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Citation: Collinet, M. and Grove, T.L. (2020), Incremental melting in the ureilite parent body: Initial composition, melting temperatures, and melt compositions. *Meteorit Planet Sci*, 55: 832-856.

As Published: 10.1111/MAPS.13471

Publisher: Wiley

Persistent URL: <https://hdl.handle.net/1721.1/132712>

Version: Original manuscript: author's manuscript prior to formal peer review

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METEORITICS & PLANETARY SCIENCE

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Journal:	<i>Meteoritics & Planetary Science</i>
Manuscript ID	MAPS-3307.R1
Manuscript Type:	Article
Date Submitted by the Author:	20-Feb-2020
Complete List of Authors:	Collinet, M; Massachusetts Institute of Technology, Earth, Atmospheric and Planetary Sciences Grove, Timothy; MIT, Earth, Atmospheric and Planetary Sciences
Keywords:	Geothermometry, Melting experiments, ureilite(s) < Meteorite(s), Planetesimal
Note: The following files were submitted by the author for peer review, but cannot be converted to PDF. You must view these files (e.g. movies) online.	
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Incremental melting in the ureilite parent body: initial composition, melting temperatures and melt compositions

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Abstract:

Ureilites are carbon-rich ultramafic achondrites that have been heated above the silicate solidus, do not contain plagioclase, and represent the melting residues of an unknown planetesimal (i.e. the ureilite parent body, UPB). Melting residues identical to pigeonite-olivine ureilites (representing 80 % of ureilites) have been produced in batch melting experiments of chondritic materials not depleted in alkali elements relative to the sun's photosphere (e.g. CI, H, LL chondrites), but only in a relatively narrow range of temperature (1120-1180 °C). However, many ureilites are thought to have formed at higher temperature (1200-1280 °C). New experiments, described in this study, show that pigeonite can persist at higher temperature (up to 1280 °C) when CI and LL chondrites are melted incrementally and while partial melts are progressively extracted. The melt productivity decreases dramatically after the exhaustion of plagioclase with only 5-9 wt.% melt being generated between 1120 and 1280 °C. The relative proportion of pyroxene and olivine in experiments are compared to 12 ureilites, analyzed for this study, together with ureilites described in the literature to constrain the initial Mg/Si ratio of the UPB (0.98-1.05). Experiments are also used to develop a new thermometer based on the partitioning of Cr between olivine and low-Ca pyroxene that is applicable to all ureilites. The equilibration temperature of ureilites increases with decreasing Al_2O_3 and Wo contents of pyroxene and decreasing bulk REE concentrations. The UPB melted incrementally, at different $f\text{O}_2$, and did not cool significantly (0-30 °C) prior to its disruption. It remained isotopically heterogeneous but the initial concentration of major elements (SiO_2 , MgO , CaO , Al_2O_3 , alkali elements) was similar in the different mantle reservoirs.

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4 31 **1. Introduction**

5 32 Ureilites are the largest group of ultramafic achondrites (550 samples at the time of writing)
6
7 33 and the second largest group of achondrites after howardites-eucrites-diogenites. Most ureilites are
8
9 34 composed of olivine and pigeonite with a significant fraction of metal (<5 wt.%), graphite (~3
10
11 35 wt.%), and traces of sulfide concentrated along silicate grain boundaries. Fewer samples contain
12
13 36 orthopyroxene and augite instead of or in addition to pigeonite (e.g. Takeda et al. 1989; Goodrich
14
15 37 et al. 2001). With the exception of brecciated ureilites (i.e. polymict ureilites) and a single
16
17 38 monomict ureilite (Goodrich et al., 2016), they are completely devoid of plagioclase. Their bulk
18
19 39 composition is depleted in incompatible lithophile elements and chalcophile elements relative to
20
21 40 all chondrites.

22 41 While they were once described as ultramafic igneous cumulates (Berkley et al. 1980;
23
24 42 Goodrich et al. 1987), most ureilites are now recognized as residues of partial melting representing
25
26 43 the mantle of a planetesimal that lost abundant FeNi-sulfide eutectic melts (e.g. Warren et al. 2006;
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28 44 Goodrich et al. 2013a; Barrat et al. 2015) and silicate melts (Warren and Kallemeyn 1992; Scott
29
30 45 et al. 1993; Kita et al. 2004, Goodrich et al. 2007; Warren 2012; Barrat et al. 2016). The Ureilite
31
32 46 Parent Body (UPB) was violently disrupted while its internal temperature was still high (1150-
33
34 47 1300 °C; e.g. Takeda et al. 1989; Sinha et al. 1997; Goodrich et al. 2004; Downes et al. 2008;
35
36 48 Herrin et al. 2010). Because cooling was rapid following the destruction of the UPB (2-20 °C/h;
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38 49 Miyamoto 1985; Takeda et al. 1989; Goodrich et al. 2001; Herrin et al. 2010), the high temperature
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40 50 mineral assemblage has been preserved. Ureilites represent “quenched melting residues”, the only
41
42 51 ones in the meteorite record, and document how planetesimals melted in the early solar system.
43
44 52 However, the initial bulk composition of the UPB, the melting conditions and the composition of
45
46 53 the melts that were extracted have proved challenging to decipher. No chondritic sample
47
48 54 representing the primitive material has been found and remnants of the associated silicate melts
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50 55 are extremely scarce. They include feldspatic clasts in polymict ureilites (Ikeda and Prinz 2001;
51
52 56 Cohen et al. 2004; Goodrich et al. 2004; Kita et al. 2004), a single 24 g trachyandesitic clast
53
54 57 (Bischoff et al. 2014) and ambiguous pyroxene-rich samples bearing melt inclusions (Goodrich et
55
56 58 al. 2001, 2009; Rosén et al. 2019) or plagioclase (Goodrich et al. 2016).

57
58 59 It is clear that the UPB was initially isotopically heterogeneous and remained so during
59
60 60 partial melting until it was disrupted. Ureilites have heterogeneous oxygen isotope compositions
61
and form a trend of mass-independent fraction (i.e. slope 1 in the three-isotope plot) which

correlates with the Mg# ($\text{MgO}/(\text{MgO}+\text{FeO})$ in mol.%) of ferromagnesian silicates (Clayton and Mayeda 1988; Greenwood et al. 2017). The forsterite content of olivine cores (Fo, equivalent to the Mg#) varies between 74 and 96 with increasing ^{16}O contents. In addition, ureilites are characterized by heterogeneities in C (Barrat et al. 2017), Cr (van Kooten et al. 2017; Zhu et al. 2020) and noble gas isotopes (Broadley et al. 2020), which also appear to correlate with O isotope compositions.

The initial composition of the UPB in major elements has long been unknown. The abundance of graphite and the oxygen isotopes point to a connection with carbonaceous chondrites (Clayton and Mayeda 1988; Goodrich et al. 2015) while nucleosynthetic anomalies in Cr, Ti and Ni are more akin to ordinary chondrites and characteristic of the “non-carbonaceous group” (Warren 2011). We have recently described batch melting experiments of a representative spectrum of chondritic materials (LL, H, CI, CM, and CV) and shown that pigeonite-olivine residues resembling ureilites can be produced by melting high NaK# compositions such as CI, H and LL chondrites (50; with $\text{NaK\#} = (\text{Na}+\text{K})/(\text{Na}+\text{K}+\text{Ca})$ in atom.%). The UPB could not have been depleted in alkali elements because low NaK# compositions (CV and CM) produce orthopyroxene-olivine residues (Collinet and Grove 2020b).

Batch melting experiments of H, LL and CI chondrites produce pigeonite-olivine residues in a relatively narrow temperature interval, between 1120 and 1180 °C, while many ureilites are thought to have formed at 1200-1270 °C (e.g. Takeda et al. 1989; Sinha et al. 1997). Here, we investigate whether pigeonite can persist at higher temperature when CI and LL chondrites are melted incrementally and while partial melts are progressively extracted. Experimental results are compared to 12 ureilites that were analyzed for this study and ureilites described in the literature to constrain the initial composition of the UPB and its Mg/Si ratio in particular. Then, we develop a new thermometer based on the partitioning of Cr between olivine and Low-Calcium Pyroxene (LCP; orthopyroxene or pigeonite) that can be used to constrain the temperature of equilibration of all ureilites. The $D_{\text{Cr}}^{\text{oliv} - \text{LCP}}$ thermometer is used to distinguish between ureilites representing simple melting residues and ureilites representing cumulates or the products of more complex igneous processes. We compare the former (most pigeonite-olivine ureilites) to our experiments to constrain melting processes and estimate the composition of the last silicate melts that were in equilibrium with ureilites. We also use the new temperature of equilibration of ureilites to discuss

the thermal history of the UPB and the nature of the heterogeneity in fO_2 and oxygen isotopes of the UPB.

2. Methods

2.1. Experimental approach

All experiments were performed in the MIT Experimental Petrology Laboratory in a Molybdenum Hafnium Carbide pressure vessel (MHC-PV) placed in a Deltech vertical furnace following the same overall approach described in Walker and Grove (1993) and Singletary and Grove (2006). One key difference is that several starting compositions (~10 mg each) were equilibrated simultaneously in individual graphite capsules positioned vertically in a single Pt outer capsule. The temperature gradient is small at the base of the MHC-PV (< 3 °C over the length of the Pt capsule) and all experimental charges were equilibrated under the same P-T- fO_2 conditions. The outer Pt capsule was loosely crimped but not sealed to allow for equilibration with the CO pressure medium and set the fO_2 to the CCO buffer (French and Eugster 1965). In all but two experiments, the temperature was fixed for the duration of the experiments. In the other two experiments (CHS 46 and 71), the temperature was first raised 20-50 °C above the final temperature for two hours. After seven hours to five days, experiments were terminated by pulling the MHC-PV out of the furnace, inverting it and hitting it with a wrench to ensure that the capsule dropped into the water-cooled head of the pressure vessel and quench. Experiment conditions are summarized in Table 1.

This study is a complement to a larger series of melting experiments of chondritic materials (Collinet and Grove 2020a, 2020b). In previous manuscripts, we reported on experimental charges that contain strictly chondritic bulk compositions (H, LL, CI, CM and CV) and simulate the batch melting of planetesimals. In this study, we describe distinct experimental configurations that simulate incremental melting processes. The batch melt compositions of (Collinet and Grove 2020a) were mathematically subtracted from the CI and LL compositions to obtain melting residue analogs. These new compositions were synthesized by mixing high purity oxides, silicates (CaSiO₃), carbonates (Na₂CO₃, K₂CO₃) and Fe metal (Grove and Bence 1977). Starting materials were mixed for four hours in an automatic mortar and pestle and pre-conditioned for three days in a gas-mixing furnace at 1000 °C and QFM –1 (IW + 2.5). These starting compositions (CIR1, CIR2, CIR3 and LLR1, LLR2, Table 2) represent melting residues of either a LL or a CI

composition from which increasing fractions of batch melts were removed (11, 15.5, 19.5 wt.% and 8.5, 16 wt.%, respectively).

During incremental melting, successive or “instantaneous” melt compositions are expected to vary as melting progresses and become rapidly depleted in incompatible elements (e.g. K_2O , Na_2O) relative to batch melts. However, the composition of the aggregate melt (i.e. the sum of all melt increments) and the composition of the residue are virtually identical to the composition of the melt and residue produced by batch melting, after the same total degree of melting (e.g. see pMELTS simulations in the supplementary material; Ghiorso et al., 2002). Accordingly, CIR1-3 and LLR1-2 compositions can also be understood as the melting residues from which the melt was extracted as successive increments of any size. The LL and CI compositions were selected because, like the initial composition of the UPB (Collinet and Grove 2020b), they are not depleted in alkali-elements relative to the sun’s photosphere ($NaK\# = 50$). Yet, they are characterized by contrasting Mg/Si (0.92 and 1.03, respectively) and Ca/Si ratios (0.052 and 0.06). In addition, CI and LL compositions are relatively close in Mg# (68.5-70) to the most FeO-rich ureilites (74), after removal of sulfur as FeS. Two additional compositions, RCa and RCa2, represent arbitrary “near-chondritic” compositions depleted in Al_2O_3 , Na_2O and K_2O but enriched in CaO. They were included to produce two-pyroxene assemblages that could be compared to orthopyroxene-augite ureilites.

2.2. EPMA analyses

The JEOL-JXA-8200 SuperProbe electron probe micro analyzer (EPMA) of the MIT Electron Microprobe Facility was used to acquire quantitative analyses of experimental run products as well as olivine and pyroxene in 12 ureilites. Silicates were analyzed with a focused beam, a voltage of 15 kV and a current intensity of 10 nA. All elements were measured for 40 s (20 s for backgrounds) except Na, which was counted first for 8 s (4 s for backgrounds). In many experiments, the small quantity of melt (<5 wt.%) wetting grain boundaries, but rarely pooling, precluded reliable measurement of its composition (quenched-modified glass). When attempted, the analysis of experimental glasses was done with a 5 nA current intensity, a spot size of 2-4 μm and a counting time of 4 s for Na.

Elemental X-ray maps of Si, Mg, Fe, Ca and Al were acquired for 10 of the 12 ureilites. The maps were performed with a beam size of 5-10 μm , a step size of 15-25 μm , a voltage of 15

152 kV, a current intensity of 30 nA and a counting time of 40 ms. Elemental intensities were acquired
 153 simultaneously with five wavelength dispersive spectrometers (WDS).

154 **2.3. Calculation of the mode of ureilites and experiments**

155 Elemental X-ray maps were compiled with MultiSpec© and used to create automatically
 156 50 clusters of pixels in 5-dimensions with the algorithm ISODATA. Pixel clusters were then
 157 manually grouped into classes representing the different phases: (1) Fe-metal, sulfide and oxides,
 158 (2) olivine, (3) low-Ca pyroxene and, if present, (4) augite. Graphite, the residual porosity and all
 159 ambiguous clusters were grouped as background. The number of pixels in each class, representing
 160 the modal composition of ureilites in vol.%, were counted with ImageJ. This treatment was applied
 161 to the 10 ureilite thin sections mapped for this study and to eight additional ureilites studied by
 162 Singletary and Grove (2003) using their original X-ray maps.

163 The phase proportions in experiments were estimated by mass balancing the composition
 164 of the different phases against the bulk composition of the starting material with the function fitlm
 165 in Matlab©. However, because the chemical composition of most melts could not be analyzed, we
 166 first had to estimate their composition by altering the composition of experimental batch melts at
 167 the same temperature and fO_2 conditions (Collinet and Grove 2020a). The concentrations of SiO_2 ,
 168 Al_2O_3 and alkali elements were progressively lowered while the concentrations of CaO and, to a
 169 lesser extent, FeO and MgO were slightly increased. Five to ten multiple linear regressions were
 170 performed and the one with the lowest sum of squared residuals was selected. In practice, we find
 171 that as long as a melt is included in mass balance calculations, the relative proportions of olivine
 172 and pyroxene are not very sensitive to small variations in the estimated melt composition.
 173 However, when no melt is included, mass balances tend to overestimate the proportion of pyroxene
 174 relative to olivine by ~ 5 wt.% (or 5 py unit; where py is the ratio $px/(px+oliv)*100$ in wt.%) even
 175 in experiments that apparently contain only a few percent of liquid (e.g. LLR2 in CHS 47, 51).
 176 Finally, to confirm that this method provides reasonable estimates of py , we calculated phase
 177 proportions in vol.% by image analysis of 10 backscattered electron (BSE) images for two
 178 experimental charges: LLR2 in CHS 47 and CIR2b in CHS 63. Modal compositions in vol.% were
 179 converted in wt.% by multiplying the phase proportions by the densities of olivine and pyroxene
 180 of appropriate composition. The py calculated by images analyses are consistent (within ~ 3 py
 181 units) with the ones calculated by mass balance: 23.1 (vs. 21.2) and 42 (vs. 44.6) for CHS 47 and
 182 CHS 63, respectively.

3. Results

3.1. Experiments on chondritic residues

Most experimental charges performed using the LLR1-2 and CIR1-3 bulk compositions (chondritic residues) contain olivine, orthopyroxene or pigeonite, glass and metal (Fig. 1). Only one charge, CIR2 in CHS 65 (1201 °C, IW -2.3), has minor augite in addition to pigeonite. All crystalline phases are homogenous in composition. Experimental runs using CIR1 and LLR1, the least refractory residues, contain more melt than CIR2-3 and LLR2 runs at a given temperature. We calculate the aggregate melt fraction (i.e. the total amount of melt produced from the initial chondritic composition), by adding the melt fraction in CIR1-3 or LLR1-2 experiments to the melt fractions associated with the different starting compositions (Table 2). At a given temperature, aggregate melt fractions are significantly smaller (15-22 wt.%) in re-melted residues than in batch melting experiments (up to 35 wt.%; Fig. 2). The main effect of removing partial melts as melting progresses is to considerably lower the melt productivity at higher temperature.

Because melt removal slows down the melting process considerably, the pigeonite (i.e. LCP with Wo content greater than 5, when defined based on composition) can persist in experimental residues up to ~1280 °C, 80 °C higher than in batch melting experiments (Fig. 3). The bulk CIR2 composition corresponds to the residue immediately following the disappearance of plagioclase ($F=15.5$ wt.%; Collinet and Grove 2020a) and produces pigeonite with a high wollastonite content (Wo) at the highest temperature (1280 °C). LLR1-2 charges contain a pigeonite with lower Wo content and pigeonite disappears at ~1220 °C (as opposed to 1170 °C in batch melting experiments of H and LL compositions).

The pigeonite in LLR1-2 charges has a lower Wo content because py is significantly larger than in CIR1-3 charges (Fig. 4). The larger proportion of pyroxene relative to olivine in experimental charges with LLR1-2 compositions results from their lower bulk Mg/Si ratios (Table 2). In other words, in LLR1-2 charges, similar bulk CaO concentrations are “diluted” into a larger volume of pyroxene than in CIR1-3 charges. As melting progresses, pyroxene is consumed and py decreases along with the Wo content in the remaining pyroxene. However, at a given total extent of melting, there is ~ 20 wt.% less pyroxene in CI residues, with a higher Wo content, than in LL residues. The $Mg\#$ of olivine and pyroxene increases with decreasing fO_2 conditions from 75 at IW -1.3 to 95 at IW -2.5. In parallel, the fraction of Fe metal increases by 12 wt.% (Fig. S1).

Experimental charges with CaO-rich bulk compositions that do not correspond to known chondritic residues (DCa1 and DCa2) contain olivine, pigeonite, augite, glass and metal. No attempt was made to estimate the melt fractions in those experiments. They are used in section 4.3 and 4.7, together with other experimental charges, to constrain the temperature of equilibration of ureilites and the thermal history of the UPB. The detailed composition of experimental runs products are reported in the supplementary material (Table S1-S5).

3.2. Mineral composition and petrographic description of ureilites

Among the 12 ureilites analyzed for this study, eight are US Antarctic samples provided by the Meteorite Working Group. Several of them have been described to various extents in the literature: ALHA 82106 (Takeda et al. 1989), EET 96293 (Singletary and Grove 2003), LAP 03721 (paired to LAP 03587; Warren and Rubin, 2010), EET 90019 (Warren 2012). Four additional samples from Northwest Africa were acquired from private collectors and have not been previously described in detail. The phase compositions and the phase proportions are reported in Table 3 and the BSE images of six samples are shown in Fig. 5.

Three of the nine ureilites that we studied are orthopyroxene-augite-olivine ureilites (EET 96293, NWA 11754 and ALHA 82106), one is an orthopyroxene-pigeonite-olivine ureilite (NWA 5555) and the remaining eight are pigeonite-olivine ureilites. EET 90019 and NWA 4852 are some of the most FeO-poor pigeonite-olivine ureilites ($Fe_{89.4}$ and $Fe_{87.7}$, respectively). We observed a single 80- μ m pigeonite in section 15 (0.6 cm²) of DOM 08012. Chromite was only found in MIL 07447 as two 50- μ m spherical crystals: one zoned and enclosed in olivine and the other relatively homogenous and enclosed in pigeonite (Fig. S2). In addition to the grain boundary metal and sulfide, spherules of variable size (1-20 μ m), made of C-rich metal (possibly cohenite), C-poor metal, sulfides, and frequently all three, occur as inclusions in pigeonite and olivine (Fig. S2). Most samples also display the typical 10-50 μ m rims of MgO-rich olivine containing sub- μ m metal blebs and produced by reduction during the disruption of the UPB (i.e. “secondary smelting”, see section 4.8).

In four of the eight pigeonite-olivine ureilites (NWA 11755, LAP 03721, MIL 090076 and EET96042), the pyroxene has been extensively smelted (Fig. 5e-f), probably due to a brief episode of shock heating that followed the impact (Warren and Rubin 2010). In those samples, but not in the other four, olivines contain sub- μ m metal inclusions in their cores (Fig. S4). Unlike the metal-sulfide spherules mentioned in the previous paragraph, they are homogeneously distributed and

exclusively sub- μm . NWA 11755 (Fig. 5e) displays particularly clear evidence of this reduction process, including the same bimodal porosity affecting “relic” pigeonite cores and reduced FeO-poor pyroxene as in LAR 04315 (Warren and Rubin 2010). Because the pyroxene was almost completely smelted and is porous, we did not attempt to estimate the pre-impact phase proportions of NWA 11755. Unlike smelted rims, the core of olivine and pyroxenes are homogeneous in composition.

NWA 11754 has a large fraction of pyroxene, including abundant augite, and a large fraction of metal that fills fractures and silicate grain boundaries (Fig. 5d). NWA 11754 is in every way identical to Hughes 009 and other members of the “Hughes cluster”, a group of ureilites interpreted as igneous cumulates rather than melting residues (Goodrich et al. 2001, 2009, 2016). EET 96293 has the same Fo content in olivine as Hughes 009 and, while it contains less augite than Hughes 009/NWA 11754, it contains a similar fraction of total pyroxene (Fig. 6).

ALHA 82106 is a unique orthopyroxene-augite-olivine ureilite that contains wavy exsolution lamellae of augite in orthopyroxene (Fig. 5c). Because ALHA 82106 also contains coarse-grained augite, its silicate phase assemblage at peak temperature is believed to have been augite-pigeonite-olivine (Takeda et al. 1989).

The element maps used to calculate the modal composition of ten of the twelve ureilites and eight of the ureilites analyzed by Singletary and Grove (2003) are available in the supplementary material. All but three ureilites form an overall trend of increasing py with Fo in olivine (Fig. 6; $R^2 = 0.74$), similar to the py -Fo trends previously documented by Singletary and Grove (2003) and Goodrich et al. (2007). Only the two anomalous “Hughes cluster” samples and MET 01083, which displays a poikilitic texture (Fig. S11), were excluded. The trend formed by ureilites in Fig. 6 has the same slope as residues with constant Mg/Si ratios, having lost the same aggregate melt fraction at various fO_2 . It is also parallel to similar trends formed by ordinary chondrites and other groups of primitive achondrites (Collinet and Grove 2020b).

4. Discussion

4.1. The initial composition of the UPB

Ureilites have long been thought to be related to carbonaceous chondrites due to their high graphite content. In support of this view, Clayton et al. (1976) and Clayton and Mayeda (1988) showed that ureilites form a trend of mass-independent fractionation in three-isotope plots and are coarsely aligned with CV chondrites (Greenwood et al., 2017). However, the discovery of

nucleosynthetic anomalies in Cr, Ni and Ti isotopes in bulk ureilites has questioned the connection between ureilites and carbonaceous chondrites (Qin et al. 2010; Warren 2011). On a $\Delta^{17}\text{O} - \epsilon^{54}\text{Cr}$ diagram, ureilites, which have higher concentrations of neutron-rich isotopes (e.g. ^{54}Cr and ^{50}Ti) relative to carbonaceous chondrites, are part of the “non-carbonaceous” (NC) group along with enstatite and ordinary chondrites. Goodrich et al. (2015) inferred that ureilites still formed in the same region of the nebula as carbonaceous chondrites (greater heliocentric distance) but at a time when nucleosynthetic anomalies were distinct (~ 0.5 Ma after CAIs). However, the NC and CC reservoirs are thought to have been present and isolated from each other very early (< 1 Ma after CAIs), presumably by a proto-Jupiter, based on the Mo isotopes of NC and CC iron meteorites (Kruijer et al. 2017). If this is true, and if the UPB accreted relatively late (1.4-1.7 Ma after CAIs) as suggested by Budde et al. (2015), then the UPB would have accreted within the orbit of proto-Jupiter together with the other NC parent bodies. However, the accretion time of the UPB is still debated and van Kooten et al. (2017) and Zhu et al. (2020) argued that the UPB was partly differentiated earlier than 1 Ma after CAIs and (as early as 0.6 Ma ΔCAIs), an older age compatible with the thermal evolution models of Wilson et al. (2008) and Goodrich et al. (2015). Based on a presumed relationship between Si isotopes and the Mg/Si ratio of parent bodies, Dauphas et al. (2015) calculated a value of 1.00 ± 0.08 for the UPB, but the uncertainty overlaps with both carbonaceous and ordinary chondrites. In summary, at present, isotopic systematics do not provide strong constraints on the initial major element composition of the UPB.

Using MAGPOX (Longhi 1991), Goodrich (1999) and Goodrich et al. (2007) showed that superchondritic $\text{CaO}/\text{Al}_2\text{O}_3$ ratios were required to stabilize pigeonite in ureilites. They extensively discussed whether this could represent a primary feature of the UPB. Other workers have commented this issue and the consensus that emerged is that the superchondritic $\text{CaO}/\text{Al}_2\text{O}_3$ ratios resulted from parent body processes rather than nebula processes (Kita et al. 2004; Warren 2012; Goodrich et al. 2013c). In a recent experimental study (Collinet and Grove 2020b), we show that the UPB was initially characterized by a chondritic $\text{CaO}/\text{Al}_2\text{O}_3$ and only became depleted in Al_2O_3 following the extraction of low-degree silicate melts. We also show that the initial composition of the UPB was not depleted in alkali elements relative to the sun’s photosphere ($\text{NaK}\# = 50$). All high $\text{NaK}\#$ chondritic compositions (H, LL, CI) produce pigeonite-olivine residues upon melting, contrary to CM (33) and CV (25) compositions. Therefore, the assumption that the precursor material of FeO-poor ureilites contained more CAIs than the one of FeO-rich ureilites (Singletary

and Grove, 2006; Goodrich et al. 2013a) appear to be no longer justified. Any significant addition of CAIs to FeO-poor ureilites would drive down the NaK# towards the value of CM (33) and CV chondrites (25), which do not form pigeonite-olivine residues by partial melting.

The new incremental melting experiments and additional analyses of ureilites performed for this study can be used to constrain the initial Mg/Si ratio of the UPB in more detail. Warren (2011) argued that, despite being carbon-rich, ureilites could derive from “non-carbonaceous” chondritic materials with Mg/Si ratios resembling ordinary chondrites (0.92-0.95) but lower Fe/Si ratios. Goodrich (1999) also concluded that the initial Mg/Si ratio was close to the one of ordinary chondrites and Goodrich and Wilson (2013) proposed that it was even lower, close to the Mg/Si ratio of enstatite chondrites (0.74-0.88).

The olivine and pigeonite of CIR2 experimental charges are identical, both in terms of chemical composition and relative proportions (*py*), to the olivine and pigeonite in ureilites (compare Fig. 4 to Fig. 6). On the other hand, LLR1-2 experimental charges contain an overabundance of low-Ca pyroxene, with relatively lower Wo contents (4-6.5) compared to ureilites (up to Wo₁₂). Therefore, the Mg/Si ratio of the LL starting composition (0.92) places a firm lower limit on the initial Mg/Si ratio of the UPB. Any composition with a lower ratio would contain no pigeonite at all (Wo < 5). The Mg/Si ratio of the CI composition (1.03) is likely much closer to the average initial ratio of the UPB. The positive correlation between the Fo content in olivine and the *py* of ureilites displays some scatter that allows for a certain degree of heterogeneity in the initial Mg/Si ratio (i.e. 0.98-1.05). However, the slope of the Fo-*py* trend, which is identical to experimental trends of constant aggregate melt fraction, suggest that, if present, slight variations in the Mg/Si ratio were independent of the Fo content in olivine. Therefore, ureilites with Fo₇₅ and Fo₉₅ olivine were characterized by the same overall initial Mg/Si ratio.

If the initial Fe/Mg of the UPB had been homogenous, high-Fo ureilites, that formed under more reducing conditions (IW -2 / -2.5) should also contain more metal, unless some of the metal was extracted from the residues along with FeS (Barrat et al. 2015), in which case highly siderophile element concentrations (HSE) would be expected to be lower (Warren and Huber 2006; Warren et al. 2006). Ureilites with Fo₈₅ and Fo₉₅ olivine should contain 6 wt.% and 12 wt.% more Fe⁰ than ureilites with Fo₇₅ olivine (Fig. S1). Because neither metal fractions nor siderophile element concentrations correlate with Fo content in olivine, the UPB is often thought of as initially heterogenous in Fe/Mg. However, the siderophile element concentrations of ureilites are still not

well understood despite extensive studies. It remains possible that the UPB was heterogenous in siderophile elements or that a fraction of exogenous metal overprinted siderophile element concentrations (Goodrich et al. 2013a; Goodrich and Desch 2019).

4.2. Literature estimates of ureilite equilibration temperatures

Estimating the temperature of equilibration (T_E) of individual ureilite samples has been a central issue in interpreting their petrogenesis. As most ureilites only contain pigeonite and olivine, two-pyroxene (Lindsley 1983; Lindsley and Andersen 1983; Sack and Ghiorso 1994) and olivine-chromite thermometry (Evans and Frost 1975; Wlotzka 2005) can only be applied to relatively small sub-groups of ureilites. Most two-pyroxene temperatures that have been calculated for orthopyroxene-augite or pigeonite-orthopyroxene ureilites are in the range 1200 – 1270 °C (Takeda 1989; Takeda et al. 1989; Sinha et al. 1997; Chikami et al. 1997; Weber et al. 2003) while olivine-chromite temperatures are much lower 1040 – 1060 °C (Goodrich et al. 2014).

Singletary and Grove (2003) made the first attempt at estimating the temperature of pigeonite-olivine ureilites by calibrating an empirical thermometer based on the MgO and CaO concentrations of pigeonite. Our batch and incremental melting experiments of chondritic compositions can be used to evaluate its utility. Out of 22 experimental charges containing olivine, pigeonite, metal and melt, only five have a calculated temperature within ± 20 °C of the experimental temperature. Five experiments have a calculated temperature within ± 50 °C and the last ten experiments are within ± 100 °C. The pigeonite thermometer of Singletary and Grove (2003) overestimates the actual temperatures of most of our experiments (Fig. S8). Pyroxene phase diagrams (Lindsley and Andersen 1983; Sack and Ghiorso 1994), previous experiments on basaltic compositions (Grove and Juster 1989) and the experiments of this study show that the minimum temperature at which pigeonite is stable increases with the Mg#. The maximum temperature of pigeonite stability (i.e. melting temperature in simple systems) is also a function of the Mg#: 1409 °C for a pigeonite $En_{90}Wo_{10}$ but probably ~ 1350 °C for the pigeonite of FeO-rich ureilites ($En_{74}Wo_{10}$; Presnall 1995; Warren and Rubin 2010). Therefore, if permitted by the bulk composition of the system, pigeonite with given MgO and CaO concentrations (or En and Wo contents) can be stable over a temperature interval of 150-250 °C. The apparent temperature dependence of MgO and CaO concentrations in pigeonite used by Singletary and Grove (2003) reflected the restricted temperature and compositional range of the melts produced in earlier experiments. The UPB was initially rich in alkalis (Kita et al. 2004; Bischoff et al. 2014; Collinet

and Grove 2020b) but the absence of Na₂O in ureilite pyroxenes did not allow Singletary and Grove (2003) to include this important element in the calibration of their single-pyroxene thermometer. Therefore, the thermometer of Singletary and Grove (2003) should be replaced by the thermometer described in the next section.

4.3. New thermometer based on the partitioning of Cr between olivine and low-Ca pyroxene

The compatibility of Cr in olivine and LCP strongly decreases with increasing temperature in batch melting experiments of chondrites (Fig. 7 a-b). At 1100 °C, the Cr₂O₃ content of olivine is less than half of the Cr₂O₃ content of coexisting LCP. However, the $D_{Cr}^{LCP-liq}$ (Cr₂O₃^{LCP}/Cr₂O₃^{liq} in wt.%) decreases faster with increasing temperature than the $D_{Cr}^{oliv-liq}$ and the Cr₂O₃ content of olivine becomes nearly identical to that of the coexisting LCP at 1300 °C. Therefore, with increasing temperature, the ratio of $D_{Cr}^{oliv-liq}$ and $D_{Cr}^{LCP-liq}$ (i.e. $D_{Cr}^{oliv-LCP}$) increases with increasing temperature and follows a linear relationship (Fig. 7 c-d). The $D_{Cr}^{oliv-liq}$, $D_{Cr}^{LCP-liq}$ and $D_{Cr}^{oliv-LCP}$ do not seem to be influenced by the fO_2 over the range of IW -1.3 and IW -2.5 (Fig. 7c).

Because the melt does not have to be analyzed to calculate the $D_{Cr}^{oliv-LCP}$, we can use all of our experimental charges, including residues with only a few percent of melt, to calibrate a $D_{Cr}^{oliv-LCP}$ thermometer. In practice, we use a subset of 60 charges for which Cr₂O₃ contents were the most accurately determined (Table S6). The 60 experimental charges include all ten starting compositions (Table 1): H, LL and CI (31), LLR1-2 and CIR1-3 (23) and RCa1-2 (6). All experimental charges, whether they contain one LCP (orthopyroxene or pigeonite) or one LCP and augite, visible traces of chromite, and regardless of the experimental fO_2 fall on the same linear trend with an accuracy of ±15 °C (Fig. 7 c-d). The $D_{Cr}^{oliv-LCP}$ also appears to be largely independent of the wollastonite content in LCP (Fig. 7d). Nonetheless, a few experiments with very low Wo content in pyroxene (<3), not observed in ureilites, could be characterized by slightly higher $D_{Cr}^{oliv-LCP}$ and were not included in the linear regression.

The independence of the $D_{Cr}^{oliv-LCP}$ to fO_2 is not intuitive because the average valence state of Cr decreases with decreasing fO_2 . Most Cr is present as Cr³⁺ at QFM +1 (~IW +4.5) but Cr²⁺ predominates at conditions more reducing than IW -1 (Hanson and Jones 1998; Papike et al. 2005) and Cr⁰ becomes the dominant oxidation state under IW -5/-6 (Holzheid and O'Neill 1995). Cations with contrasting charge and size such as Cr²⁺ and Cr³⁺ would be expected to partition

differently in the M1 and M2 octahedral sites of the olivine and pyroxene crystal structures. Presumably, this is not the case in olivine, for which the partition coefficients $D_{\text{Cr}^{3+}}$ and $D_{\text{Cr}^{2+}}$ appear to be identical and largely independent of the $f\text{O}_2$ (Papike et al. 2005), as suggested by our experimental results. In pyroxene, Cr^{3+} should fit easily in the smaller M1 site and can be charged balanced by Al^{3+} in the tetrahedral site (Tschermak substitution) while Cr^{2+} would only fit in the larger M2 site. Therefore, $D_{\text{Cr}}^{\text{oliv} - \text{LCP}}$ could be expected to decrease with $f\text{O}_2$, a behavior that has been shown experimental between IW +1 and IW -1 (Karner et al. 2007) but that should become negligible under IW -1 as most Cr is already in the Cr^{2+} state, as suggested by our experiments (Fig. S6). The predominance of Cr^{2+} in ureilites is also indicated by XANES analyses (calculated valence of 2.0-2.2; Goodrich et al., 2013b; Sutton et al., 2017). In summary, the $D_{\text{Cr}}^{\text{oliv} - \text{LCP}}$ thermometer can be used at conditions more reducing than IW -1 without any $f\text{O}_2$ dependent term, as the influence of $f\text{O}_2$ is undetectable. The $D_{\text{Cr}}^{\text{oliv} - \text{LCP}}$ thermometer is specifically designed for ureilites and is likely not directly applicable to more oxidizing systems in its current form.

The $D_{\text{Cr}}^{\text{oliv} - \text{LCP}}$ thermometer can be used to calculate the temperature of equilibration (T_E) of all ureilites, regardless of their mineral assemblage, as long as they contain olivine and at least one LCP. It can be directly applied to 76 samples with detailed EPMA analyses of both olivine and LCP (Table S8). Their T_E cover a large range of temperature, 1054-1276 °C, which is identical to the range of previous T_E estimates (section 4.2). However, the $D_{\text{Cr}}^{\text{oliv} - \text{LCP}}$ T_E offer a new highly reliable representation of the temperature distribution in the UPB and provide important new constraints on the petrogenesis of ureilites.

Ureilites with low Fo content in olivine (Fo_{75-80}) have the most contrasting T_E (1054-1276 °C) while ureilites with high Fo content in olivine (Fo_{90-95}) have more restricted and higher T_E of 1180-1250 °C (Fig. 8). Histograms of the T_E of ureilites with either Fo_{74-85} or Fo_{87-96} olivine display overlapping modes at 1180-1220 °C. In other words, FeO-poor and FeO-rich ureilites are characterized by identical average T_E and eight of the nine samples with T_E higher than 1240 °C are FeO-rich. This result is in stark contrast with the positive correlation between T_E and olivine Fo contents (i.e. Mg#) that was predicted by the pigeonite thermometer of Singletary and Grove (2003). Because the new T_E no longer correlate with the Mg# of ureilite samples, a strict separation between FeO-poor and FeO-rich ureilites appears arbitrary. In the remainder of the discussion, these two groups should be thought of as the end members of a continuum.

All ureilites with two pyroxenes show a $D_{Cr}^{oliv-LCP}$ equilibration temperature that is lower than the minimum temperature of pigeonite stability of Sack and Ghiorso (1994). The few ureilite samples containing euhedral chromite, such as NWA 766, LEW 88774, and NWA 3109, display low olivine-chromite temperatures (1037, 1042 and 1058 °C; Goodrich et al. 2014) and low $D_{Cr}^{oliv-LCP}$ temperatures (1054, 1085 and 1141 °C) that correlate with the Cr# (i.e. Cr/(Cr+Al) in mol.%) of chromites: 0.65, 0.71 and 0.76, respectively.

4.4. Anomalous low-temperature ureilites

Ureilites containing orthopyroxene and augite, characterized by low T_E (open symbols in Fig. 8), display poikilitic textures with large (up to 15 mm) orthopyroxene oikocrysts enclosing rounded olivine and augite crystals: LEW 88774 (Chikami et al. 1997), MET 78008 and Y 74130 (Takeda et al. 1989), RaS 517 (Rosén et al. 2019) and HaH 064 (Weber et al. 2003). Many such samples contain unusually large fractions of pyroxene (e.g. LEW 88774, the “Hughes cluster” samples, RaS 517) or, more rarely, unusually little pyroxene (MET 01083). Those anomalous low-temperature ureilites are the only ones that do not plot on the trend of increasing py with increasing Fo content in olivine (Fig. 6b). In addition, the Al_2O_3 concentrations of their LCP are significantly higher (1.3-2.2 wt.%) than in other ureilites (0.3-1.0 wt.%; Fig 9a). The “Hughes cluster” samples traditionally contain a large fraction of augite (> 30 vol.%) and their bulk Ca/Mg ratios are superchondritic (Goodrich et al. 2001, 2009).

Due to their large pyroxene fractions and poikilitic textures, some anomalous ureilites have been interpreted as cumulates or older residues that reacted with silicate melts (Downes et al. 2008; Goodrich et al. 2009). All of the aforementioned samples are characterized by lower T_E than the temperature of plagioclase disappearance in chondritic materials (Fig. 8). If they represented simple melting residues quenched at peak temperature following the destruction of the UPB, they would be expected to contain plagioclase. Yet, only one sample contain plagioclase (Goodrich et al. 2016). This sample, equilibrated at 1144 °C, is part of the Hughes cluster. It contains a plagioclase much more calcic ($An_{68.4}$) than the plagioclase of the melting residues of high NaK# chondrites (An_{20-40} ; Collinet and Grove 2020a).

Collectively, those observations are consistent with the idea that all anomalous low-temperature samples do not represent melting residues but cumulates or products of melt-rock reactions during melt migration. RaS 517 was recently described as containing primary melts and recording the ureilite anatexis (Rosén et al. 2019). Due to its very low T_E (1072 °C) and high augite

fraction, this scenario does not seem viable. Instead, the silicate melt could represent a residual liquid crystallizing in a superficial intrusion or magma conduit. Some brachinite-like meteorites display similar coarse poikilitic textures and are inferred to form by partial crystallization of the melt during melt migration (Goodrich et al. 2017a).

4.5. Temperatures and extents of melting recorded by pigeonite-olivine ureilites

The anomalous samples described in the previous section are in part responsible for the once popular view that ureilites were igneous cumulates (Berkley et al. 1980; Goodrich et al. 1987). However, since the 1990s, pigeonite-olivine ureilites have largely been regarded as melting residues (Warren and Kallemeyn 1992; Scott et al. 1993). If FeO-rich pigeonite-olivine ureilites (Fo₇₆₋₈₂), which represent 70-80 % of all ureilites, are melting residues from a relatively homogeneous chondritic material, the samples that equilibrated at high temperature should have experienced larger extents of melting than the samples that equilibrated at lower temperature. The chemical composition and petrologic features of pigeonite-olivine ureilites should correlate with T_E. Several such correlations are indeed observed. In ureilites with Fo₇₆₋₈₂ olivine, the Al₂O₃ content of LCP decreases linearly with increasing T_E from 1 wt.% at 1150 °C to 0.35 wt.% at 1276 °C (Fig. 9b). The Wo content in LCP decreases in parallel, albeit with more scatter, from Wo_{10.5-12} to Wo_{4.5-5} (Fig. 9c). The variability of Wo contents at a given temperature could result from the interaction of migrating CaO-rich “late stage melts” with melting residues (see next section). Finally, Barrat et al. (2016) have recently analyzed the rare earth element (REE) concentrations of a large number of ureilites. They identified two main patterns: (1) “group B” with steeper LREE profiles and high HREE concentrations, representing MgO-rich ureilites (Fo₈₉₋₉₅), and (2) “group A” with less steep LREE profiles and more variable bulk REE depletions, representing FeO-rich ureilites. The REE concentrations, as well as the Zr concentrations of the ureilites of “group A” (bulk leachates) correlate negatively with T_E (Fig. 10).

The ureilites of “group B” (Fo₈₉₋₉₅ olivine) could represent the melting residues, produced under more reducing conditions, of a chondritic material with initial NaK# and Mg/Si ratios similar to the chondritic material that produced FeO-rich ureilites (section 4.1). In addition, FeO-poor ureilites were equilibrated over the same range of temperature as FeO-rich ureilites (1180-1220 °C; Fig. 8). Because the melting temperature of ferro-magnesian silicates increases with the Mg#, FeO-poor ureilites should represent residues that melted to a lower extent than the FeO-rich

ureilites equilibrated at the same temperature. A lower bulk degree of melting could explain in part the higher REE concentrations of group B ureilites (Fig. 10).

4.6. Incremental melting and composition of “late-stage” silicate melts

As discussed in section 4.1, a chondritic composition with a high NaK# (50) and a relatively homogenous Mg/Si ratio (0.98-1.05) can produce ureilite-like residues by partial melting (F=15-24 wt.%) under variable fO_2 conditions. In batch melting experiments, pigeonite melts out at 1170-1200 °C while it persists up to 1280 °C in incremental melting experiments. The ureilite samples with the highest T_E (Goalpara, 1276 °C, and Dingo Pup Donga, 1267 °C) have a LCP with low Wo content (4.7-4.6, i.e. orthopyroxene). However, many FeO-rich samples with T_E between 1240 and 1260 °C contain Wo_{6.5-10} pigeonite: RC 027, Y 74123, NWA 11755, Y 82100 and GRO 95575 (Table S8). Residues containing pigeonite (Wo_{6.5-10}) at 1250 °C cannot form by batch melting of any chondritic material (Fig. 3) and, instead, suggest that silicate melts were extracted in several steps (i.e. incremental melting).

The composition of the first silicate melts produced in ureilites have been constrained by batch melting experiments of high NaK# chondritic materials (Collinet and Grove 2020a). Low-degree partial melts (< 15 wt.%), rich in alkali elements, Al₂O₃ and SiO₂ were likely extracted from their source and formed the ALM-A trachyandesite (Bischoff et al. 2014) and the albite/oligoclase-rich clasts of polymict ureilites (Ikeda and Prinz 2001; Cohen et al. 2004; Kita et al. 2004). If pigeonite-ureilites melted incrementally, the melts that were produced in later melting stages would have been radically different in composition from the melts of batch melting experiments. Because the melt productivity drops significantly following the extraction of alkali-rich melts (~5 wt.% between 1120 and 1250/1280 °C; Fig. 2), ureilites would have contained very little melt at the time of the destruction of the parent body. Similarly, LLR2 and CIR2-3 experimental charges contain very little melt (2-5 wt.%) that could not be analyzed successfully. These “late-stage” melts are expected to be depleted in alkali elements relative to batch melts. Following the disappearance of plagioclase, Na₂O concentrations decrease rapidly with successive increments of melting (see pMELTS simulations in supplementary material).

Pigeonite-olivine ureilites with Fo₇₅₋₈₂ olivine and high T_E (1200-1260 °C) have the highest concentrations of CaO in olivine (0.36-0.44; Fig. 11b). The partition coefficient of CaO between olivine and liquid ($D_{Ca}^{oliv-liq} = CaO_{oliv}/CaO_{liq}$) decreases with increasing temperature (Fig 11a), and can be parametrized to calculate the CaO content of the late-stage melts that were in equilibrium

with specific samples (Fig. 11c). At low temperature (<1170 °C), the CaO content of the melts in equilibrium with ureilites were identical to batch melts of high NaK# chondrites (H, LL and CI). However, at higher temperature, the late-stage melts were increasingly CaO-rich (8-12 wt.%; Fig. 11d) while the CaO content of batch melts plateaus at 7-8 wt.% and then decreases from 1220 °C. The high CaO content of late-stage melts cannot result from batch melting and supports an origin of ureilites as residues of incremental melting.

The Al_2O_3 content of late-stage melts could be estimated based on the $D_{\text{Al}}^{\text{LCP} - \text{liq}}$. In practice, due to the low Al_2O_3 content of pyroxene in experiments, the $D_{\text{Al}}^{\text{LCP} - \text{liq}}$ is difficult to calculate precisely. In batch melting experiments Collinet and Grove (2020a), and the experiments of Singletary and Grove (2006), the $D_{\text{Al}}^{\text{LCP} - \text{liq}}$ varies between 0.04 and 0.07 at 2-13 MPa for a pigeonite with Wo_{6-10} but we could not identify systematic variations. In Collinet et al. (2015), the $D_{\text{Al}}^{\text{LCP} - \text{liq}}$ strongly increases with pressure but is constant over a 150 °C temperature interval (i.e. 0.11 at 0.5 GPa, 0.2 at 1.0 GPa and 0.3 at 2.0 GPa). Therefore, we estimate the Al_2O_3 concentrations of late-stage melts with a constant $D_{\text{Al}}^{\text{LCP} - \text{liq}}$ of 0.055 and find that, within the ± 20 wt.% relative uncertainty, they are indistinguishable from the Al_2O_3 concentrations of batch melts. Our best estimate of the composition of a late-stage melt, produced at 1240 °C by incremental melting and in equilibrium with olivine Fo_{79} , is reported in Table 4. The MgO content is estimated as a function of the temperature and is used to constrain the FeO content, assuming a $K_{\text{D, Fe} - \text{Mg}}^{\text{oliv} - \text{liq}}$ of 0.31, from batch melting experiments.

The Na_2O content of late stage melts is more difficult to constrain. Following the disappearance of plagioclase, Na behaves as an incompatible element and is rapidly exhausted from the residue. Therefore, the Na_2O content of the melts produced at this stage was likely variable and decreased rapidly (from ~ 2 to 0 wt.% Na_2O). Late-stage melts such as the one reported in Table 4 could easily have crystallized the labradorite-rich clasts observed in polymict ureilites (Cohen et al. 2004; Kita et al. 2004; Goodrich et al. 2017b). With the algorithm MELTS (Ghiorso and Sack 1995), equilibrium and fractional crystallization of a late-stage melt containing 1 wt.% Na_2O produce ~ 20 -30 wt.% labradorite plagioclase (An_{50-70}). This is also consistent with the composition of plagioclase (An_{68}) in one Hughes cluster sample and with the idea that this sample represents an olivine-rich residue in which a silicate melt migrated and partly re-crystallized (Goodrich et al. 2016). Cumulate-residue hybrids have occasionally been advocated to explain the origin of ureilites under the term “paracumulates” (Warren and Kallemeyn 1989; Goodrich et al.

2004). The Hughes cluster samples and other anomalous low-T ureilites rich in augite could represent such lithologies and derive from the crystallization of CaO-rich and alkali-poor late stage melts.

The behavior of melting of the UPB has been disputed based on differing interpretations of REE patterns (Warren and Kallemeyn 1992; Goodrich et al. 2007, 2013c; Warren and Huber 2006; Warren 2012). Most recently, Barrat et al. (2016) used REE melting models to argue that ureilites formed by continuous/dynamic melting, a specific case of incremental melting during which a fixed melt fraction, referred to as the critical melt fraction, is retained in the residue. We reproduced their calculations using the modal proportions and melting coefficients of Collinet and Grove (2020a) and find that the REE patterns of FeO-rich ureilites can be explained by 15-24 wt.% of continuous melting with a 2 wt.% critical melt fraction (Fig. 12a), a range that is identical to the aggregate melt fractions of experimental charges containing ureilite-like residues (section 4.1, Fig. 4). In agreement with their more restricted T_E , FeO-poor ureilites can be explained by 16-18 wt.% of continuous melting. In detail, REE melting models are non-unique and the REE patterns can be reproduced by different incremental melting processes, such as the complete removal of larger melt increments (e.g. 5-10 wt.%; Fig. S10). On the other hand, near-fractional melting would produce stronger depletions in LREE (Fig. 11c), unless melting was faster than LREE diffusion in pyroxene (Goodrich et al. 2007). Because all ureilites represent melting residues that melted enough to exhaust plagioclase ($F > 15.5$ wt.%), and because REE melting models produce model-dependent results (Fig. S10), it is impossible to constrain uniquely the size of the melt increments. We infer that they were in the range 2-5 wt.%.

4.7. Peak temperatures of ureilites and thermal history of the UPB

Ureilites are sometimes described as having a two-stage cooling history: an initial episode of slow-cooling followed by an episode of rapid-cooling simultaneous with the destruction of the UPB (e.g. Goodrich 1992). It could be argued that the T_E of ureilites, estimated with the $D_{Cr}^{oliv-LCP}$ thermometer, does not represent the peak temperature. Cr diffuses relatively fast in olivine and in orthopyroxene. Diffusion coefficients are only one order of magnitude slower than the ones of Fe-Mg interdiffusion (Ito and Ganguly 2006; Dohmen et al. 2007; Ganguly et al. 2007). If main group ureilites had stopped melting and started cooling following the extinction of ^{26}Al , the $D_{Cr}^{oliv-LCP}$ thermometer would have recorded a lower final temperature rather than the peak melting temperature. However, the cores of olivine and pyroxene display homogeneous Cr_2O_3 contents.

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3 582 Either slow cooling did not occur or ureilites systematically and completely re-equilibrated at an
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5 583 intermediate temperature. Because olivine and pyroxene cores are also homogenous in Fe, Mg and
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7 584 Ca, two-pyroxene thermometers should record the same final temperature of equilibration. Only a
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9 585 handful of samples have actually been suggested to record two distinct temperatures of
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11 586 equilibration and, therefore, a slow-cooling event. In the next paragraphs, we use our experiments,
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13 587 the new $D_{Cr}^{oliv-LCP}$ thermometer and the phase diagrams and two-pyroxene temperatures of Sack
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15 588 and Ghiorso (1994) to re-evaluate whether some ureilites, and more generally the UPB, went
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17 589 through a stage of slow cooling.

17 590 As long as pigeonite-olivine ureilites remained in the field of pigeonite stability, a stage of
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19 591 slow-cooling could be undetectable. However, many pigeonite-olivine ureilites with $Wo_{6.5-10}$ were
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21 592 last equilibrated at 1240-1260 °C, a temperature which is close to the highest temperature at which
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23 593 we observe pigeonite ($Wo_{5.5-7}$) in incremental melting experiments (1274 °C). At higher
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25 594 temperature (1300 °C), melting residues become extremely CaO-poor and pigeonite is replaced by
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27 595 orthopyroxene. If ureilites had cooled slowly over a significant interval of temperature (from 1300
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29 596 to 1240 °C), they would have contained less CaO and orthopyroxene would have replaced
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31 597 pigeonite. In addition, if the final T_E had been significantly lower than the peak temperatures, the
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33 598 correlation of T_E with bulk REE concentrations and Al_2O_3 contents in pyroxene might have been
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35 599 erased. This is not the case.

36 600 Only one ureilite displays inverted pigeonite with augite lamellae in a orthopyroxene host:
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38 601 ALH 82106 (Takeda et al. 1989; Fig. 5c). This unique sample has a high T_E of 1245 ± 15 °C, just
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40 602 under the minimum temperature of pigeonite stability (see symbol 22 in Fig. 8). One of our
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42 603 experiments (CHS 66; RCa1) performed at 1250 °C contains olivine ($Fo_{95.3}$ vs. $Fo_{95.5}$ in ALH
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44 604 82106), augite and a pigeonite that is almost identical to the bulk unmixed pyroxene of ALH 82106
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46 605 (opx+augite lamellae; $Wo_{10.5}$, Table S7). This experiment suggests that ALH 82106 reached its
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48 606 peak temperature at 1250 ± 10 °C and only had to cool by a few to 20 °C to form inverted pigeonite.
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50 607 The large majority of ureilites that equilibrated below the minimum temperature of pigeonite
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52 608 stability contain distinct, randomly orientated, orthopyroxene and augite crystals. We posit that if
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54 609 those ureilites had cooled slowly by more than 0-20 °C below their peak temperatures many more
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56 610 samples should instead contain inverted pigeonite.

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58 611 To our knowledge, only one other ureilite displays clear signs of slow cooling: the highly
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60 612 anomalous sample LEW 88774 (Chikami et al. 1997), which contains coarse ~50 µm intergrown

lamellae of orthopyroxene and augite. Based on the bulk composition of the pyroxene (Wo_{15-20} ; sub-calcic augite), Chikami et al. (1997) argued that the pyroxene progressively recrystallized during slow cooling, starting at 1280 °C. The model of Sack and Ghiorso (1994), which agrees closely with our experimental results (Fig. S5), suggests that a bulk pyroxene with a composition of $En_{66}Wo_{15}$ is stable at 1180-1200 °C as opposed to 1280 °C (Chikami et al. 1997). LEW 88774 might have cooled slowly from 1200 °C to 1100 °C, the $D_{Cr}^{oliv-LCP}$ temperature. Because LEW 88774 is a highly unusual pyroxenite ($py = 76$) it could represent a cumulate formed within an outer layer of the UPB and its thermal history is likely not representative of the conditions recorded by most ureilites in the deeper interior of the UPB.

Several other “anomalous low-temperature ureilites” (section 4.4), are thought to have equilibrated at two distinct temperatures. For example, Goodrich et al. (2001) suggest that Hughes 009, containing orthopyroxene and augite (Mg# of 88-89), cooled from 1250 °C to 1050 °C. One of our experiments places an upper limit to the peak temperature of Hughes 009. CIR1-CHS 48 contain pigeonite (Mg# 88) at 1194 °C, which is close to the minimum temperature of pigeonite stability at that Mg# (1190 °C; Sack and Ghiorso 1994). Because Hughes 009 contain no exsolution lamellae but “standard” anhedral crystals of orthopyroxene and augite, it probably reached a peak temperature lower than the temperature of pigeonite stability (1190 °C), while the final temperature recorded by the $D_{Cr}^{oliv-LCP}$ thermometer is 1167 ± 15 °C. Those two temperatures suggest that Hughes 009 cooled, at most, over 30 °C instead of 200 °C. Using a similar reasoning, and comparing the ureilite HaH 064 to experiment CI-CHS 54 performed at 1129 °C (Collinet and Grove 2020a), we infer that its peak temperature was in the range 1125 °C (Sack and Ghiorso 1994) – 1089 °C ($D_{Cr}^{oliv-LCP} T_E$) rather than 1200 °C (Weber et al. 2003) and that, therefore, it did not cool significantly.

In summary, ALH 82106 is the only sample that has recorded an episode of slow cooling at high temperature and in the region of the UPB representing the residual mantle. It cooled by only ~ 20 °C. The bulk of evidence suggests that the ureilites that represent melting residues were maintained very close to their peak temperature, and might have been actively melting, at the time of the disruption of the UPB.

4.8. Variable fO_2 during melting and mantle heterogeneity of the UPB

The metal and carbon polymorphs present along silicate grain boundaries have occasionally been interpreted as being added by the impactor that disrupted the UPB (Boynton et al. 1976; Wasson et al. 1976; Day et al. 2017). However, the weakly shocked ureilite ALHA 78019 (Berkley and Jones 1982) contains euhedral crystals of graphite intergrown with metal and sulfide. In addition, mixed cohenite-metal-sulfide inclusions occur in the silicates of many ureilites (Goodrich et al. 2013a; Fig. S2) and strongly suggest that graphite and metal were in equilibrium with silicates during partial melting. The presence of metal in equilibrium with olivine places strict constraints on the fO_2 of the system (OSI equilibrium; Nitsan, 1974). A composition of Fe_{75-95} in olivine and $Fe_{90-95}Ni_{10-5}$ in the metal at 1100-1300 °C correspond to ΔIW -1.3 to -2.5, which is identical to our experimental range of fO_2 . In experiments, the fO_2 relevant to the formation of ureilites is imposed by the C-CO buffer. Its position relative to the IW buffer is strongly dependent on both the temperature and the pressure. If the temperature is increased or the pressure is decreased, Fe^{2+} is progressively reduced to Fe^0 .

Because ureilites contain abundant graphite, it has long been assumed that the C-CO buffer imposed the fO_2 of equilibration of ureilites, like in our experiments, and was responsible for the variable Fo content of olivine cores (Fe_{75-95}) between samples (e.g. Berkley et al. 1980; Berkley and Jones 1982). In metallurgy, the process by which Fe metal is produced by heating FeO ores in the presence of C is called “smelting”. By analogy, a primary control of the C-CO buffer on the composition of silicate cores (i.e. Mg#) in ureilites has been called smelting by Walker and Grove (1993). It is now called “primary smelting” or “equilibrium smelting” in the recent ureilite literature, by opposition to “secondary smelting” or “disequilibrium smelting”, which refer to the reduction event associated with the disruption of the parent body. Despite minor semantic disagreement (Warren and Huber 2006), disequilibrium smelting has not been a source of contention. It is widely recognized as having produced the ubiquitous 10-50 μm wide rims of olivine that are increasingly FeO-poor outward and contain small metal blebs and free SiO_2 (Fig. 5a), following the pressure drop and rapid cooling at 2-20 °C/h caused by the destruction of the UPB (Miyamoto 1985; Takeda et al. 1989; Herrin et al. 2010). In most cases, the pyroxene is less affected by this process, probably due to the slower rate of Fe-Mg interdiffusion. However, a few samples display pyroxene smelted to different extents (Fig. 5e-f and Fig. S3) associated with olivine crystals containing homogeneously distributed sub- μm metal inclusions (Fig. S4). Such

samples are interpreted as briefly shock-heated by the impact before being rapidly cooled (Warren and Rubin 2010).

Equilibrium smelting, which would control the composition of olivine cores, has been more controversial. It has been invoked in the context of cumulate and residue models alike and encompasses different processes (Walker and Grove 1993; Goodrich et al. 1987, 2007; Sinha et al. 1997; Singletary and Grove 2006). In Goodrich et al. (1987), equilibrium smelting is envisioned as affecting the parental melts of ureilites as they migrate upwards and decompress before pooling at various depths. In the model of Singletary and Grove (2006), smelting affects partly molten diapirs heated more rapidly than the surrounding mantle due to their higher CAIs contents, and higher ^{26}Al concentrations. In the model of Goodrich et al. (2007), primary smelting is driven by an increase in temperature at constant pressure and happens simultaneously with partial melting.

In detail, those simple primary smelting models appear to be inconsistent with the new $D_{\text{Cr}}^{\text{oliv} - \text{LCP}}$ thermometer. For example, the model of Goodrich et al. (2007) assumed that ureilites, regardless of their Mg#, reached peak temperatures of 1250-1275 °C and that FeO-rich ureilites (Fo_{75-80}) melted deeper in the parent body than FeO-poor ureilites, as a consequence of the pressure sensitivity of the C-CO buffer (Warren 2012; Goodrich et al. 2013c). However, it is now clear that the equilibration temperatures of FeO-rich pigeonite-olivine ureilites span a 150 °C interval (1130-1280 °C). The Fo content of olivine in our experiments, which is a function of the temperature and of the CO pressure, is used to calculate the pressure of equilibration of ureilites, assuming that the $f\text{O}_2$ is primarily controlled by the C-CO buffer (Fig. 13a and Fig. S9). Because FeO-rich ureilites formed over a large range of temperature, they would also have melted over a large range of pressure (4-14 MPa), which would overlap with the pressure of equilibration of FeO-poor ureilites (2-8 MPa; Fig 13a).

If melting occurred at higher pressure (> 15 MPa), where graphite can be stable with olivine of various compositions (Fo_{74-95}) over the same temperature interval (1130-1280 °C), the Fo content in olivine and the T_E based on the $D_{\text{Cr}}^{\text{oliv} - \text{LCP}}$ would not be sufficient to constrain the pressure of ureilite equilibration (Warren and Kallemeyn 1992; Walker and Grove 1993; Warren and Huber 2006). Nevertheless, the temperature interval over which pigeonite-olivine ureilites equilibrated (1130-1280 °C) could represent a radial temperature gradient within the UPB. The slope of such a temperature profile is unconstrained as it would be influenced by many parameters

(e.g. Neumann et al. 2012). In any case, the identical equilibration temperatures of FeO-poor and FeO-rich ureilites could indicate that they melted at the same depth (Fig. 13b).

There is little doubt that the UPB accreted from isotopically heterogeneous materials. The fact that C (Barrat et al. 2017), Cr (Zhu et al. 2020) and O isotopes (Clayton and Mayeda 1988) all correlate with the Mg# of silicates suggests that the intrinsic fO_2 of ureilites was at least partly inherited from the precursor chondritic materials. The origin of the intrinsic fO_2 of meteorite parent bodies is an active area of research. The formation of FeO in the nebula setting is non-trivial and could require high dust-to-gas ratios and dust rich in H_2O (e.g. Fedkin and Grossman, 2016). Similar correlations between the Mg# of silicates (i.e. the intrinsic fO_2) and the $\Delta^{17}O$ are observed in various meteorite groups other than ureilites. Such trends could be connected to the distribution of ice and/or water vapor, either in the accretion disk or within parent bodies (e.g. Tenner et al. 2015; Sanders et al. 2017). Regardless of their origin, the initial heterogeneities in intrinsic fO_2 of ureilites do not preclude that reduction processes involving C and various gas species occurred during melting and melt migration. Such “equilibrium smelting” reactions could have partly overprinted the initial heterogeneities in intrinsic fO_2 . The presence of a gas phase could explain the incremental nature of the melting process (Wilson et al. 2008) but it is no longer seen as a requirement to drive rapid melt extraction from the residues (Goodrich et al. 2015). New models for the origin of ureilites should evaluate which primary/equilibrium smelting processes are compatible with the presence of initial heterogeneities in fO_2 and the T-P conditions of equilibration of ureilites (Fig. 13a). The large span in T_E and extent of melting recorded in ureilites suggests a UPB that was heterogeneous in intrinsic fO_2 , Mg# and isotopic compositions, resulting in a residual mantle where both FeO-rich and FeO-poor ureilites were generated over a range of depths (Fig. 14).

5. Acknowledgements

This research project was funded by a NASA Emerging Worlds grant (NNX16AD29J). US Antarctic meteorite samples are recovered by the Antarctic Search for Meteorites (ANSMET) program which has been funded by NSF and NASA, and characterized and curated by the Department of Mineral Sciences of the Smithsonian Institution and the Astromaterials Curation Office at NASA Johnson Space Center. We thank Kevin Righter for his editorial handling as well as Cyrena Goodrich and Akira Yamaguchi for their thorough and constructive reviews.

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33 929 **Figure captions:**

34 930
35 931 **Figure 1.** Backscattered images of experimental charges containing ureilite-like residues (olivine-
36 932 pigeonite-metal). (a) CI residue with *py* of 21. (b) LL residue with *py* of 39.5. *py* =
37 933 $px/(px+oliv)*100$ in wt.%.

38 934
39 935 **Figure 2.** Aggregate melt fraction as a function of the experimental temperature in batch (Collinet
40 936 and Grove, 2020a) and incremental melting experiments (this study; IW -1.4/-1.7). The melt
41 937 productivity is low following the extraction of 15 wt.% melt (plag-out temperature).

42 938
43 939 **Figure 3.** Composition of low Ca pyroxene (LCP) as a function of the temperature in batch melting
44 940 experiments (shaded areas; Collinet and Grove, 2020a) and incremental melting experiments (this
45 941 study). (a) ordinary chondritic compositions. (b) CI chondritic compositions.

46 942
47 943 **Figure 4.** Composition (a-b) and fraction of pyroxene (c-d) as a function of the Fo content in
48 944 olivine in batch and incremental melting experiments of ordinary (a-c) and CI (b-d) chondritic
49 945 compositions. The Fo content in olivine and *py* increase with decreasing *fO₂*. From Fo₇₅ (IW -1.3)
50 946 to Fo₉₅ (IW -2.5), ~12 wt.% Fe metal is produced (Figure S1). Wo content and *py* decrease with
51 947 increasing aggregate melt fraction (colored bar). The lines represent the modal and chemical
52 948 compositions of residues for a given degree of melting (Collinet and Grove, 2020b).

Figure 5. Backscattered images of a subset of the ureilites analyzed for this study. See text for detail. All samples display reduction rims in olivine. Pyroxene is almost completely “smelted” in NWA 11755 (e). It displays a coarse porosity in the FeO-poor area and a fine porosity in the FeO-rich area. MIL 090076 displays a similar texture but the pyroxene cores are homogeneous and free of porosity (f). NWA 11754 is characteristic of the “Hughes cluster” (d).

Figure 6. Composition (Wo content) and fraction of pyroxene (py) as a function of the Fo content in olivine in ureilites. All X-ray maps used to calculate the modal compositions are available in the supplementary material. Leaving out the Hughes cluster samples and the anomalous sample MET 01083, ureilites form of similar trend of increasing py with Fo (dotted line, equation $R^2=0.74$). The solid lines (b) and shaded areas (a) represent the experimental trends of Fig. 4: CI and CI residues (CIR1-3, light gray) and LL and LL residues (LLR1-2, dark gray).

Figure 7. Calibration of the $D_{Cr}^{oliv-LCP}$ thermometer. (a-b) different temperature dependence of the Cr partition coefficients between olivine (a) or low Ca pyroxene (b) and silicate melt in batch melting experiments of a CI composition (fO_2 : IW -1.3/-2.2; Collinet and Grove, 2020a). (c-d) Linear relationship between the $D_{Cr}^{oliv-LCP}$ and the experimental temperature of 60 charges with various bulk compositions (supplementary material and see text for detail). The linear relationship is used as a simple mineral thermometer that is independent of the fO_2 (within the experimental range) and the Wo content.

Figure 8. Equilibration temperature (T_E) of 76 ureilites as a function of the Fo content in olivine. All pigeonite-olivine ureilites have a higher T_E (within ± 15 °C uncertainty) than the minimum temperature of pigeonite stability of Sack and Ghiorso (1994). Two-pyroxene ureilites have a lower T_E than pigeonite-olivine ureilites at a given Fo content in olivine (or Mg# in pyroxene, see supplementary material). FeO-rich ureilites (e.g. Fo₇₇₋₈₀) cover a large range of temperature (210 °C). The temperature at which plagioclase melts out during batch melting of CI, H, LL and CM chondrites is shown for reference (green lines). Ureilites with Fo₇₄₋₈₅ and Fo₈₇₋₉₆ olivine are equilibrated at similar temperatures (mode at 1180-1220 °C). Open symbols are considered “anomalous” in the sense that they do not represent simple melting residues. Key samples: [1] NWA 766, [2] MET 78008, [3] Y 74130, [4] RaS 517, [5-6] LEW 88774, [7] HaH 064, [8] NWA 7349, [9] LAP 02382, [10] Haverro, [11] EET 96293, [12] FRO 90054, [13] EET 96314, [14] NWA 11754, [15] Hughes 009, [16] MIL 091004, [17] MET01083, [18] NWA 5555, [19-20] ALHA 82130, [21] ALHA 84136, [22] ALHA 82106, [23] MS-MU-012. See text and supplementary material for references.

Figure 9. (a) Concentration of Al_2O_3 in low-Ca pyroxene (orthopyroxene or pigeonite) as a function of the $D_{Cr}^{oliv-LCP}$ temperature (ureilites) or the experimental temperature (+). See caption of Fig. 8 for sample numbers. (b) Same as (a) but only keeping FeO-rich pigeonite-olivine ureilites. (c) Wo content in LCP as a function of the $D_{Cr}^{oliv-LCP}$ temperature of FeO-rich pigeonite-olivine ureilites (same samples as in (b)). The R^2 value in parentheses characterizes the linear regression without the three open symbols.

Figure 10. (a) Rare earth element concentrations of ureilites from Barrat et al. (2016). We excluded one sample from group A (LAP 03587) and one from group B (LAR 04315) as their pyroxene is thought to have been shock-melted (Warren and Rubin, 2010), a process that probably lowered the bulk concentration of incompatible elements. (b-d) Bulk incompatible element concentrations (leachates) as a function of the $D_{Cr}^{oliv-LCP}$ temperature. Incompatible element concentrations of Group A samples correlate negatively with T_E . Yamato 790981 (open symbol) might represent an anomalous low-temperature sample.

Figure 11. (a) Temperature dependence of Ca partition coefficients between olivine and liquid in CI, H and LL batch melting experiments (Collinet and Grove, 2020a). (b) Concentration of CaO in olivine as a function of the Fo content and the temperature (colored bar). (c) Estimated concentration of CaO in the “late-stage melts” that were last equilibrated with FeO-rich ureilites and comparison with batch melts (IW -1.3/-1.7) of H, LL, CI, CM and CV chondrites at the same temperature. The “late-stage melts” of ureilites show a CaO-enrichment trend that is characteristic of incremental melting. (d) pMELTS simulation (CI, IW -1.5) illustrating how instantaneous melts, produced by incremental melting (e.g. continuous melting, 1 and 3 wt.% critical melt fraction) become CaO-rich with increasing temperature relative to batch melts. Batch melt compositions and melting temperatures are not consistent with experiments in detail.

Figure 12. Rare earth element modeling using the data and the same overall approach as Barrat et al. (2016) but with the melting coefficients and phase proportions of Collinet and Grove (2020a). The blue field represents the “Group A” and the red field the “Group B” see Figure 10. Following Barrat et al. (2016), merrillite is stable in the residue and melts out simultaneously with plagioclase. (a) dynamic melting at IW -1.5 reproducing the REE patterns of FeO-rich ureilites, with a 0.02 critical melt fraction until the last increment, where either all the melt is removed (black lines) or 0.015 (75 wt.% of the final melt fraction) is removed (blue lines). (b) same as (a) but with a pyroxene-rich starting composition (IW -2) and 0.001 of the final melt fraction (5 wt.%) retained. (c) fractional melting. REE melting model results are non-unique, see supplementary material.

Figure 13. (a) Pressure of equilibration of ureilites assuming that the fO_2 is primarily controlled by the C-CO buffer. The pressure of equilibration (P_{C-CO}) is fixed by the Fo content of olivine of individual samples and their $D_{Cr}^{oliv-LCP}$ temperature of equilibration (T_E ; Fig. S9). (b) If the actual pressure of equilibration of all ureilites is larger than 15 MPa, the P_{C-CO} of (a) is no longer relevant and the fO_2 is not controlled by the C-CO buffer. The intrinsic fO_2 of ureilites is variable (IW -1.3/-2.5) and is higher than the fO_2 of the C-CO buffer. Graphite is stable and there are no precise constraints on the pressure of equilibration (P_E). The various T_E of ureilites can still be interpreted as representing a “geotherm” (a temperature profile). Although the slope of the geotherm is unconstrained, ureilites with different Fo content in olivine but identical T_E could be assumed to have equilibrated at the same pressure.

Figure 14. Summary: origin of ureilites and possible internal structure of the UPB. The large majority of ureilites, equilibrated at 1150-1280 °C, represent the residual mantle of a chondritic planetesimal (green) from which 15-24 wt.% of silicate melt was incrementally extracted (pink). The diversity of ureilites in terms of T_E and extent of melting suggests a vertical temperature gradient in the mantle. Ureilites equilibrated at higher temperature could have originated from deeper in the UPB. Because the T_E of more oxidized ureilites (light green) and more reduced ureilites (dark green) overlap, the residual mantle of the UPB was heterogeneous in terms of Mg# (and intrinsic fO_2 , isotopic composition) but not stratified. The UPB melted incrementally. Alkali-rich melts (Collinet and Grove 2020a), and late-stage melts rich in CaO but poor in alkali were successively extracted from the melting residues. Late stage melts produced the anomalous low-T ureilites (1060-1170 °C) rich in augite, which represent cumulates or derive from melt-rock reactions in magma conduits (i.e. Hughes cluster samples). They were likely emplaced in a shallow region of the UPB.

Figure 1. Backscattered images of experimental charges containing ureilite-like residues (olivine-pigeonite-metal). (a) CI residue with py of 21. (b) LL residue with py of 39.5. $py = px/(px+oliv)*100$ in wt.%.

247x96mm (150 x 150 DPI)

Figure 2. Aggregate melt fraction as a function of the experimental temperature in batch (Collinet and Grove, 2020a) and incremental melting experiments (this study; IW -1.4/-1.7). The melt productivity is low following the extraction of 15 wt.% melt (plag-out temperature).

86x75mm (300 x 300 DPI)

Figure 3. Composition of low Ca pyroxene (LCP) as a function of the temperature in batch melting experiments (shaded areas; Collinet and Grove, 2020a) and incremental melting experiments (this study). (a) ordinary chondritic compositions. (b) CI chondritic compositions.

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Figure 4. Composition (a-b) and fraction of pyroxene (c-d) as a function of the Fo content in olivine in batch and incremental melting experiments of ordinary (a-c) and CI (b-d) chondritic compositions. The Fo content in olivine and py increase with decreasing fO_2 . From Fo75 (IW -1.3) to Fo95 (IW -2.5), ~12 wt.% Fe metal is produced (Figure S1). Wo content and py decrease with increasing aggregate melt fraction (colored bar). The lines represent the modal and chemical compositions of residues for a given degree of melting (Collinet and Grove, 2020b).

311x307mm (300 x 300 DPI)

Figure 5. Backscattered images of a subset of the ureilites analyzed for this study. See text for detail. All samples display reduction rims in olivine. Pyroxene is almost completely “smelted” in NWA 11755 (e). It displays a coarse porosity in the FeO-poor area and a fine porosity in the FeO-rich area. MIL 090076 displays a similar texture but the pyroxene cores are homogeneous and free of porosity (f). NWA 11754 is characteristic of the “Hughes cluster” (d).

248x300mm (150 x 150 DPI)

Figure 6. Composition (Wo content) and fraction of pyroxene (py) as a function of the Fo content in olivine in ureilites. All X-ray maps used to calculate the modal compositions are available in the supplementary material. Leaving out the Hughes cluster samples and the anomalous sample MET 01083, ureilites form of similar trend of increasing py with Fo (dotted line, equation R2=0.74). The solid lines (b) and shaded areas (a) represent the experimental trends of Fig. 4: CI and CI residues (CIR1-3, light gray) and LL and LL residues (LLR1-2, dark gray).

206x272mm (300 x 300 DPI)

Figure 7. Calibration of the $D_{\text{Cr}}^{\text{(oliv-LCP)}}$ thermometer. (a-b) different temperature dependence of the Cr partition coefficients between olivine (a) or low Ca pyroxene (b) and silicate melt in batch melting experiments of a CI composition (fO_2 : IW -1.3/-2.2; Collinet and Grove, 2020a). (c-d) Linear relationship between the $D_{\text{Cr}}^{\text{(oliv-LCP)}}$ and the experimental temperature of 60 charges with various bulk compositions (supplementary material and see text for detail). The linear relationship is used as a simple mineral thermometer that is independent of the fO_2 (within the experimental range) and the Wo content.

202x373mm (300 x 300 DPI)

Figure 8. Equilibration temperature (TE) of 76 ureilites as a function of the Fo content in olivine. All pigeonite-olivine ureilites have a higher TE (within ± 15 °C uncertainty) than the minimum temperature of pigeonite stability of Sack and Ghiorso (1994). Two-pyroxene ureilites have a lower TE than pigeonite-olivine ureilites at a given Fo content in olivine (or Mg# in pyroxene, see supplementary material). FeO-rich ureilites (e.g. Fo77-80) cover a large range of temperature (210 °C). The temperature at which plagioclase melts out during batch melting of CI, H, LL and CM chondrites is shown for reference (green lines). Ureilites with Fo74-85 and Fo87-96 olivine are equilibrated at similar temperatures (mode at 1180-1220 °C). Open symbols are considered “anomalous” in the sense that they do not represent simple melting residues. Key samples: [1] NWA 766, [2] MET 78008, [3] Y 74130, [4] RaS 517, [5-6] LEW 88774, [7] HaH 064, [8] NWA 7349, [9] LAP 02382, [10] Haverro, [11] EET 96293, [12] FRO 90054, [13] EET 96314, [14] NWA 11754, [15] Hughes 009, [16] MIL 091004, [17] MET01083, [18] NWA 5555, [19-20] ALHA 82130, [21] ALHA 84136, [22] ALHA 82106, [23] MS-MU-012. See text and supplementary material for references.

227x170mm (300 x 300 DPI)

Figure 9. (a) Concentration of Al_2O_3 in low-Ca pyroxene (orthopyroxene or pigeonite) as a function of the $D_{\text{Cr}}^{\text{oliv-LCP}}$ temperature (ureilites) or the experimental temperature (+). See caption of Fig. 8 for sample numbers. (b) Same as (a) but only keeping FeO-rich pigeonite-olivine ureilites. (c) Wo content in LCP as a function of the $D_{\text{Cr}}^{\text{oliv-LCP}}$ temperature of FeO-rich pigeonite-olivine ureilites (same samples as in (b)). The R^2 value in parentheses characterizes the linear regression without the three open symbols.

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Figure 10. (a) Rare earth element concentrations of ureilites from Barrat et al. (2016). We excluded one sample from group A (LAP 03587) and one from group B (LAR 04315) as their pyroxene is thought to have been shock-melted (Warren and Rubin, 2010), a process that probably lowered the bulk concentration of incompatible elements. (b-d) Bulk incompatible element concentrations (leachates) as a function of the $D_{Cr}^{(oliv-LCP)}$ temperature. Incompatible element concentrations of Group A samples correlate negatively with TE. Yamato 790981 (open symbol) might represent an anomalous low-temperature sample.

372x298mm (300 x 300 DPI)

Figure 11. (a) Temperature dependence of Ca partition coefficients between olivine and liquid in CI, H and LL batch melting experiments (Collinet and Grove, 2020a). (b) Concentration of CaO in olivine as a function of the Fo content and the temperature (colored bar). (c) Estimated concentration of CaO in the "late-stage melts" that were last equilibrated with FeO-rich ureilites and comparison with batch melts (IW -1.3/-1.7) of H, LL, CI, CM and CV chondrites at the same temperature. The "late-stage melts" of ureilites show a CaO-enrichment trend that is characteristic of incremental melting. (d) pMELTS simulation (CI, IW -1.5) illustrating how instantaneous melts, produced by incremental melting (e.g. continuous melting, 1 and 3 wt.% critical melt fraction) become CaO-rich with increasing temperature relative to batch melts. Batch melt compositions and melting temperatures are not consistent with experiments in detail.

320x340mm (300 x 300 DPI)

Figure 12. Rare earth element modeling using the data and the same overall approach as Barrat et al. (2016) but with the melting coefficients and phase proportions of Collinet and Grove (2020a). The blue field represents the “Group A” and the red field the “Group B” see Figure 10. Following Barrat et al. (2016), merrillite is stable in the residue and melts out simultaneously with plagioclase. (a) dynamic melting at IW -1.5 reproducing the REE patterns of FeO-rich ureilites, with a 0.02 critical melt fraction until the last increment, where either all the melt is removed (black lines) or 0.015 (75 wt.% of the final melt fraction) is removed (blue lines). (b) same as (a) but with a pyroxene-rich starting composition (IW -2) and 0.001 of the final melt fraction (5 wt.%) retained. (c) fractional melting. REE melting model results are non-unique, see supplementary material.

179x379mm (300 x 300 DPI)

Figure 13. (a) Pressure of equilibration of ureilites assuming that the fO_2 is primarily controlled by the C-CO buffer. The pressure of equilibration (PC-CO) is fixed by the Fo content of olivine of individual samples and their $D_{Cr}^{oliv-LCP}$ temperature of equilibration (TE; Fig. S9). (b) If the actual pressure of equilibration of all ureilites is larger than 15 MPa, the PC-CO of (a) is no longer relevant and the fO_2 is not controlled by the C-CO buffer. The intrinsic fO_2 of ureilites is variable (IW -1.3/-2.5) and is higher than the fO_2 of the C-CO buffer. Graphite is stable and there are no precise constraints on the pressure of equilibration (PE). The various TE of ureilites can still be interpreted as representing a "geotherm" (a temperature profile). Although the slope of the geotherm is unconstrained, ureilites with different Fo content in olivine but identical TE could be assumed to have equilibrated at the same pressure.

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Figure 14. Summary: origin of ureilites and possible internal structure of the UPB. The large majority of ureilites, equilibrated at 1150-1280 °C, represent the residual mantle of a chondritic planetesimal (green) from which 15-24 wt.% of silicate melt was incrementally extracted (pink). The diversity of ureilites in terms of TE and extent of melting suggests a vertical temperature gradient in the mantle. Ureilites equilibrated at higher temperature could have originated from deeper in the UPB. Because the TE of more oxidized ureilites (light green) and more reduced ureilites (dark green) overlap, the residual mantle of the UPB was heterogenous in terms of Mg# (and intrinsic fO₂, isotopic composition) but not stratified. The UPB melted incrementally. Alkali-rich melts (Collinet and Grove 2020a), and late-stage melts rich in CaO but poor in alkali were successively extracted from the melting residues. Late stage melts produced the anomalous low-T ureilites (1060-1170 °C) rich in augite, which represent cumulates or derive from melt-rock reactions in magma conduits (i.e. Hughes cluster samples). They were likely emplaced in a shallow region of the UPB.

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Table 1: summary of experimental conditions

exp #	P (CO)	T int	log fO_2	Δ IW	t (h)	Δ T*	starting compositions per experiment				
	MPa	°C	(CCO)			°C	1	2	3	4	5
CHS 46	6.2	1191	-13.7	-1.61	72	20	LLR1	<i>H</i>			
CHS 47	6.6	1200	-13.6	-1.63	72	0	CIR1	LLR1	LLR2		
CHS 48	4.1	1194	-14.0	-1.98	72	0	CIR1	LLR1	<i>H</i>		
CHS 49	10.0	1248	-13.0	-1.62	24	0	CIR1	CIR3	LLR1	LLR2	
CHS 50	7.2	1248	-13.3	-1.89	27	0	CIR1	CIR2a	CIR3		
CHS 51	8.1	1217	-13.3	-1.58	68	0	CIR2a	CIR1	CIR3	LLR1	LLR2
CHS 53	12.1	1248	-12.8	-1.47	46	0	CIR2a	CIR1	CIR3		
CHS 63	9.1	1202	-13.3	-1.38	72	0	CIR2b	<i>Cl</i>	<i>H</i>		
CHS 65	3.1	1201	-14.2	-2.27	52	0	CIR2b	CIR1	RCa	<i>Cl</i>	<i>H</i>
CHS 66	3.4	1250	-13.9	-2.53	42	0	CIR1	CIR2b	RCa	<i>Cl</i>	
CHS 67	13.3	1274	-12.6	-1.59	20	0	CIR2b	CIR1	<i>Cl</i>		
CHS 68	5.2	1159	-12.5	-1.53	100	0	CIR2b	RCa2	RCa	<i>Cl</i>	
CHS 69	5.9	1301	-13.2	-2.44	9	0	RCa	RCa2	CIR2b	<i>H</i>	<i>Cl</i>
CHS 70	3.8	1205	-14.2	-1.84	96	0	RCa2	RCa	<i>Cl</i>		
CHS 71	7.9	1216	-13.3	-1.59	90	58	LLR1	CIR1	CIR2b	<i>Cl</i>	

fO_2 calculated based on the CCO buffer (extended formulation, French and Eugter, 1965) and expressed relative to the IW buffer (Huebner 1971)

*T (°C) of initial 2 h step - final T (°C); 0 = isothermal

Experimental charges with starting chondritic compositions (*italic*) are described in Collinet and Grove (2020a)

Table 2. Experimental starting compositions and bulk ureilite compositions

	CI*	CIR1	CIR2a	CIR2b	CIR3	PCA 82506	North Haig	H*	LL*	LLR1	LLR2	DPD	RCa1	RCa2
F=	11	15.5	15.5	19.5	mmict		(pmict)			8.5	16	mmict	n.a.	n.a.
SiO ₂	40.8	39.7	37.1	38.1	38.3	42.0	40.3	37.6	43.5	41.6	40.3	41.8	37.8	41.5
TiO ₂	0.13	0.09	0.07	0.05	0.05	0.04	0.09	0.12	0.12	0.09	0.05	0.09	0.07	0.06
Al ₂ O ₃	2.86	1.15	0.65	0.37	0.38	0.10	0.20	2.22	2.44	1.20	0.39	0.37	0.85	1.26
Cr ₂ O ₃	0.69	0.60	0.79	0.60	0.62	0.82	0.73	0.51	0.59	0.63	0.65	0.81	0.65	0.88
FeO	21.8	22.4	24.6	22.7	23.3	19.2	18.4	30.8	21.9	23.5	24.5	21.2	21.5	19.3
MnO	0.44	0.36	0.36	0.36	0.37	0.41	0.44	0.31	0.36	0.38	0.39	0.36	0.34	0.30
MgO	28.2	32.1	32.4	34.2	34.1	36.3	37.9	23.8	26.8	28.8	30.7	33.2	33.9	30.4
CaO	2.30	1.87	1.99	2.00	1.34	1.00	1.38	1.78	2.11	1.96	1.41	1.54	3.58	5.14
Na ₂ O	1.20	0.54	0.26	0.24	0.25	0.03	0.09	0.89	0.99	0.56	0.26	0.07	0.23	0.21
K ₂ O	0.12	0.03	0.00	0.01	0.01	0.01	0.03	0.10	0.10	0.03	0.01	0.03	0.05	0.08
P ₂ O ₅	0.39	0.20	0.32	0.19	0.19	0.04	0.23	0.26	0.22	0.21	0.20	0.21	0.18	0.16
NiO	1.13	1.04	1.31	1.10	1.12	0.10	0.15	1.48	1.01	1.09	1.18	0.32	1.04	0.93
Mg/Si	1.03	1.21	1.30	1.34	1.33	1.29	1.40	0.94	0.92	1.03	1.14	1.18	1.33	1.09
(Ca/Mg) _{CI}	1.01	0.72	0.76	0.73	0.49	0.34	0.45	0.93	0.98	0.84	0.57	0.58	1.31	2.10
(Al/Mg) _{CI}	1.00	0.35	0.20	0.11	0.11	0.03	0.05	0.92	0.90	0.41	0.13	0.11	0.25	0.41
Mg#	69.7	71.9	70.1	72.9	72.3	77.1	78.6	57.9	68.6	68.7	69.1	73.6	73.8	73.8

*Average chondritic starting compositions of Lodders and Fegley (1998), renormalized without FeS or volatile elements;

starting compositions of batch melting experiments (Collinet and Grove, 2020a)

F= estimated melt fraction extracted from residues relative to chondritic composition CI and LL, respectively

mmict and pmict = bulk composition of monomict and polymict ureilites from Jarosewich (2006), DPD: Dingo Pup Donga

Table 3. Mineralogy of ureilites analyzed for this study

	<i>py</i>	<i>px</i> ₂ / <i>px</i> _{tot}	<i>oliv</i>			<i>px</i> ₁ (LCP)			<i>px</i> ₂ (aug or opx)			<i>chromite</i>	
			Fo	Cr ₂ O ₃	CaO	Wo	Cr ₂ O ₃	Al ₂ O ₃	Wo	Cr ₂ O ₃	Al ₂ O ₃	Fe#	Cr#
<i>Antartic</i>													
LAP 03721	9.1		75.1	0.61	0.32	9.3	1.22	0.92					
EET 90019	36.7		89.4	0.64	0.34	9.0	1.04	0.81					
ALHA 82106	49.5	19.0	95.5	0.57	0.28	4.8	0.80	0.46	36.1	0.87	0.83		
MIL 090076	29.0		78.8	0.72	0.35	10.6	1.36	0.93					
EET 96293	57.0	3.5	87.1	0.48	0.29	4.8	1.07	1.22	37.2	1.33	1.87		
DOM 08012			77.6	0.71	0.36	7.1	1.08	0.51					
EET 96042	15.1		81.5	0.84	0.36	9.1	1.37	0.73					
MIL 07447	25.6		82.2	0.71	0.34	10.5	1.25	0.89				0.48	0.77
<i>Northwest Africa</i>													
NWA 5555	39.0	21.1	90.9	0.64	0.31	8.7	1.03	0.55	4.8	0.95	0.57		
NWA 4852	41.8		87.5	0.64	0.31	8.4	1.04	0.75					
NWA 11754	60.5	33.6	87.5	0.51	0.28	4.9	0.98	1.23	36.5	1.19	1.91		
NWA 11755			78.3	0.73	0.36	8.2	1.01	0.44					

py is the ratio (pyroxene total)/(pyroxene total + olivine)*100, wt.% percent pyroxene relative to olivine;
calculated from modal composition (X-ray maps) and phase densities
px₂/px_{tot} is the ratio px₂/(px₁+px₂)
for full EPMA analyses and analytical uncertainties, see Table S2

Table 4. Average composition of melt extracted

	1	2	
	1104 °C	1240 °C	
SiO ₂	62.44	55.7	<i>by diff</i>
TiO ₂	0.52	0.3	±0.1
Al ₂ O ₃	15.99	10	±2
Cr ₂ O ₃	0.09	0.65	±0.15
FeO	5.05	12.1	±1.5
MnO	0.17	0.55	±0.10
MgO	3.09	8	±1
CaO	4.13	11.5	±1.0
Na ₂ O	6.78	1	±1
K ₂ O	0.82	0.05	±0.05
P ₂ O ₅	0.86	0.1	±0.1

1 Aggregate melt extracted immediately

prior to plagioclase exhaustion (Collinet and Grove, 2020a)

2 Average composition of "late-stage" melt. SiO₂ calculated by difference; see text for detail.

Incremental melting in the ureilite parent body: initial composition,
melting temperatures and melt compositions

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Supplementary material

Figure S1. Metal fraction in experiments as a function of the forsterite content in olivine.

Figure S2. (a) zoned chromite inclusion in olivine with metal blebs surrounding the outer Mg-rich rims, indicative of “disequilibrium smelting”. (b) a nearly identical chromite inclusion but in pigeonite instead of olivine is not zoned/smelted. The compositions at the centers of both chromite inclusions are identical. (c and e) Composite C-rich metal (cohenite?), FeNi C-poor metal and sulfide spherules (see (Goodrich et al., 2013)). (d) inclusions of immiscible sulfide and metal. Presumably both phases were liquid, which is consistent with T_E of DOM 08012 (1220 °C, 70 °C above the FeC eutectic). (f) smaller inclusions in MIL 07447. Even the trails of μm to sub- μm inclusions in pigeonite can contain FeNi metal and sulfide.

Figure S3. Additional BSE images of disequilibrium pyroxene smelting. Similar textures affecting the rims of homogenous pyroxene crystals can be observed in MIL 090076 and LAP 03721. Also see (Warren and Rubin, 2010).

Figure S4. (*next page*) sub-µm metal inclusions in the olivine cores of ureilites that suffered pyroxene smelting (EET 96042, LAP 03721, MIL 090076, NWA 11755), whether pyroxene smelting is limited to the rims or is extensive (NWA 11755). The last two pictures represent the homogeneous cores of two samples that do not display pyroxene smelting (MIL 07447 and DOM 08012). Also see Figure 5 of the main manuscript.

EET 96042

NWA 11755

LAP 03721

MIL 090076

MIL 07447

DOM 08012

Figure S4. (caption on previous page)

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Figure S5. Composition of pyroxene in experiments and ureilites compared to the phase diagram of (Sack and Ghiorso, 1994). The shaded areas represent the pyroxene assemblage in all other primitive achondrites (lodranites, brachinites, acapulcoites), which, contrary to ureilites cooled slowly. The color code represents the experimental temperature or the $D_{Cr}^{oliv-LCP}$ temperature of equilibration (for ureilites).

Figure S6. Oxygen fugacity dependence of the partitioning coefficients of Cr between pyroxene and silicate melt extrapolated from Karner et al. (2007) experiments at 1170 °C. Our experiments (Collinet and Grove, submitted), do not show a clear fO_2 dependence at more reducing conditions.

Figure S7. Equilibration temperature (T_E ; based on the partitioning of Cr) of 75 ureilites as a function of the Mg# in low-Ca pyroxene. The black line represents the minimum temperature of pigeonite stability from (Sack and Ghiorso, 1994). Compare with Figure 8 in the main manuscript. Key samples: [1]NWA 766 [2]MET 78008 [3]Y 74130 [4]RaS 517 [5-6]LEW 88774 [7]HaH 064 [8]NWA 7349 [9]LAP 02382 [10]Havero [11]EET 96293 [12]FRO 90054 [13]EET 96314 [14]NWA 11754 [15] Hughes 009 [16]MIL 091004 [17]MET01083 [18]NWA 5555 [19-20]ALHA 82130 [21]ALHA 84136 [22]ALHA 82106.

Figure S8. Offset between calculated temperature, using the thermometer of (Singletary and Grove, 2003), and the experimental temperature of pigeonite-olivine-liquid-metal experiments of this study. Offsets are large and generally overestimate the temperature (by up to 100 °C) with no systematics. The pigeonite temperature of (Singletary and Grove, 2003) was calibrated in Na₂O-free melts, unlike the partial melts shown to be in equilibrium with ureilites. This thermometer should be replaced with the $D_{Cr}^{oliv-LCP}$ thermometer calibrated in this study (see main manuscript).

Figure S9. (a) Relationship between experimental pressure and temperature and the forsterite content in olivine of experiments. A subset of the experiments (CI composition) are used to calculate a linear regression of the pressure as a function of the temperature and Fo content in olivine: $P \text{ (MPa)} = -62.3 - 0.02993 \text{ Fo} + 0.07828 \text{ T (°C)}$. (b) Pressure of equilibration of ureilites assuming that the fO_2 is controlled by the CCO buffer (primary smelting). The pressure is calculated from the temperature of equilibration ($D_{Cr}^{oliv-LCP}$) and the Fo content of olivine in ureilites.

Figure S10. Additional REE models. Blue field represent the “group A” ureilites and red field represent the “group B” ureilites from Barrat et al. (2016). Continuous melting equations are from (Zou, 1998).

Figure S10. (continued)

Figure S11. RGB (FeMgCa) image of anomalous (poikilitic) sample MET 01083 Blue=augite, dark green=opx, light green=olivine.

pMELTS simulations (1): melting of a LL chondrite at IW -1

The melting temperatures, temperatures of phase disappearance, the composition of batch melts and the composition of mineral phases calculated with pMELTS deviate significantly from experimental results. This is not surprising as pMELTS is not optimized to simulate the melting of chondritic material at low pressure. Nevertheless, pMELTS simulations are valuable for visualizing some of the relative differences between batch and fractional melting. For near-fractional melting, the melt is continuously extracted from the residue while maintaining a 0.2 wt.% critical melt fraction (i.e. retained melt fraction). The composition of cumulates and aggregate melts at a given total melt fraction are identical (except for K_2O contents) for the different melting behaviors (right column). Following the exhaustion of plagioclase ($F = 17$ wt.%), the melt productivity essentially drops to zero in near-fractional melting. Note that the concentration of alkali elements in instantaneous near-fractional melts becomes negligible after the exhaustion of plagioclase and the concentration of Al_2O_3 decreases as well. At the same time, the CaO contents of instantaneous melts increase sharply (right column). However, due to the low melt productivity, this transition is stretched over ~ 100 °C where very little melt is actually produced (left column). The equivalence of melting residues between the different melting behavior shows that increasing fractions of batch melts can be extracted from a chondritic composition to simulate incremental melting, which is how we calculate CIR1-3 and LLR1-2

compositions (Table 2 of the main manuscript). Following the exhaustion of plagioclase and the extraction of low-degree melts, ureilites essentially stopped melting, not just because they became depleted in ^{26}Al , but also because the melt productivity became negligible over a significant temperature interval and pigeonite remained stable. The few percent of “late-stage melts” of ureilites, produced after the exhaustion of plagioclase were enriched in CaO, depleted in alkali elements and probably slightly depleted in Al_2O_3 .

pMELTS simulations (2): melting of a CI chondrite at IW -1.5

All supplementary tables are also available in an Excel file included in the .zip folder

Table S1 modal composition of experiments

comp.	exp #	T (°C)	Δ IW	total px	oliv	liq	agg. liq	metal	py	Fo	Wo px1	Wo px2
LLR1	CHS 46	1191	-1.61	31.5	48.4	8.0	15.8	9.0	39.45	78.73	6.30	
LLR1	CHS 47	1200	-1.63	35.7	42.1	9.9	17.5	11.0	45.90	81.34	5.49	
LLR1	CHS 48	1194	-1.98	42.1	32.6	7.5	15.3	16.2	56.40	88.35	5.73	
LLR1	CHS 49	1248	-1.62	34.8	39.1	12.1	19.6	12.6	47.09	83.90	4.05	
LLR1	CHS 51	1217	-1.58	33.3	43.7	10.4	18.0	11.1	43.23	80.85	4.72	
LLR1	CHS 71	1216	-1.59	34.8	41.0	10.7	18.4	12.1	45.91	82.88	4.11	
LLR2	CHS 47	1200	-1.63	37.3	46.4	2.8	18.0	12.1	44.57	81.64	5.22	
LLR2	CHS 49	1248	-1.62	37.0	43.1	4.6	19.4	14.0	46.16	84.24	4.15	
LLR2	CHS 51	1217	-1.58	33.5	50.1	3.3	18.4	11.6	40.07	80.58	5.27	
CIR1	CHS 47	1200	-1.63	16.8	62.7	10.6	21.4	8.5	19.13	81.34	7.20	
CIR1	CHS 48	1194	-1.98	27.4	49.0	9.3	19.6	14.5	33.83	88.35	6.69	
CIR1	CHS 49	1248	-1.62	14.0	62.1	12.4	22.9	9.2	16.45	83.90	4.05	
CIR1	CHS 50	1248	-1.89	21.9	51.2	11.2	21.9	14.2	27.96	88.37	5.04	
CIR1	CHS 51	1217	-1.58	15.0	64.2	10.6	21.4	8.7	16.89	81.19	6.39	
CIR1	CHS 53	1248	-1.47	19.8	59.1	10.5	20.9	8.4	20.07	81.04	4.33	
CIR1	CHS 65	1201	-2.27	31.3	41.8	9.5	20.1	17.4	40.76	92.59	6.53	
CIR1	CHS 66	1250	-2.53	33.4	37.3	9.5	20.2	18.9	45.27	94.64	4.73	
CIR1	CHS 67	1274	-1.59	21.0	52.1	13.5	24.0	11.8	26.78	85.39	6.83	
CIR1	CHS 71	1216	-1.59	18.1	59.2	11.1	21.4	10.1	21.47	83.40	5.15	
CIR2a	CHS 50	1248	-1.89	22.6	57.1	3.8	19.2	14.8	28.35	88.94	9.49	
CIR2a	CHS 51	1217	-1.58	17.8	69.5	2.3	17.9	8.5	20.35	80.99	10.4	
CIR2a	CHS 53	1248	-1.47	16.5	70.6	3.7	19.1	7.4	18.96	80.06	10.3	
CIR2a	CHS 53	1248	-1.47	17.8	68.4	3.9	19.3	8.1	20.61	80.86	9.83	
CIR2b	CHS 63	1202	-1.38	17.5	65.4	3.5	19.0	10.0	21.15	80.02	11.5	
CIR2b	CHS 65	1201	-2.27	30.2	43.0	3.0	18.5	19.8	41.22	92.80	7.89	34.6
CIR2b	CHS 66	1250	-2.53	30.0	41.1	5.0	19.9	21.3	42.18	94.96	6.95	
CIR2b	CHS 67	1274	-1.59	17.3	57.6	8.6	23.4	13.4	23.08	84.50	5.18	
CIR2b	CHS 68	1159	-1.53	16.5	67.1	2.5	18.0	10.2	19.71	79.91	8.86	
CIR2b	CHS 69	1301	-2.44	21.1	45.3	9.8	24.4	20.5	31.79	94.50	5.03	
CIR2b	CHS 71	1216	-1.59	17.2	61.6	5.5	20.3	12.8	21.82	83.75	8.09	
CIR3	CHS 49	1248	-1.62	19.9	63.0	4.6	22.8	11.0	21.03	83.60	5.58	
CIR3	CHS 50	1248	-1.89	23.9	55.8	3.8	22.1	14.7	27.01	88.27	5.58	
CIR3	CHS 51	1217	-1.58	16.6	69.0	3.5	21.8	9.0	16.41	81.34	6.79	
CIR3	CHS 53	1248	-1.47	17.9	67.6	4.2	22.4	8.9	17.91	81.11	5.63	

Table S2 olivine composition in experiments

comp.	exp #	T (°C)	Δ IW	SiO ₂	TiO ₂	Al ₂ O ₃	Cr ₂ O ₃	FeO	MnO	MgO	CaO	P ₂ O ₅	NiO	total	n
LLR1	CHS 46	1191	-1.61	38.7 <i>0.14</i>	bdl	bdl	0.56 <i>0.01</i>	19.5 <i>0.21</i>	0.32 <i>0.02</i>	40.5 <i>0.09</i>	0.31 <i>0.01</i>	0.06 <i>0.02</i>	0.08 <i>0.07</i>	101.0	6
LLR1	CHS 47	1200	-1.63	38.5 <i>0.07</i>	bdl	bdl	0.55 <i>0.02</i>	17.4 <i>0.09</i>	0.37 <i>0.02</i>	42.6 <i>0.21</i>	0.30 <i>0.01</i>	0.06 <i>0.02</i>	0.13 <i>0.03</i>	101.5	5
LLR1	CHS 48	1194	-1.98	40.4 <i>0.48</i>	bdl	bdl	0.54 <i>0.01</i>	11.1 <i>0.16</i>	0.37 <i>0.03</i>	47.2 <i>0.30</i>	0.28 <i>0.04</i>	0.04 <i>0.02</i>	0.06 <i>0.04</i>	101.2	5
LLR1	CHS 49	1248	-1.62	39.0 <i>0.28</i>	bdl	bdl	0.56 <i>0.02</i>	15.2 <i>0.19</i>	0.37 <i>0.05</i>	44.4 <i>0.18</i>	0.30 <i>0.01</i>	0.05 <i>0.04</i>	0.08 <i>0.06</i>	101.7	4
LLR1	CHS 51	1217	-1.58	39.0 <i>0.18</i>	bdl	bdl	0.57 <i>0.02</i>	17.7 <i>0.41</i>	0.36 <i>0.03</i>	41.9 <i>0.38</i>	0.31 <i>0.01</i>	bdl	0.11 <i>0.09</i>	101.3	5
LLR1	CHS 71	1216	-1.59	38.9 <i>0.87</i>	bdl	bdl	0.57 <i>0.03</i>	16.1 <i>0.09</i>	0.40 <i>0.04</i>	43.7 <i>0.30</i>	0.31 <i>0.01</i>	bdl	bdl	101.4	6
LLR2	CHS 47	1200	-1.63	38.8 <i>0.19</i>	bdl	bdl	0.59 <i>0.02</i>	17.1 <i>0.15</i>	0.44 <i>0.02</i>	42.7 <i>0.19</i>	0.30 <i>0.01</i>	0.02 <i>0.01</i>	bdl <i>0.08</i>	101.3	7
LLR2	CHS 49	1248	-1.62	39.1 <i>0.17</i>	bdl	bdl	0.61 <i>0.03</i>	14.8 <i>0.07</i>	0.46 <i>0.04</i>	44.5 <i>0.20</i>	0.30 <i>0.02</i>	0.03 <i>0.02</i>	0.05 <i>0.08</i>	101.0	6
LLR2	CHS 51	1217	-1.58	39.5 <i>0.56</i>	bdl	bdl	0.60 <i>0.02</i>	17.8 <i>0.26</i>	0.44 <i>0.02</i>	41.3 <i>0.93</i>	0.29 <i>0.02</i>	bdl	bdl	100.9	6
CIR1	CHS 47	1200	-1.63	38.7 <i>0.07</i>	bdl	bdl	0.56 <i>0.01</i>	17.4 <i>0.06</i>	0.32 <i>0.02</i>	42.5 <i>0.15</i>	0.32 <i>0.01</i>	0.03 <i>0.02</i>	0.06 <i>0.07</i>	101.7	6
CIR1	CHS 48	1194	-1.98	40.1 <i>0.20</i>	bdl	bdl	0.53 <i>0.01</i>	11.1 <i>0.10</i>	0.35 <i>0.02</i>	47.4 <i>0.21</i>	0.27 <i>0.01</i>	0.03 <i>0.01</i>	0.10 <i>0.07</i>	100.9	5
CIR1	CHS 49	1248	-1.62	39.0 <i>0.13</i>	bdl	bdl	0.56 <i>0.02</i>	15.2 <i>0.15</i>	0.37 <i>0.03</i>	44.4 <i>0.18</i>	0.30 <i>0.01</i>	0.05 <i>0.03</i>	0.08 <i>0.06</i>	101.7	4
CIR1	CHS 50	1248	-1.89	40.7 <i>0.13</i>	bdl	bdl	0.52 <i>0.02</i>	11.0 <i>0.14</i>	0.34 <i>0.03</i>	47.1 <i>0.13</i>	0.29 <i>0.01</i>	0.03 <i>0.02</i>	bdl	100.2	4
CIR1	CHS 51	1217	-1.58	39.1 <i>0.09</i>	bdl	bdl	0.59 <i>0.03</i>	17.4 <i>0.05</i>	0.32 <i>0.02</i>	42.2 <i>0.14</i>	0.33 <i>0.01</i>	bdl	0.06 <i>0.05</i>	101.4	5
CIR1	CHS 53	1248	-1.47	37.8 <i>0.60</i>	bdl	bdl	0.61 <i>0.04</i>	17.9 <i>0.13</i>	0.34 <i>0.04</i>	43.0 <i>0.20</i>	0.31 <i>0.01</i>	bdl	bdl	100.7	4
CIR1	CHS 65	1201	-2.27	40.4 <i>0.07</i>	bdl	bdl	0.48 <i>0.03</i>	7.29 <i>0.10</i>	0.33 <i>0.03</i>	51.1 <i>0.17</i>	0.27 <i>0.01</i>	bdl	bdl	100.6	6
CIR1	CHS 66	1250	-2.53	40.4 <i>0.27</i>	bdl	bdl	0.55 <i>0.02</i>	5.35 <i>0.08</i>	0.37 <i>0.04</i>	53.0 <i>0.16</i>	0.28 <i>0.02</i>	bdl	bdl	100.5	7
CIR1	CHS 67	1274	-1.59	38.0 <i>1.14</i>	bdl	bdl	0.57 <i>0.02</i>	14.2 <i>0.20</i>	0.36 <i>0.03</i>	46.5 <i>0.23</i>	0.40 <i>0.02</i>	bdl	bdl	99.9	7
CIR1	CHS 71	1216	-1.59	39.1 <i>0.18</i>	bdl	bdl	0.57 <i>0.03</i>	15.6 <i>0.13</i>	0.35 <i>0.04</i>	44.0 <i>0.09</i>	0.33 <i>0.02</i>	bdl	bdl	101.2	7

numbers in italic represent the analytical standard errors (1 σ)

bdl: below detection limit. Detection limits as follow (in ppm): Si 195, Ti 296, Al 161, Cr 406, Fe 223, Mn 217, Mg 212, Ca 145, Na 459, K 111, P 254, Ni 341

Table S2 olivine composition in experiments
(continued)

continued

comp.	exp #	T (°C)	Δ IW	SiO ₂	TiO ₂	Al ₂ O ₃	Cr ₂ O ₃	FeO	MnO	MgO	CaO	P ₂ O ₅	NiO	total	n							
CIR2a	CHS 50	1248	-1.89	40.7	0.29	bdl	bdl	0.50	0.02	10.5	0.16	0.40	0.02	47.5	0.07	0.39	0.05	bdl	bdl	100.1	3	
CIR2a	CHS 51	1217	-1.58	38.9	0.08	bdl	bdl	0.60	0.02	17.6	0.13	0.40	0.02	42.0	0.23	0.37	0.02	bdl	bdl	101.3	5	
CIR2a	CHS 53	1248	-1.47	38.1	0.32	bdl	bdl	0.66	0.00	18.6	0.01	0.40	0.01	41.8	0.08	0.45	0.02	bdl	bdl	101.5	3	
CIR2a	CHS 53	1248	-1.47	38.1	0.31	bdl	bdl	0.59	0.02	17.9	0.14	0.41	0.02	42.4	0.35	0.47	0.01	bdl	bdl	101.2	5	
CIR2b	CHS 63	1202	-1.38	38.1	0.25	bdl	bdl	0.68	0.02	18.6	0.28	0.28	0.03	41.8	0.23	0.38	0.02	bdl	0.07	0.05	99.7	5
CIR2b	CHS 65	1201	-2.27	40.4	0.28	bdl	bdl	0.66	0.04	7.06	0.06	0.39	0.04	51.0	0.23	0.33	0.04	bdl	0.07	0.02	101.1	6
CIR2b	CHS 66	1250	-2.53	40.7	0.40	bdl	bdl	0.70	0.02	4.99	0.06	0.39	0.02	52.8	0.42	0.38	0.08	bdl	0.05	0.02	100.8	7
CIR2b	CHS 67	1274	-1.59	38.7	0.12	bdl	bdl	0.73	0.02	14.7	0.07	0.38	0.03	45.1	0.22	0.34	0.01	bdl	bdl	101.0	5	
CIR2b	CHS 68	1159	-1.53	38.9	0.11	bdl	bdl	0.47	0.03	18.5	0.11	0.38	0.02	41.3	0.20	0.36	0.05	bdl	bdl	101.4	7	
CIR2b	CHS 69	1301	-2.44	40.6	0.15	bdl	bdl	0.74	0.02	5.44	0.04	0.44	0.04	52.4	0.16	0.35	0.02	bdl	bdl	100.8	7	
CIR2b	CHS 71	1216	-1.59	39.1	0.27	bdl	0.09	0.77	0.02	15.2	0.21	0.41	0.04	43.9	0.73	0.43	0.12	bdl	bdl	101.4	7	
CIR3	CHS 49	1248	-1.62	38.9	0.22	bdl	bdl	0.59	0.03	15.47	0.21	0.41	0.02	44.2	0.06	0.34	0.01	bdl	bdl	100.8	3	
CIR3	CHS 50	1248	-1.89	40.4	0.19	bdl	bdl	0.56	0.01	11.14	0.10	0.44	0.03	47.0	0.53	0.33	0.02	bdl	0.07	0.06	99.4	3
CIR3	CHS 51	1217	-1.58	39.0	0.10	bdl	bdl	0.59	0.01	17.30	0.16	0.42	0.01	42.3	0.25	0.34	0.03	bdl	bdl	101.4	5	
CIR3	CHS 53	1248	-1.47	38.4	0.02	bdl	bdl	0.64	0.02	17.64	0.14	0.42	0.01	42.5	0.12	0.36	0.01	bdl	bdl	101.2	3	
RCa	CHS 65	1201	-2.27	40.6	0.37	bdl	bdl	0.49	0.02	6.97	0.09	0.38	0.03	51.1	0.41	0.34	0.08	bdl	bdl	100.0	8	
RCa	CHS 66	1250	-2.53	40.8	0.31	bdl	bdl	0.52	0.03	4.69	0.07	0.37	0.02	53.2	0.45	0.40	0.09	bdl	bdl	100.6	9	
RCa	CHS 68	1159	-1.53	38.6	0.71	bdl	bdl	0.49	0.01	17.70	0.16	0.32	0.02	42.3	0.31	0.40	0.01	bdl	0.04	0.01	100.8	7
RCa	CHS 69	1301	-2.44	41.2	0.32	bdl	bdl	0.53	0.01	5.65	0.08	0.37	0.05	51.7	0.24	0.46	0.02	bdl	0.05	0.02	100.2	6
RCa	CHS 70	1205	-1.84	38.9	0.20	bdl	bdl	0.52	0.01	15.23	0.21	0.35	0.04	44.5	0.21	0.34	0.01	bdl	bdl	101.6	3	
RCa2	CHS 68	1159	-1.53	39.2	0.15	bdl	bdl	0.40	0.02	17.7	0.08	0.36	0.04	42.0	0.17	0.39	0.09	bdl	bdl	101.4	7	
RCa2	CHS 69	1301	-2.44	41.3	0.23	bdl	bdl	0.71	0.04	5.15	0.08	0.33	0.03	51.9	0.38	0.48	0.06	bdl	bdl	100.3	6	
RCa2	CHS 70	1205	-1.84	39.1	0.17	bdl	bdl	0.53	0.03	14.0	0.02	0.41	0.04	45.5	0.10	0.31	0.03	bdl	bdl	101.4	4	

numbers in italic represent the analytical standard errors (1 σ)

bdl: below detection limit. Detection limits as follow (in ppm): Si 195, Ti 296, Al 161, Cr 406, Fe 223, Mn 217, Mg 212, Ca 145, Na 459, K 111, P 254, Ni 341

Table S3 low Ca pyroxene composition in experiments

comp.	exp #	T (°C)	Δ IW	SiO ₂		TiO ₂		Al ₂ O ₃		Cr ₂ O ₃		FeO		MnO		MgO		CaO		P ₂ O ₅		NiO	total	n
LLR1	CHS 46	1191	-1.61	54.6	<i>0.76</i>	0.09	<i>0.02</i>	0.47	<i>0.10</i>	0.91	<i>0.04</i>	12.7	<i>1.19</i>	0.37	<i>0.02</i>	27.6	<i>0.54</i>	3.24	<i>0.43</i>	0.11	<i>0.04</i>	bdl	101.0	17
LLR1	CHS 47	1200	-1.63	55.2	<i>0.26</i>	0.09	<i>0.02</i>	0.38	<i>0.06</i>	0.87	<i>0.03</i>	11.3	<i>0.16</i>	0.35	<i>0.01</i>	28.8	<i>0.29</i>	2.84	<i>0.13</i>	0.08	<i>0.02</i>	bdl	101.0	8
LLR1	CHS 48	1194	-1.98	56.5	<i>0.48</i>	0.11	<i>0.03</i>	0.44	<i>0.10</i>	0.86	<i>0.05</i>	7.66	<i>0.22</i>	0.39	<i>0.02</i>	31.0	<i>0.59</i>	2.98	<i>0.28</i>	0.08	<i>0.03</i>	bdl	100.7	14
LLR1	CHS 49	1248	-1.62	55.7	<i>0.71</i>	0.07	<i>0.02</i>	0.28	<i>0.06</i>	0.85	<i>0.02</i>	10.2	<i>0.22</i>	0.34	<i>0.02</i>	30.4	<i>0.56</i>	2.12	<i>0.20</i>	0.08	<i>0.05</i>	bdl	100.4	8
LLR1	CHS 51	1217	-1.58	55.1	<i>0.25</i>	0.05	<i>0.02</i>	0.34	<i>0.05</i>	0.85	<i>0.04</i>	11.2	<i>0.21</i>	0.38	<i>0.01</i>	29.5	<i>0.80</i>	2.47	<i>0.07</i>	0.09	<i>0.03</i>	bdl	99.8	3
LLR1	CHS 71	1216	-1.59	56.0	<i>0.24</i>	0.08	<i>0.02</i>	0.28	<i>0.06</i>	0.89	<i>0.04</i>	10.6	<i>0.09</i>	0.37	<i>0.01</i>	29.5	<i>0.32</i>	2.11	<i>0.34</i>	0.12	<i>0.02</i>	bdl	101.1	5
LLR2	CHS 47	1200	-1.63	55.4	<i>0.48</i>	0.10	<i>0.02</i>	0.34	<i>0.11</i>	0.90	<i>0.02</i>	11.2	<i>0.45</i>	0.44	<i>0.01</i>	28.7	<i>0.36</i>	2.68	<i>0.06</i>	0.09	<i>0.03</i>	bdl	101.0	6
LLR2	CHS 49	1248	-1.62	56.2	<i>0.27</i>	0.05	<i>0.01</i>	0.15	<i>0.04</i>	0.84	<i>0.03</i>	9.72	<i>0.14</i>	0.42	<i>0.02</i>	30.2	<i>0.56</i>	2.14	<i>0.12</i>	0.09	<i>0.04</i>	bdl	101.1	6
LLR2	CHS 51	1217	-1.58	55.3	<i>0.18</i>	bdl		0.22	<i>0.05</i>	0.83	<i>0.03</i>	11.0	<i>0.29</i>	0.43	<i>0.00</i>	29.3	<i>0.14</i>	2.75	<i>0.14</i>	0.12	<i>0.02</i>	bdl	99.8	2
CIR1	CHS 47	1200	-1.63	55.4	<i>0.48</i>	0.08	<i>0.02</i>	0.42	<i>0.05</i>	0.90	<i>0.03</i>	11.2	<i>0.31</i>	0.35	<i>0.01</i>	28.0	<i>0.43</i>	3.69	<i>0.18</i>	0.09	<i>0.03</i>	bdl	101.1	10
CIR1	CHS 48	1194	-1.98	56.2	<i>0.66</i>	0.11	<i>0.01</i>	0.49	<i>0.05</i>	0.92	<i>0.03</i>	7.56	<i>0.06</i>	0.37	<i>0.02</i>	30.7	<i>0.36</i>	3.49	<i>0.12</i>	0.08	<i>0.01</i>	bdl	100.7	9
CIR1	CHS 49	1248	-1.62	55.7	<i>0.31</i>	0.07	<i>0.02</i>	0.28	<i>0.06</i>	0.85	<i>0.04</i>	10.2	<i>0.33</i>	0.34	<i>0.01</i>	30.4	<i>0.34</i>	2.12	<i>0.16</i>	0.08	<i>0.02</i>	bdl	100.4	8
CIR1	CHS 50	1248	-1.89	56.5	<i>0.57</i>	0.07	<i>0.02</i>	0.32	<i>0.07</i>	0.70	<i>0.02</i>	7.64	<i>0.49</i>	0.35	<i>0.01</i>	31.7	<i>0.21</i>	2.66	<i>0.16</i>	0.06	<i>0.04</i>	bdl	99.9	10
CIR1	CHS 51	1217	-1.58	55.3	<i>0.10</i>	0.09	<i>0.03</i>	0.33	<i>0.05</i>	0.90	<i>0.03</i>	10.9	<i>0.11</i>	0.35	<i>0.01</i>	28.8	<i>0.44</i>	3.32	<i>0.25</i>	0.08	<i>0.01</i>	bdl	99.9	6
CIR1	CHS 53	1248	-1.47	54.9	<i>0.80</i>	0.06	<i>0.03</i>	0.28	<i>0.05</i>	0.84	<i>0.03</i>	11.3	<i>0.06</i>	0.33	<i>0.01</i>	30.0	<i>0.18</i>	2.28	<i>0.21</i>	0.06	<i>0.02</i>	bdl	99.4	5
CIR1	CHS 65	1201	-2.27	56.6	<i>0.20</i>	0.12	<i>0.01</i>	0.51	<i>0.07</i>	0.86	<i>0.03</i>	5.15	<i>0.17</i>	0.40	<i>0.09</i>	32.8	<i>0.39</i>	3.47	<i>0.16</i>	0.09	<i>0.02</i>	bdl	100.4	7
CIR1	CHS 66	1250	-2.53	57.2	<i>0.49</i>	0.11	<i>0.01</i>	0.26	<i>0.03</i>	0.76	<i>0.03</i>	4.07	<i>0.56</i>	0.40	<i>0.02</i>	34.6	<i>0.11</i>	2.55	<i>0.01</i>	0.08	<i>0.04</i>	bdl	100.8	4
CIR1	CHS 67	1274	-1.59	55.9	<i>0.24</i>	0.04	<i>0.02</i>	0.11	<i>0.01</i>	0.76	<i>0.02</i>	8.94	<i>0.12</i>	0.40	<i>0.02</i>	30.1	<i>0.46</i>	3.59	<i>0.42</i>	0.07	<i>0.03</i>	bdl	101.0	10
CIR1	CHS 71	1216	-1.59	56.1	<i>0.40</i>	0.08	<i>0.02</i>	0.28	<i>0.04</i>	0.89	<i>0.04</i>	10.1	<i>0.11</i>	0.33	<i>0.02</i>	29.4	<i>0.23</i>	2.65	<i>0.16</i>	0.09	<i>0.04</i>	bdl	101.0	7

numbers in italic represent the analytical standard errors (1 σ)

bdl: below detection limit. Detection limits as follow (in ppm): Si 195, Ti 296, Al 161, Cr 406, Fe 223, Mn 217, Mg 212, Ca 145, Na 459, K 111, P 254, Ni 341

Table S3 low Ca pyroxene composition in experiments (continued)

comp.	exp #	T (°C)	Δ IW	SiO ₂	TiO ₂	Al ₂ O ₃	Cr ₂ O ₃	FeO	MnO	MgO	CaO	P ₂ O ₅	NiO	total	n									
CIR2a	CHS 50	1248	-1.89	56.3	0.44	0.06	0.01	0.24	0.04	0.70	0.04	7.23	0.21	0.43	0.02	29.9	0.31	4.96	0.20	0.10	0.02	bdl	99.9	11
CIR2a	CHS 51	1217	-1.58	55.0	0.38	0.08	0.02	0.32	0.04	0.89	0.03	10.9	0.41	0.39	0.02	27.0	0.64	5.32	0.46	0.12	0.05	bdl	100.2	6
CIR2a	CHS 53	1248	-1.47	55.4	0.48	0.05	0.02	0.17	0.01	0.83	0.04	10.8	0.18	0.38	0.01	27.0	0.59	5.30	0.43	0.07	0.04	bdl	100.7	8
CIR2a	CHS 53	1248	-1.47	55.4	0.29	0.03	0.02	0.13	0.02	0.81	0.04	10.5	0.27	0.38	0.02	27.6	0.33	5.08	0.45	0.09	0.02	bdl	100.6	8
CIR2b	CHS 63	1202	-1.38	54.3	0.48	0.11	0.03	0.44	0.06	1.13	0.04	10.9	0.34	0.42	0.02	26.6	0.54	5.92	0.56	0.10	0.03	bdl	99.7	4
CIR2b	CHS 65	1201	-2.27	55.6	0.97	0.12	0.02	0.65	0.05	1.09	0.02	5.28	0.30	0.51	0.02	32.4	0.43	4.22	0.26	0.12	0.04	bdl	99.4	2
CIR2b	CHS 66	1250	-2.53	56.8	0.75	0.07	0.02	0.33	0.09	0.99	0.03	3.71	0.17	0.47	0.01	33.8	0.85	3.73	0.23	0.09	0.02	bdl	101.0	14
CIR2b	CHS 67	1274	-1.59	55.6	0.42	0.05	0.02	0.20	0.01	0.97	0.02	9.55	0.09	0.39	0.01	30.4	0.10	2.72	0.12	0.08	0.04	bdl	100.4	8
CIR2b	CHS 68	1159	-1.53	54.8	0.54	0.12	0.02	0.47	0.04	0.84	0.03	11.8	0.27	0.43	0.04	26.8	0.34	4.52	0.15	0.14	0.03	bdl	99.7	5
CIR2b	CHS 69	1301	-2.44	57.3	0.31	0.04	0.02	0.16	0.03	0.92	0.03	4.24	0.28	0.45	0.02	34.2	0.32	2.69	0.12	0.04	0.02	bdl	101.0	7
CIR2b	CHS 71	1216	-1.59	55.8	0.74	0.06	0.02	0.31	0.07	1.12	0.03	9.59	0.24	0.41	0.03	28.4	0.41	4.14	0.35	0.11	0.03	bdl	101.2	12
CIR3	CHS 49	1248	-1.62	55.7	0.46	0.04	0.02	0.17	0.02	0.84	0.03	10.2	0.12	0.40	0.02	29.6	0.34	2.91	0.20	0.08	0.03	bdl	100.7	8
CIR3	CHS 50	1248	-1.89	56.3	0.46	0.06	0.02	0.25	0.06	0.76	0.03	7.75	0.41	0.44	0.02	31.4	0.33	2.94	0.10	0.14	0.06	bdl	100.0	7
CIR3	CHS 51	1217	-1.58	55.8	1.20	0.08	0.02	0.32	0.09	0.84	0.03	10.9	0.25	0.41	0.01	28.1	0.71	3.46	0.26	0.13	0.03	bdl	100.1	4
CIR3	CHS 53	1248	-1.47	55.6	0.15	0.06	0.01	0.23	0.07	0.85	0.02	11.1	0.23	0.40	0.02	28.8	0.38	2.91	0.26	0.06	0.03	bdl	100.7	15
RCa	CHS 65	1201	-2.27	56.5	0.31	0.05	0.01	0.72	0.07	0.84	0.04	4.68	0.15	0.51	0.04	32.0	0.36	4.53	0.38	0.09	0.04	bdl	100.5	5
RCa	CHS 66	1250	-2.53	57.1	0.20	bdl		0.39	0.04	0.74	0.04	3.25	0.08	0.44	0.03	32.6	0.43	5.38	0.40	0.08	0.02	bdl	100.9	9
RCa	CHS 68	1159	-1.53	55.1	0.31	0.05	0.01	0.96	0.26	0.92	0.08	10.4	0.25	0.34	0.02	25.8	0.68	6.23	0.87	0.16	0.09	bdl	100.1	12
RCa	CHS 69	1301	-2.44	56.5	0.73	bdl		0.12	0.01	0.66	0.04	5.28	1.16	0.39	0.01	32.3	0.59	4.69	0.37	bdl		bdl	99.8	5
RCa	CHS 70	1205	-1.84	55.6	0.26	0.05	0.02	0.81	0.11	0.89	0.04	9.44	0.35	0.34	0.01	27.8	0.40	4.97	0.20	0.13	0.05	bdl	100.4	8
RCa2	CHS 69	1301	-2.44	57.4	0.15	bdl		0.14	0.03	0.87	0.03	4.26	0.51	0.33	0.03	32.4	0.30	4.77	0.09	bdl		bdl	99.4	6

numbers in italic represent the analytical standard errors (1 σ)

bdl: below detection limit. Detection limits as follow (in ppm): Si 195, Ti 296, Al 161, Cr 406, Fe 223, Mn 217, Mg 212, Ca 145, Na 459, K 111, P 254, Ni 341

Table S4 augite composition in experiments

comp.	exp #	T (°C)	Δ IW	SiO ₂	TiO ₂		Al ₂ O ₃		Cr ₂ O ₃		FeO		MnO		MgO		CaO		Na ₂ O		P ₂ O ₅		NiO	total	n	
CIR2b	CHS 65	1201	-2.27	54.4		0.11		0.98		0.86		2.90		0.34		22.5		17.8		0.19		0.19		bdl	101.1	1
RCa	CHS 65	1201	-2.27	54.2	0.24	0.12	0.02	1.28	0.09	0.82	0.02	2.78	0.12	0.33	0.02	21.7	0.21	18.5	0.28	0.18	0.02	0.18	0.02	bdl	100.8	7
RCa	CHS 66	1250	-2.53	55.2	0.33	0.08	0.04	0.75	0.13	0.66	0.03	2.29	0.15	0.33	0.04	23.4	1.17	17.0	1.59	0.16	0.06	0.16	0.06	bdl	101.3	11
RCa	CHS 68	1159	-1.53	53.4	0.91	0.09	0.02	1.44	0.09	1.14	0.09	6.93	0.16	0.24	0.01	20.4	0.25	16.1	0.66	0.24	0.02	0.24	0.02	bdl	100.3	2
RCa	CHS 69	1301	-2.44	55.2	0.04	0.15	0.01	0.97	0.25	1.01	0.11	3.43	0.02	0.36	0.04	25.7	0.35	13.1	0.32	0.09	0.04	0.09	0.04	bdl	100.8	3
RCa	CHS 70	1205	-1.84	53.0	0.04	0.10	0.01	1.60	0.11	1.17	0.04	5.92	0.03	0.24	0.02	19.5	0.51	18.3	0.53	0.21	0.00	0.21	0.00	bdl	101.3	8
RCa2	CHS 68	1159	-1.53	52.9	0.10	0.22	0.01	1.50	0.06	1.09	0.02	6.39	0.09	0.24	0.02	18.0	0.59	19.2	0.44	0.27	0.05	0.27	0.05	bdl	100.3	3
RCa2	CHS 69	1301	-2.44	55.7	0.14	0.09	0.01	0.81	0.08	1.37	0.09	3.21	0.17	0.35	0.00	25.6	0.13	12.8	0.06	0.09	0.01	0.09	0.01	bdl	100.3	4
RCa2	CHS 70	1205	-1.84	52.8	0.78	0.18	0.03	1.58	0.09	1.19	0.11	5.80	0.16	0.25	0.01	20.4	0.03	17.6	0.10	0.20	0.05	0.20	0.05	bdl	101.4	5

numbers in italic represent the analytical standard errors (1 σ)

bdl: below detection limit. Detection limits as follow (in ppm): Si 195, Ti 296, Al 161, Cr 406, Fe 223, Mn 217, Mg 212, Ca 145, Na 459, K 111, P 254, Ni 341

Table S5 melt compositon in experiments

comp.	exp #	T (°C)	Δ IW	SiO ₂	TiO ₂	Al ₂ O ₃	Cr ₂ O ₃	FeO	MnO	MgO	CaO	Na ₂ O	K ₂ O	P ₂ O ₅	total	n	K _d ^{oliv-liq}	K _d _{T05}
LLR1	CHS 46	1191	-1.61	57.61	0.58	11.15	0.38	10.71	0.24	5.69	8.68	4.27	0.35	0.33	100.1	6	0.26	0.31
				<i>0.47</i>	<i>0.03</i>	<i>0.16</i>	<i>0.06</i>	<i>0.20</i>	<i>0.02</i>	<i>0.26</i>	<i>0.34</i>	<i>0.11</i>	<i>0.02</i>	<i>0.05</i>				
CIR1	CHS 47	1200	-1.63	57.12	0.65	10.51	0.43	10.35	0.24	6.51	9.70	3.89	0.32	0.25	101.3	2	0.26	0.31
				<i>0.04</i>	<i>0.02</i>	<i>0.08</i>	<i>0.06</i>	<i>0.19</i>	<i>0.00</i>	<i>0.16</i>	<i>0.07</i>	<i>0.04</i>	<i>0.03</i>	<i>0.01</i>				
CIR1	CHS 50	1248	-1.89	57.94	0.62	9.61	0.55	7.85	0.31	9.37	10.06	3.26	0.33	0.08	100.5	3	0.28	0.31
				<i>0.36</i>	<i>0.01</i>	<i>0.08</i>	<i>0.02</i>	<i>0.08</i>	<i>0.00</i>	<i>0.19</i>	<i>0.17</i>	<i>0.06</i>	<i>0.00</i>	<i>0.00</i>				
CIR1	CHS 71	1216	-1.59	57.17	0.69	9.79	0.59	10.33	0.25	6.38	10.72	3.46	0.30	0.32	100.0	3	0.22	0.32
				<i>1.19</i>	<i>0.02</i>	<i>0.32</i>	<i>0.03</i>	<i>0.66</i>	<i>0.09</i>	<i>0.33</i>	<i>0.21</i>	<i>0.06</i>	<i>0.04</i>	<i>0.11</i>				
H	CHS 46	1191	-1.61	58.3	0.45	11.8	0.36	10.5	0.25	6.05	7.00	4.58	0.43	0.21	101.1	6	0.31	0.31
				<i>0.1</i>	<i>0.02</i>	<i>0.1</i>	<i>0.02</i>	<i>0.1</i>	<i>0.01</i>	<i>0.06</i>	<i>0.06</i>	<i>0.09</i>	<i>0.01</i>	<i>0.02</i>				

the numbers in italic represent the standard deviation of EPMA analyses

Kd T05 are the Kd oliv-liq calculated with Toplis (2005), note that several compositions display lower apparent Kd and are likely quenched modified

H melt from Collinet and Grove (submitted) for comparison

Table S6 experiments used for the calibration of the DCr oliv-LCP thermometer

Name	Mat	T	dIW	DCr	olivine											NIO
					SiO2	TiO2	Al2O3	Cr2O3	FeO	MnO	MgO	CaO	Na2O	K2O	P2O5	
CHS 71	LLR1	1216	-1.59	0.64	38.88	bdl	bdl	0.57	16.08	0.40	43.68	0.31	n.a.	n.a.	bdl	bdl
CHS 71	CIR1	1216	-1.59	0.65	39.07	bdl	bdl	0.57	15.62	0.35	44.03	0.33	n.a.	n.a.	bdl	bdl
CHS 71	CIR2b	1216	-1.59	0.69	39.11	bdl	bdl	0.77	15.19	0.41	43.93	0.43	n.a.	n.a.	bdl	bdl
CHS 71	CI	1216	-1.59	0.72	39.03	bdl	bdl	0.77	15.08	0.55	44.22	0.28	n.a.	n.a.	bdl	bdl
CHS 70	CI	1167	-1.84	0.52	38.96	bdl	bdl	0.59	13.94	0.54	45.60	0.30	n.a.	n.a.	bdl	bdl
CHS 70	RCa2	1167	-1.84	0.58	38.87	bdl	bdl	0.52	15.23	0.35	44.49	0.34	n.a.	n.a.	bdl	bdl
CHS 69	H	1301	-2.44	0.87	40.55	bdl	bdl	0.55	6.70	0.42	51.48	0.22	n.a.	n.a.	bdl	bdl
CHS 69	RCa	1301	-2.44	0.81	41.22	bdl	bdl	0.53	5.65	0.37	51.70	0.46	n.a.	n.a.	bdl	bdl
CHS 69	RCa2	1301	-2.44	0.81	41.32	bdl	bdl	0.71	5.15	0.33	51.94	0.48	n.a.	n.a.	bdl	bdl
CHS 69	CIR2b	1301	-2.44	0.81	40.62	bdl	bdl	0.74	5.44	0.44	52.36	0.35	n.a.	n.a.	bdl	bdl
CHS 69	CI	1301	-2.44	0.88	40.47	bdl	bdl	0.70	5.82	0.56	52.14	0.21	n.a.	n.a.	bdl	bdl
CHS 68	CI	1159	-1.53	0.57	38.84	bdl	bdl	0.48	19.34	0.51	40.45	0.34	n.a.	n.a.	bdl	bdl
CHS 68	CIR2b	1159	-1.53	0.56	38.93	bdl	bdl	0.47	18.51	0.38	41.30	0.36	n.a.	n.a.	bdl	bdl
CHS 68	RCa	1159	-1.53	0.55	39.27	bdl	bdl	0.48	17.54	0.31	41.88	0.41	n.a.	n.a.	bdl	bdl
CHS 67	CIR2b	1274	-1.59	0.75	38.67	bdl	bdl	0.73	14.74	0.38	45.10	0.34	n.a.	n.a.	bdl	bdl
CHS 67	CIR1	1274	-1.59	0.75	37.99	bdl	bdl	0.57	14.17	0.36	46.46	0.40	n.a.	n.a.	bdl	bdl
CHS 66	CIR1	1250	-2.53	0.71	40.41	bdl	bdl	0.55	5.35	0.37	52.97	0.28	n.a.	n.a.	bdl	bdl
CHS 66	CIR2b	1250	-2.53	0.71	40.66	bdl	bdl	0.70	4.99	0.39	52.79	0.38	n.a.	n.a.	bdl	bdl
CHS 66	RCa	1250	-2.53	0.71	40.78	bdl	bdl	0.52	4.69	0.37	53.17	0.40	n.a.	n.a.	bdl	bdl
CHS 65	CIR2b	1201	-2.27	0.60	40.36	bdl	bdl	0.66	7.06	0.38	51.13	0.32	n.a.	n.a.	bdl	0.06
CHS 65	CIR1	1201	-2.27	0.58	40.45	bdl	bdl	0.50	7.29	0.33	51.10	0.27	n.a.	n.a.	bdl	bdl
CHS 65	RCa	1201	-2.27	0.58	40.63	bdl	bdl	0.49	6.97	0.38	51.15	0.34	n.a.	n.a.	bdl	bdl
CHS 64	LL	1202	-1.08	0.64	37.17	bdl	bdl	0.50	24.53	0.38	37.11	0.24	n.a.	n.a.	bdl	bdl
CHS 63	H	1202	-1.38	0.64	37.86	bdl	bdl	0.59	20.22	0.37	40.65	0.26	n.a.	n.a.	bdl	bdl
CHS 63	CIR2b	1202	-1.38	0.63	38.12	bdl	bdl	0.68	18.62	0.28	41.84	0.38	n.a.	n.a.	bdl	0.07
CHS 63	CI	1202	-1.38	0.64	38.31	bdl	bdl	0.65	18.47	0.45	41.81	0.29	n.a.	n.a.	bdl	bdl
CHS 62	CI	1192	-2.02	0.64	38.35	bdl	bdl	0.68	14.71	0.50	45.37	0.31	n.a.	n.a.	bdl	0.08
CHS 61	H	1301	-1.77	0.82	39.32	bdl	bdl	0.55	12.56	0.43	46.88	0.20	n.a.	n.a.	bdl	bdl
CHS 60	CI	1104	-1.63	0.39	38.28	bdl	bdl	0.32	19.44	0.56	41.05	0.25	n.a.	n.a.	bdl	0.07
CHS 54	CI	1129	-1.55	0.47	37.84	bdl	bdl	0.38	20.36	0.53	40.42	0.30	n.a.	n.a.	bdl	0.06
CHS 54	CIR1	1129	-1.55	0.51	37.70	bdl	bdl	0.36	22.46	0.31	38.75	0.31	n.a.	n.a.	bdl	bdl
CHS 53	CIR1	1250	-1.47	0.71	37.75	bdl	bdl	0.61	17.93	0.34	43.01	0.31	n.a.	n.a.	bdl	bdl
CHS 53	CIR2a	1250	-1.47	0.72	38.14	bdl	bdl	0.59	17.91	0.41	42.44	0.47	n.a.	n.a.	bdl	bdl
CHS 51	CIR2a	1217	-1.58	0.67	38.94	bdl	bdl	0.60	17.60	0.40	42.04	0.37	n.a.	n.a.	bdl	bdl
CHS 51	CIR1	1217	-1.58	0.64	39.11	bdl	bdl	0.59	17.42	0.32	42.17	0.33	n.a.	n.a.	bdl	0.06
CHS 50	CIR1	1248	-1.89	0.73	40.65	bdl	bdl	0.52	11.04	0.34	47.08	0.29	n.a.	n.a.	bdl	bdl
CHS 50	CIR2a	1248	-1.89	0.72	40.67	bdl	bdl	0.50	10.52	0.40	47.46	0.39	n.a.	n.a.	bdl	bdl
CHS 49	CIR1	1248	-1.62	0.72	38.98	bdl	bdl	0.59	16.33	0.33	43.36	0.31	n.a.	n.a.	bdl	0.07
CHS 49	CIR3	1248	-1.62	0.70	38.91	bdl	bdl	0.59	15.47	0.41	44.23	0.34	n.a.	n.a.	bdl	bdl
CHS 49	LLR2	1248	-1.62	0.72	39.14	bdl	bdl	0.61	14.84	0.46	44.52	0.30	n.a.	n.a.	bdl	0.08
CHS 48	CIR1	1194	-1.98	0.59	40.14	bdl	bdl	0.54	11.14	0.35	47.40	0.27	n.a.	n.a.	bdl	0.10
CHS 48	H	1194	-1.98	0.61	39.80	bdl	bdl	0.53	11.68	0.42	47.24	0.24	n.a.	n.a.	bdl	bdl
CHS 47	CIR1	1200	-1.63	0.62	38.73	bdl	bdl	0.56	17.40	0.32	42.55	0.32	n.a.	n.a.	bdl	0.06
CHS 46	CIR1	1191	-1.61	0.60	38.68	bdl	bdl	0.56	19.50	0.32	40.49	0.31	n.a.	n.a.	0.06	0.08
CHS 45	LLR1	1182	-1.60	0.54	38.77	bdl	bdl	0.52	18.59	0.33	41.39	0.30	n.a.	n.a.	bdl	bdl
CHS 44	H	1161	-1.86	0.55	39.76	bdl	bdl	0.47	13.59	0.48	45.34	0.24	n.a.	n.a.	0.08	bdl
CHS 43	H	1163	-2.06	0.50	40.59	bdl	bdl	0.41	9.65	0.47	48.46	0.22	n.a.	n.a.	0.08	0.07
CHS 42	H	1085	-1.65	0.34	38.64	bdl	bdl	0.29	17.21	0.43	42.95	0.22	n.a.	n.a.	0.10	0.05
CHS 41	H	1142	-2.00	0.48	40.02	bdl	bdl	0.37	12.30	0.45	46.42	0.23	n.a.	n.a.	bdl	0.07
CHS 40	H	1108	-1.81	0.38	39.08	bdl	bdl	0.28	15.57	0.42	44.31	0.21	n.a.	n.a.	bdl	0.05
CHS 38	LL	1086	-1.30	0.37	37.60	bdl	bdl	0.23	24.73	0.35	36.67	0.25	n.a.	n.a.	0.09	0.07
CHS 35	H	1079	-1.04	0.37	35.75	bdl	bdl	0.21	33.26	0.33	29.96	0.30	n.a.	n.a.	0.05	0.09
CHS 33	H	1112	-1.18	0.41	36.04	bdl	bdl	0.26	31.08	0.36	31.87	0.37	n.a.	n.a.	0.15	0.08
CHS 34	LL	1108	-1.16	0.43	37.41	bdl	bdl	0.25	25.94	0.29	35.54	0.32	n.a.	n.a.	0.06	bdl
CHS 31	H	1202	-1.56	0.64	38.40	bdl	bdl	0.50	18.66	0.41	41.65	0.28	n.a.	n.a.	bdl	0.05
CHS 31	HDep	1202	-1.56	0.63	38.70	bdl	bdl	0.49	19.19	0.41	40.60	0.37	n.a.	n.a.	0.07	bdl
CHS 26	H	1158	-1.42	0.49	37.25	bdl	bdl	0.36	25.24	0.40	36.34	0.30	n.a.	n.a.	bdl	bdl
CHS 37	LL	1063	-0.98	0.33	36.74	bdl	bdl	0.17	28.06	0.38	34.20	0.24	n.a.	n.a.	0.09	0.08
CHS 59	CI	1100	-1.60	0.39	38.28	bdl	bdl	0.32	19.44	0.56	41.05	0.25	n.a.	n.a.	bdl	0.07
CHS 58	CI	1136	-2.27	0.49	40.32	bdl	bdl	0.54	10.31	0.60	47.83	0.27	n.a.	n.a.	0.06	bdl

n.a. not analyzed

bdl: below detection limit

see Table S2, S3 and S4 for standard deviations

Table S6 experiments used for the calibration of the DCr oliv-LCP thermometer (continued)

low Ca pyroxene																
Name	Mat	T	dIW	DCr	SiO2	TiO2	Al2O3	Cr2O3	FeO	MnO	MgO	CaO	Na2O	K2O	P2O5	NiO
CHS 71	LLR1	1216	-1.59	0.639	56.04	0.08	0.28	0.89	10.63	0.37	29.48	2.11	0.12	bdl	bdl	bdl
CHS 71	CIR1	1216	-1.59	0.645	56.12	0.08	0.28	0.89	10.15	0.33	29.40	2.65	0.09	bdl	bdl	bdl
CHS 71	CIR2b	1216	-1.59	0.686	55.85	0.06	0.31	1.12	9.59	0.41	28.42	4.14	0.11	bdl	bdl	bdl
CHS 71	CI	1216	-1.59	0.719	55.98	0.05	0.32	1.07	9.70	0.55	30.71	1.56	0.10	bdl	bdl	bdl
CHS 70	CI	1167	-1.84	0.524	55.09	0.10	0.62	1.13	9.39	0.60	28.64	4.32	0.13	bdl	bdl	bdl
CHS 70	RCa2	1167	-1.84	0.585	55.56	0.05	0.81	0.89	9.44	0.34	27.80	4.97	0.13	bdl	bdl	bdl
CHS 69	H	1301	-2.44	0.871	58.29	0.05	0.22	0.63	4.09	0.40	35.83	0.49	0.00	bdl	bdl	bdl
CHS 69	RCa	1301	-2.44	0.81	56.50	0.03	0.12	0.66	5.28	0.39	32.29	4.69	0.05	bdl	bdl	bdl
CHS 69	RCa2	1301	-2.44	0.815	57.39	0.01	0.14	0.87	4.26	0.33	32.38	4.58	0.03	bdl	bdl	bdl
CHS 69	CIR2b	1301	-2.44	0.809	57.29	0.04	0.16	0.92	4.24	0.45	34.16	2.69	0.04	bdl	bdl	bdl
CHS 69	CI	1301	-2.44	0.878	57.67	0.06	0.19	0.80	5.34	0.43	35.08	0.40	0.02	bdl	bdl	bdl
CHS 68	CI	1159	-1.53	0.569	55.11	0.09	0.47	0.84	12.64	0.53	26.16	4.05	0.10	bdl	bdl	bdl
CHS 68	CIR2b	1159	-1.53	0.557	54.82	0.12	0.47	0.84	11.82	0.43	26.83	4.52	0.14	bdl	bdl	bdl
CHS 68	RCa	1159	-1.53	0.551	55.20	0.04	0.97	0.88	10.47	0.34	25.82	6.09	0.16	bdl	bdl	bdl
CHS 67	CIR2b	1274	-1.586	0.75	55.61	0.05	0.20	0.97	9.55	0.39	30.43	2.72	0.08	bdl	bdl	bdl
CHS 67	CIR1	1274	-1.586	0.75	55.93	0.04	0.11	0.76	8.94	0.40	30.14	3.59	0.07	bdl	bdl	bdl
CHS 66	CIR1	1250	-2.535	0.713	57.19	0.11	0.26	0.76	4.07	0.40	34.57	2.55	0.08	bdl	bdl	bdl
CHS 66	CIR2b	1250	-2.535	0.709	56.81	0.07	0.33	0.99	3.71	0.47	33.82	3.73	0.09	bdl	bdl	bdl
CHS 66	RCa	1250	-2.535	0.711	57.08	0.02	0.39	0.74	3.25	0.44	32.62	5.38	0.08	bdl	bdl	bdl
CHS 65	CIR2b	1201	-2.27	0.598	56.17	0.10	0.66	1.10	5.19	0.49	32.14	4.06	0.12	bdl	bdl	bdl
CHS 65	CIR1	1201	-2.27	0.583	56.69	0.12	0.43	0.86	5.10	0.40	32.86	3.46	0.06	bdl	bdl	bdl
CHS 65	RCa	1201	-2.27	0.579	56.52	0.05	0.72	0.84	4.68	0.51	32.05	4.53	0.09	bdl	bdl	bdl
CHS 64	LL	1202	-1.08	0.639	54.46	0.02	0.35	0.78	15.21	0.32	26.76	2.08	0.03	bdl	bdl	bdl
CHS 63	H	1202	-1.375	0.637	54.83	0.08	0.44	0.92	12.78	0.45	28.68	1.82	0.02	bdl	bdl	bdl
CHS 63	CIR2b	1202	-1.375	0.634	54.36	0.08	0.44	1.07	11.19	0.40	26.74	5.61	0.10	bdl	bdl	bdl
CHS 63	CI	1202	-1.375	0.643	55.21	0.01	0.42	1.01	11.67	0.51	28.69	2.37	0.07	bdl	bdl	bdl
CHS 62	CI	1192	-2.02	0.638	54.57	0.10	0.59	1.07	10.75	0.57	29.17	3.01	0.13	bdl	bdl	bdl
CHS 61	H	1301	-1.77	0.823	57.12	0.01	0.24	0.67	8.06	0.41	32.59	0.85	0.05	bdl	bdl	bdl
CHS 60	CI	1104	-1.63	0.394	54.97	0.09	0.56	0.81	11.40	0.56	28.78	2.68	0.14	bdl	bdl	bdl
CHS 54	CI	1129	-1.55	0.471	54.28	0.10	0.73	0.80	12.72	0.55	26.16	4.62	0.13	bdl	bdl	bdl
CHS 54	CIR1	1129	-1.55	0.507	54.39	0.11	0.61	0.71	13.76	0.36	26.40	3.55	0.11	bdl	bdl	bdl
CHS 53	CIR1	1250	-1.47	0.708	54.87	0.06	0.28	0.85	11.29	0.33	29.98	2.28	0.06	bdl	bdl	bdl
CHS 53	CIR2a	1250	-1.47	0.719	55.42	0.03	0.13	0.81	10.49	0.38	27.56	5.08	0.09	bdl	bdl	bdl
CHS 51	CIR2a	1217	-1.582	0.673	55.02	0.08	0.32	0.89	10.88	0.39	26.96	5.32	0.12	bdl	bdl	bdl
CHS 51	CIR1	1217	-1.582	0.64	55.27	0.09	0.33	0.92	10.88	0.35	28.78	3.32	0.08	bdl	bdl	bdl
CHS 50	CIR1	1248	-1.89	0.728	56.53	0.07	0.32	0.71	7.64	0.35	31.66	2.66	0.06	bdl	bdl	bdl
CHS 50	CIR2a	1248	-1.89	0.723	56.33	0.06	0.24	0.70	7.23	0.43	29.95	4.96	0.10	bdl	bdl	bdl
CHS 49	CIR1	1248	-1.62	0.722	55.71	0.07	0.25	0.81	10.74	0.31	29.77	2.27	0.05	bdl	bdl	bdl
CHS 49	CIR3	1248	-1.62	0.701	55.71	0.04	0.17	0.84	10.20	0.40	29.60	2.91	0.08	bdl	bdl	bdl
CHS 49	LLR2	1248	-1.62	0.725	56.19	0.05	0.15	0.84	9.72	0.42	30.17	2.14	0.09	bdl	bdl	bdl
CHS 48	CIR1	1194	-1.98	0.594	56.25	0.11	0.49	0.91	7.56	0.37	30.71	3.49	0.08	bdl	bdl	bdl
CHS 48	H	1194	-1.98	0.613	56.47	0.12	0.51	0.86	8.12	0.49	31.08	2.29	0.05	bdl	bdl	bdl
CHS 47	CIR1	1200	-1.63	0.624	55.38	0.08	0.38	0.90	11.17	0.35	27.98	3.69	0.09	bdl	bdl	bdl
CHS 46	CIR1	1191	-1.61	0.603	54.58	0.09	0.47	0.92	12.65	0.37	27.56	3.24	0.11	bdl	bdl	bdl
CHS 45	LLR1	1182	-1.6	0.541	54.64	0.10	0.48	0.96	11.72	0.37	28.10	3.51	0.10	bdl	bdl	bdl
CHS 44	H	1161	-1.86	0.551	55.48	0.12	0.43	0.85	9.27	0.55	29.88	3.29	0.11	bdl	bdl	bdl
CHS 43	H	1163	-2.06	0.503	55.68	0.15	0.65	0.81	6.99	0.57	31.44	3.56	0.12	bdl	bdl	bdl
CHS 42	H	1085	-1.65	0.341	54.47	0.09	0.47	0.84	12.36	0.46	28.99	2.41	0.08	bdl	bdl	bdl
CHS 41	H	1142	-2	0.475	55.46	0.12	0.59	0.78	9.53	0.60	30.46	2.38	0.10	bdl	bdl	bdl
CHS 40	H	1108	-1.81	0.379	54.90	0.13	0.51	0.75	11.06	0.67	29.64	2.55	0.13	bdl	bdl	bdl
CHS 38	LL	1086	-1.3	0.375	54.60	0.08	0.47	0.62	13.90	0.41	27.40	2.39	0.12	bdl	bdl	bdl
CHS 35	H	1079	-1.04	0.368	53.24	0.07	0.39	0.57	18.33	0.38	25.45	1.45	0.07	bdl	bdl	bdl
CHS 33	H	1112	-1.18	0.413	52.79	0.13	0.76	0.64	17.72	0.37	22.46	4.99	0.13	bdl	bdl	bdl
CHS 34	LL	1108	-1.16	0.429	53.86	0.15	0.67	0.58	15.69	0.40	24.90	3.64	0.14	bdl	bdl	bdl
CHS 31	H	1202	-1.56	0.643	55.09	0.08	0.48	0.78	11.70	0.45	29.27	2.06	0.08	bdl	bdl	bdl
CHS 31	HDep	1202	-1.56	0.626	54.52	0.11	0.48	0.78	11.85	0.41	27.33	4.41	0.10	bdl	bdl	bdl
CHS 26	H	1158	-1.42	0.491	53.59	0.10	0.58	0.73	14.98	0.42	26.51	3.01	0.08	bdl	bdl	bdl
CHS 37	LL	1063	-0.98	0.325	54.16	0.06	0.38	0.52	15.08	0.38	27.13	2.17	0.10	bdl	bdl	bdl
CHS 59	CI	1100	-1.6	0.394	54.97	0.09	0.56	0.81	11.40	0.56	28.78	2.68	0.14	bdl	bdl	bdl
CHS 58	CI	1136	-2.27	0.491	56.15	0.14	0.74	1.11	6.92	0.65	30.57	3.58	0.12	bdl	bdl	bdl

Table S6 experiments used for the calibration of the DCr oliv-LCP thermometer (continued)

Name	Mat	T	dIW	DCr	augite											
					SiO2	TiO2	Al2O3	Cr2O3	FeO	MnO	MgO	CaO	Na2O	K2O	P2O5	NiO
CHS 71	LLR1	1216	-1.59	0.64												
CHS 71	CIR1	1216	-1.59	0.65												
CHS 71	CIR2b	1216	-1.59	0.69												
CHS 71	CI	1216	-1.59	0.72												
CHS 70	CI	1167	-1.84	0.52												
CHS 70	RCa2	1167	-1.84	0.58	52.96	0.10	1.60	1.17	5.92	0.24	19.47	18.30	0.21	bdl	bdl	bdl
CHS 69	H	1301	-2.44	0.87												
CHS 69	RCa	1301	-2.44	0.81	55.22	0.15	0.97	1.01	3.43	0.36	25.66	13.10	0.09	bdl	bdl	bdl
CHS 69	RCa2	1301	-2.44	0.81	55.74	0.09	0.81	1.37	3.21	0.35	25.56	12.78	0.09	bdl	bdl	bdl
CHS 69	CIR2b	1301	-2.44	0.81												
CHS 69	CI	1301	-2.44	0.88												
CHS 68	CI	1159	-1.53	0.57												
CHS 68	CIR2b	1159	-1.53	0.56												
CHS 68	RCa	1159	-1.53	0.55	53.40	0.09	1.44	1.14	6.93	0.24	20.41	16.11	0.24	bdl	bdl	bdl
CHS 67	CIR2b	1274	-1.586	0.75												
CHS 67	CIR1	1274	-1.586	0.75												
CHS 66	CIR1	1250	-2.535	0.71												
CHS 66	CIR2b	1250	-2.535	0.71												
CHS 66	RCa	1250	-2.535	0.71	55.24	0.08	0.75	0.66	2.29	0.33	23.44	17.04	0.16	bdl	bdl	bdl
CHS 65	CIR2b	1201	-2.27	0.60	54.38	0.11	0.98	0.86	2.90	0.34	22.46	17.77	0.19	bdl	bdl	bdl
CHS 65	CIR1	1201	-2.27	0.58												
CHS 65	RCa	1201	-2.27	0.58	54.20	0.12	1.28	0.82	2.78	0.33	21.74	18.53	0.18	bdl	bdl	bdl
CHS 64	LL	1202	-1.08	0.64												
CHS 63	H	1202	-1.375	0.64												
CHS 63	CIR2b	1202	-1.375	0.63												
CHS 63	CI	1202	-1.375	0.64												
CHS 62	CI	1192	-2.02	0.64												
CHS 61	H	1301	-1.77	0.82												
CHS 60	CI	1104	-1.63	0.39	52.90	0.43	2.54	1.69	6.88	0.45	18.60	15.97	0.51	bdl	bdl	bdl
CHS 54	CI	1129	-1.55	0.47												
CHS 54	CIR1	1129	-1.55	0.51												
CHS 53	CIR1	1250	-1.47	0.71												
CHS 53	CIR2a	1250	-1.47	0.72												
CHS 51	CIR2a	1217	-1.582	0.67												
CHS 51	CIR1	1217	-1.582	0.64												
CHS 50	CIR1	1248	-1.89	0.73												
CHS 50	CIR2a	1248	-1.89	0.72												
CHS 49	CIR1	1248	-1.62	0.72												
CHS 49	CIR3	1248	-1.62	0.70												
CHS 49	LLR2	1248	-1.62	0.72												
CHS 48	CIR1	1194	-1.98	0.59												
CHS 48	H	1194	-1.98	0.61												
CHS 47	CIR1	1200	-1.63	0.62												
CHS 46	CIR1	1191	-1.61	0.60												
CHS 45	LLR1	1182	-1.6	0.54												
CHS 44	H	1161	-1.86	0.55												
CHS 43	H	1163	-2.06	0.50												
CHS 42	H	1085	-1.65	0.34	52.23	0.45	1.65	1.42	8.45	0.40	18.80	16.12	0.46	bdl	bdl	bdl
CHS 41	H	1142	-2	0.48	53.18	0.35	1.38	1.17	7.23	0.51	26.05	9.79	0.34	bdl	bdl	bdl
CHS 40	H	1108	-1.81	0.38												
CHS 38	LL	1086	-1.3	0.37												
CHS 35	H	1079	-1.04	0.37	52.16	0.40	1.63	1.09	11.88	0.27	16.56	15.44	0.42	bdl	bdl	bdl
CHS 33	H	1112	-1.18	0.41												
CHS 34	LL	1108	-1.16	0.43												
CHS 31	H	1202	-1.56	0.64												
CHS 31	HDep	1202	-1.56	0.63												
CHS 26	H	1158	-1.42	0.49												
CHS 37	LL	1063	-0.98	0.33	51.80	0.39	1.96	1.01	12.01	0.35	16.42	15.64	0.41	bdl	bdl	bdl
CHS 59	CI	1100	-1.6	0.39	52.90	0.43	2.54	1.69	6.88	0.45	18.60	15.97	0.51	bdl	bdl	bdl
CHS 58	CI	1136	-2.27	0.49	53.54	0.29	1.46	1.25	5.27	0.39	18.95	18.43	0.41	bdl	bdl	bdl

Table S7a

Olivine composition of ureilites analyzed for this study

	SiO2	TiO2	Al2O3	Cr2O3	FeO	MnO	MgO	CaO	Total	n	Fo							
LAP 03721	38.11	0.3	bdl	bdl	0.63	0.08	22.82	0.15	0.40	0.02	38.80	0.47	0.33	0.04	101.1	17	75.2	ol-pig
EET 90019	39.98	0.2	bdl	bdl	0.64	0.04	10.28	0.11	0.45	0.03	48.68	0.24	0.34	0.07	100.4	10	89.4	ol-pig
NWA 4852	39.47	0.3	bdl	bdl	0.64	0.03	12.05	0.15	0.45	0.03	47.25	0.31	0.31	0.01	100.2	15	87.5	ol-pig
MIL 090076	38.37	0.1	bdl	bdl	0.73	0.06	19.92	0.21	0.43	0.05	41.58	0.14	0.36	0.03	101.5	7	78.8	ol-pig
MIL 07447_10	39.28	0.3	bdl	bdl	0.71	0.02	16.82	0.07	0.41	0.05	43.72	0.12	0.34	0.01	101.3	9	82.2	ol-pig
DOM 08012_15	39.09	0.1	bdl	bdl	0.71	0.03	20.55	0.15	0.44	0.03	39.92	0.12	0.36	0.02	101.1	5	77.6	ol-pig
EET 96042_60	39.34	0.1	bdl	bdl	0.84	0.05	17.40	0.10	0.43	0.03	42.92	0.11	0.36	0.02	101.4	10	81.5	ol-pig
NWA 11755	38.32	0.3	bdl	bdl	0.73	0.04	20.31	0.09	0.40	0.03	41.03	0.20	0.36	0.02	101.2	13	78.3	ol-pig
NWA 5555	40.35	0.1	bdl	bdl	0.64	0.03	8.86	0.07	0.44	0.04	49.44	0.15	0.31	0.02	100.1	14	90.9	ol-pig-opx
ALHA 82106	40.81	0.3	bdl	bdl	0.58	0.02	4.49	0.08	0.53	0.03	53.79	0.23	0.28	0.02	100.5	6	95.5	ol-opx-aug
NWA 11754"	39.66	0.1	bdl	bdl	0.51	0.02	12.03	0.06	0.53	0.04	47.23	0.16	0.29	0.01	100.3	12	87.5	ol-opx-aug
EET 96293"	39.33	0.1	bdl	bdl	0.48	0.05	12.34	0.10	0.56	0.03	46.93	0.11	0.29	0.02	101.4	10	87.1	ol-opx-aug

"Hughes cluster samples

numbers in italic represent the analytical standard errors (1 σ)

bdl: below detection limit. Detection limits as follow (in ppm): Si 195, Ti 296, Al 161, Cr 406, Fe 223, Mn 217, Mg 212, Ca 145, Na 459, K 111, P 254, Ni 341

Table S7b

Pigeonite composition of ureilites analyzed for this study

	SiO2		TiO2		Al2O3		Cr2O3		FeO		MnO		MgO		CaO		Na2O		Total	n	Mg#	Wo
LAP 03721	53.98	0.7	0.05	0.01	0.93	0.21	1.22	0.22	13.29	0.32	0.40	0.02	25.84	1.10	5.27	1.19	0.07	0.02	101.1	14	77.6	10.2
EET 90019	56.27	0.3	0.12	0.02	0.81	0.02	1.04	0.06	6.27	0.12	0.48	0.03	31.04	0.24	4.77	0.07	0.07	0.03	100.9	10	89.8	9.0
NWA 4852	56.33	0.2	0.11	0.02	0.75	0.01	1.04	0.04	7.26	0.10	0.47	0.02	30.27	0.21	4.36	0.11	0.05	0.03	100.6	11	88.1	8.4
MIL 090076	54.69	0.3	0.10	0.01	0.95	0.07	1.39	0.05	11.55	0.11	0.42	0.01	27.01	0.24	5.54	0.10	0.10	0.04	101.7	16	80.7	10.6
MIL 07447_10	55.01	0.4	0.08	0.02	0.89	0.01	1.25	0.03	9.78	0.09	0.45	0.02	26.93	0.10	5.28	0.09	bdl		99.7	9	83.1	10.5
DOM 08012_15	55.64	0.1	0.04	0.03	0.51	0.03	1.08	0.04	12.33	0.03	0.40	0.02	26.52	0.01	3.56	0.02	bdl		100.2	3	79.3	7.1
EET 96042_60	55.54	0.1	0.11	0.01	0.73	0.02	1.37	0.01	10.18	0.11	0.46	0.01	27.01	0.11	4.58	0.04	0.11	0.03	100.1	7	82.5	9.1
NWA 11755	55.38	0.4	0.04	0.02	0.44	0.11	1.01	0.09	11.74	0.33	0.41	0.02	27.69	0.36	4.28	0.31	bdl		101.0	24	80.8	8.2
NWA 5555	56.51	0.5	0.12	0.01	0.55	0.02	1.03	0.03	5.51	0.06	0.47	0.02	30.92	0.13	4.53	0.05	bdl		99.7	10	90.9	8.7
ALHA 82106*	57.16	0.16			0.54		0.82		2.74		0.45		33.66		5.72		0.08		101.3	n.a.	95.6	10.5

* average non-inverted pigeonite (orthopyroxene+augite exsolutions)

Table S7c
Orthopyroxene composition of ureilites analyzed for this study

	SiO2		TiO2		Al2O3		Cr2O3		FeO		MnO		MgO		CaO		Na2O		Total	n	Mg#	Wo
NWA 5555	57.20	0.9	0.13	0.01	0.57	0.02	0.95	0.04	5.41	0.04	0.42	0.01	32.92	0.10	2.53	0.02	bdl		100.2	8	91.6	4.8
ALHA 82106	57.57	0.3	0.13	0.01	0.47	0.01	0.81	0.02	2.95	0.08	0.47	0.03	36.16	0.23	2.62	0.02	bdl		101.2	5	95.6	4.8
NWA 11754"	55.97	0.5	0.15	0.01	1.21	0.02	0.96	0.04	7.25	0.11	0.51	0.02	30.96	0.13	2.52	0.02	bdl		99.5	12	88.4	4.9
EET 96293"	56.31	0.2	0.13	0.02	1.24	0.02	1.09	0.02	7.45	0.08	0.52	0.02	32.14	0.21	2.53	0.03	0.05	0.02	101.5	10	88.5	4.8

"Hughes cluster samples

Table S7d
Augite composition of ureilites analyzed for this study

	SiO2		TiO2		Al2O3		Cr2O3		FeO		MnO		MgO		CaO		Na2O		Total	n	Mg#	Wo
ALHA 82106	55.41	0.1	0.28	0.01	0.85	0.03	0.88	0.03	1.84	0.05	0.36	0.01	23.00	0.13	18.92	0.05	0.30	0.03	101.8	11	95.7	36.1
NWA 11754"	53.17	0.5	0.28	0.02	1.91	0.04	1.20	0.03	4.17	0.05	0.39	0.02	20.48	0.14	18.25	0.09	0.20	0.02	100.0	5	89.7	36.5
EET 96293"	53.75	0.0	0.27	0.03	1.90	0.06	1.35	0.05	4.22	0.07	0.40	0.02	20.44	0.13	18.78	0.03	0.23	0.03	101.3	5	89.6	37.2

"Hughes cluster samples

Table S8: phase compositions of ureilites from the literature
see .zip folder