quasicrystal-bearing Khatyrka meteorite and its relationship to other meteorites, in particular other CV chondrites. We do this by measuring the helium and neon (He, Ne) content of individual Khatyrka olivine grains, in order to determine their cosmic-ray exposure and radiogenic gas retention (based on uranium–thorium–helium = U, Th–He) ages. Due to the extremely small mass available from Khatyrka (total mass <0.1 g), destructive analyses have to be reduced to the minimum. We worked with single grains of ca. 40 μm diameter, which were all part of a chondrule from Khatyrka fragment #126 (recovered in the 2011 expedition). Because of the very small gas amounts expected, we used the high-sensitivity “compressor-source” noble gas mass spectrometer at ETH Zurich (Baur, 1999). This unique instrument has previously been used in a similar way to measure He and Ne in individual mineral grains returned from asteroid Itokawa (Meier et al., 2014).

2. Methods

2.1. Volume determination by X-ray tomography

To determine cosmic-ray exposure and radiogenic gas retention ages, we need noble gas concentrations ($\text{cm}^3_{\text{STP}}/\text{g}$), which requires the determination of the masses of the individual grains. This is not possible to do both safely and reliably on a micro-balance for such small (<1 μg) grains. To determine their masses, we

first measure their volumes using high-resolution X-ray tomography (nano-CT), and then multiply these volumes with the densities calculated from their mineralogical composition (determined with SEM-EDS) and textbook mineral densities. At the University of Florence, seven olivine grains from Khatyrka fragment #126 (named #126-01 through -07, or 1 through 7 for short in the figures), all fragments from a chondrule (judging from their size and chemistry), were transferred with a micro-manipulator needle from a TEM grid to a carbon tape. The samples were then imaged (on the tape) with a GE manufactured v|tome|x s micro/nano-CT scanner located at the UChicago PaleoCT facility, using the 180 kV nano-CT tube, an acceleration voltage of 80 kV, beam current of 70 μA with 500 ms integration time, and no filter. The voxels of the scan are isometric with a size of 5.473 μm . Grain volumes were then determined from the CT images using the “3D-object counter” plug-in in Fiji/ImageJ (Schindelin et al., 2012), which finds 3D-connected objects with a voxel brightness above a user-defined 8-bit (256) gray-scale value (called the “threshold”). We first searched a range of thresholds where the grains are well-resolved from the background, then searched for the sequence of four thresholds for which the volume change between adjacent gray-scale steps was minimal. We then corrected for surface resolution effects (i.e., for voxels close to the actual grain which are only partially in-filled by grain material, and thus have a reduced voxel brightness: they fall below the threshold although they contain a sub-voxel-sized part of the grain) by interpolating between the average brightness of unambiguous (internal) grain voxels and background voxels (e.g., if the grain material brightness is 120 and the background brightness is 40, a surface-near voxel with brightness 80 is assumed to be half in-filled with grain material). Grain #126-06 was too small, and too fragmented to be of use for this study, and was thus not further analyzed. The grain volume uncertainties in Table 1 correspond to the range in volume within the sequence of four thresholds with minimal volume changes (after correction for surface effects). Grain #126-05 fragmented after nano-CT scanning (as visible from the SEM images, see section 2.2). Only two of its fragments could be transferred to the sample holder for noble gas analysis. Their volume was estimated from their cross-sectional area in the SEM images and an empirical relationship between cross-sectional area (A) and volume (V) established by the other five grains, $A = (0.080 \pm 0.009) \times V$ ($R^2 = 0.998$), which resulted in a somewhat larger mass uncertainty for these fragments. The volume of grain #126-05 given in Table 1 represents the sum of the two fragments.

2.2. Chemical composition and grain masses

After nano-CT scanning, the bulk elemental composition of the grains was qualitatively determined by spot analysis on unpolished surfaces using an Oxford Instruments XMax-50 energy dispersive X-ray (EDX) spectrometer, mounted on a Zeiss Evo 60 Scanning Electron Microscope (SEM; acceleration voltage 20 kV, electron current 400 pA) at the Field Museum of Natural History in Chicago. Back-Scattered Electron (BSE) images of the grains are shown in Fig. S1 in the Supplementary Material. We found an average composition of $95 \pm 5\%$ forsterite (Mg_2SiO_4 ; 3.28 g/cm^3) and $5 \pm 5\%$ fayalite (Fe_2SiO_4 ; 4.39 g/cm^3), corresponding to a grain density of $3.33 \pm 0.06 \text{ g}/\text{cm}^3$ (Table 1). Since the individual measurements were done in spot analysis mode and are not resolved from their average, we only use the average density to calculate the grain masses in Table 1. The six grains (or grain fragments) were then transferred to a customized Al sample holder for noble gas analysis, using a hydraulic Narishige MMO-202ND/MMN-1 micro-manipulator attached to a Nikon Eclipse Microscope at SEAES, University of Manchester (an image of a grain in transfer is shown in Fig. S2 in the Supplementary Material).

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