Characterization of matrix material in Northwest Africa 5343: Weathering and thermal metamorphism of the least equilibrated CK chondrite

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Abstract—Based on the chemical heterogeneity of chondrule and matrix olivine, Northwest Africa (NWA) 5343 is the least metamorphosed CK chondrite reported so far. To better constrain the lower limit of metamorphism in the CK chondrites, we performed a detailed analysis of matrix material in NWA 5343, including characterization of the texture and bulk composition and analyses of individual silicate minerals. Results suggest that NWA 5343 is petrologic type 3.6 or 3.7. Although silicate minerals in the matrix seem to be equilibrated to roughly the same extent throughout the sample, there are recognizable differences in grain size and shape. These textural differences may be the result of transient heating events during impacts, which would be likely on the CK chondrite parent body. The difference between the extent of chemical equilibration and texture may also suggest that grain size and shape are still sensitive to metamorphism at petrologic subtypes where silicate mineral equilibration is nearly complete (e.g., >3.7). Carbonate material present in NWA 5343 is a product of terrestrial weathering; however, infiltration of a Ca-bearing fluid did not influence the composition of silicate minerals in the matrix. To evaluate the possibility of a continuous metamorphic sequence between the CV and CK chondrites, the bulk matrix composition of NWA 5343 is compared to the CVred chondrite, Vigarano. Although the matrix composition of NWA 5343 could be derived by secondary processing of a Vigarano-like precursor, porosity and texture of matrix olivine in NWA 5343 are hard to reconcile with a continuous metamorphic sequence.

INTRODUCTION

The Karoonda-like (CK) chondrites are a group of highly oxidized carbonaceous chondrites characterized by the presence of nickel-rich matrix olivine (>0.2 wt% NiO) and Cr$_2$O$_3$-rich magnetite (>3 wt% Cr$_2$O$_3$) (Kallemeyn et al. 1991; Noguchi 1993; Geiger and Bischoff 1995). The CK chondrites contain abundant matrix material and are dominated by porphyritic, Fe-rich olivine chondrules (Kallemeyn et al. 1991; Rubin 2010; Chaumard and Devouard 2016). Bulk lithophile elemental abundances and oxygen isotopes of the CK chondrites are similar to those of the Vigarano-like (CV) chondrites (Greenwood et al. 2010). These similarities led Greenwood et al. (2010) to suggest that the CV and CK chondrites originated from a single, thermally stratified (i.e., onion-shell) parent asteroid. The single parent body model has since gained support, though alternatives to the onion-shell model have been proposed (e.g., Chaumard et al. 2012, 2014; Wasson et al. 2013; Chaumard and Devouard 2016). However, Dunn et al. (2016a) contended that the single parent body model is unlikely, and they showed that compositions of magnetite in the CVs and CKs could not be reconciled by a continuous metamorphic sequence (i.e., CV3 → CK3 → CK4-6). Most recently, Yin et al. (2017) also argued against a single parent body model based on Cr isotopic compositions, which are clearly distinguishable between the CV and CK chondrites.
One significant difference between the CV and CK chondrite groups is the degree of metamorphism experienced by each. All CV chondrites are classified as petrologic type 3 (McSween 1977; Bonal et al. 2006), while CK chondrites exhibit the full range of thermal metamorphism, from type 3 to type 6 (Kallemeyn et al. 1991; Geiger et al. 1993). Most CK chondrites are equilibrated (petrologic types 4–6). However, 35 unequilibrated (type 3) CK chondrites have been identified since the first CK3 chondrite, Watson 002, was discovered in 1991 (Geiger et al. 1993) (see the Meteoritical Bulletin for a current list of CK3s). In meteorites that have experienced a low degree of metamorphism (i.e., petrologic type 3), subtle differences in the extent of thermal processing can be recognized. Resultantly, meteorites of this grade can be further divided into petrologic subtypes (i.e., type 3.0–3.9) (Sears et al. 1980, 1991; Chizmadia et al. 2002). It has been suggested that the unequilibrated CKs are all petrologic type 3.7 or higher based on abundances and compositions of refractory inclusions (Greenwood et al. 2010), and six unequilibrated CK chondrites have been assigned petrologic type 3.8 or 3.9 (Chaumard et al. 2014; Dunn et al. 2016a, 2016b). In contrast, determining the petrologic subtypes of the CV chondrites is complicated by their complex alteration histories, which involve fluid-assisted metamorphism and metasomatism (Brearley and Krot 2013). Although approaches based on petrography are fairly consistent (suggesting that the CVs are petrologic subtype 3.3 or lower) (Guimon et al. 1995; Grossman and Brearley 2005), Raman spectroscopy of organic material yields petrologic subtypes ≥3.6 for the CV chondrites Allende, Axtel, Bali, Mokoia, and Grosnaja (Bonal et al. 2006). It has been suggested that these inconsistencies are due to decoupling of organic matter and silicates during metamorphism (Pearson et al. 2007). Despite the uncertainty, the general consensus is that CV chondrites have experienced lower degrees of metamorphism (≤ subtype 3.6) than the unequilibrated CK chondrites (≥ subtype 3.7).

In our efforts to determine the petrologic subtypes of the unequilibrated CK chondrites, we have analyzed chondrule and matrix olivine in several CK3 and CK4 chondrites (Dunn et al. 2016b). Consistent with previous observations (e.g., Chaumard and Devouard 2016), the homogeneity and chemical composition of chondrule olivine suggests that most of these CK3s have been metamorphosed to at least petrologic subtype 3.8 conditions. However, heterogeneity of olivine in Northwest Africa 5343 (NWA 5343) suggests that it is less metamorphosed than other CK chondrites (Chaumard and Devouard 2016; Dunn et al. 2016b). Accurate determination of petrologic subtypes is critical when assessing the onion-shell model. Therefore, to better constrain the lower limit of metamorphism in the CK chondrites, we performed a detailed analysis of matrix material in NWA 5343. Here we use “metamorphism” to refer to all materials that occupy the space between chondrite components (e.g., chondrules). Following the procedures of Hurt et al. (2012), we characterized the texture and bulk composition of matrix. We also analyzed individual silicate minerals (olivine, pyroxene, and plagioclase), as these phases have been shown to record the effects of metamorphism in unequilibrated ordinary chondrites (e.g., Dodd and Van Schmus 1967; Huss et al. 1981). Finally, to assess possible links with the CV chondrites, we have compared the bulk matrix composition of NWA 5343 to that of CV_red chondrite Vigarano.

**METHODOLOGY**

The polished thin section examined in this study was prepared from a 36.7 g slice of NWA 5343, which was purchased from Mirko Graul in 2014. This thin section (NWA 5343_01) and the remaining sample mass are housed in the meteorite collection at Colby College. Backscattered electron (BSE) images were obtained using the JXA JEOL 8200 electron microprobe (EMP) at Rutgers University. These images were used to determine the textural characteristics of the sample and to measure porosity. We measured grain sizes and calculated porosity using the image processing software ImageJ. More than 300 grains were measured (along the long axis) throughout the sample. Porosity was determined by thresholding an image to highlight the pore space, converting the images to black and white, and then calculating the thresholded area of the image.

Backscattered electron images were compiled to make a mosaic image of the entire thin section (Fig. 1). We superimposed a 1 mm by 1 mm grid, with rows labeled 1–24 and columns labeled A–X, onto the mosaic BSE image (data not shown on Fig. 1) and selected five areas (L14, I21, K5, S6, and U12) for additional study. The selected areas are illustrated on Fig. 1, and BSE images of each of the five areas are provided in the supporting information Figs. S1 and S2. Wavelength-dispersive (WDS) and energy-dispersive (EDS) elemental Kα X-ray intensity maps for Fe, Al, Ca, S, and Mg (WDS) as well as Si, K, Na, and Cr (EDS) were obtained using the EPMA JEOL 8200 at Rutgers University. These maps were collected using 15 kV accelerating voltage, 25 nA beam current, 1 μm focused beam, a pixel step size of 2 μm, and a dwell time of 55 ms. X-ray maps were used to identify mineral phases in regions S6, K5, U12, L14, and I21.

Quantitative mineral chemical compositions were also determined using the JEOL 8200 EMP. Operating
conditions were: 15 kV accelerating voltage; 20 nA beam current; focused electron beam (1 μm in size) for olivine, pyroxene, and magnetite; defocused electron beam (3 μm) for plagioclase; and peak and background counting times of 10–40 s per element. Analytical standards used to determine both mineral compositions and bulk matrix composition were well-characterized synthetic oxides and natural minerals including albite (Si, Na), olivine (Mg, Fe), Ni-olivine (Ni), orthoclase (K), plagioclase (Al), chromite (Cr), troilite (S), anorthite (Ca), rutile (Ti), and rhodonite (Mn). Data quality was ensured by analyzing standard materials as unknowns.

Bulk composition of the matrix was determined using grid analyses similar to that implemented by Wasson and Rubin (2009) and Hurt et al. (2012). In this technique, several square grids measuring 150 by 150 μm were analyzed within the regions S6, K5, U12, L14, and I21. Each grid area was analyzed with a step size of 10 μm between each point and background counting times of 10–40 s per element. We used standard, fully matrix-corrected, wavelength-dispersive analyses. For further discussion, see Wasson and Rubin (2009). We used 10 μm beam at 15 nA to measure 11 elements: Na, Mg, Al, Si, S, K, Ca, Cr, Mn, Ni, and Fe. Detection limits were 0.01 wt% for Al, Mg, Si, Cr, S, K, Ti, and Ca; 0.02 wt% for Na, Ni; 0.05 wt% for Mn; and 0.08 wt% for Fe. Data quality was ensured by analyzing standard materials as unknowns.

**RESULTS**

**Petrography of the Matrix**

Two distinct regions of matrix are visible in the BSE image of the thin section (Fig. 1). In the first region, located predominately in the southwest quadrant of the thin section, the matrix is light gray and continuous. Matrix material in the second region, located primarily in the northwest quadrant, is darker and more porous. There is a sharp contact between these two matrix regions (Fig. 1 insert). We will refer to these regions henceforth as the BSE-light matrix and the BSE-dark matrix. Detailed inspection of areas within the BSE-light and BSE-dark matrix indicates that there are noteworthy textural differences between the two regions. BSE-dark matrix material (areas K5, S6, and U12) mainly consists of subhedral to anhedral grains of olivine with an average grain size of 19.5 ± 16.5 μm (Fig. 2). Minor, euhedral, high-Ca pyroxene (16.1 ± 9.5 μm) and euhedral plagioclase grains (18.3 ± 8.6 μm) are also present; however, olivine is the
dominant phase. Grains are separated by pore space, which is sometimes filled with submicron-sized silicates and oxides (Fig. 2). Porosity of areas within the darker matrix ranges from 10 to 14%. Much of the BSE-dark matrix is typical of a breccia and is similar to textures seen in the CV chondrite Vigarano (Abreu and Brearley 2011). We also identified a 1.5 × 3 mm area within the BSE-dark region that is characterized by smaller (<10 μm), more angular olivine grains intermixed with a few large olivine clasts (90–100 μm) (Fig. 3). This area most likely represents a crushed chondrule rather than actual matrix material. Fragmental matrix textures are common in the CK chondrites and have been attributed to impact-induced crushing events prior to metamorphism (Noguchi 1993; Wasson et al. 2013). Some of the larger grains adjacent to the crushed chondrule exhibit characteristic 120° triple junctions that are attributed to thermally driven metamorphism (Fig. 3 insert).

Fig. 2. Representative areas of the BSE-dark matrix. This matrix is dominated by subhedral to anhedral olivine (olv), mostly < 30 μm in size. Some euhedral high-Ca pyroxene (cpx) and plagioclase (plag) is also present.

The BSE-light matrix (areas I21 and L14) consists mostly of anhedral olivine grains surrounded by an interstitial Ca-rich material that is responsible for the light color of this area in BSE images (Fig. 4). This material, identified as calcium carbonate using EDS, is a common terrestrial weathering product in desert regions. The source of this interstitial material is a large vein (150–300 μm wide) in area L14 (Fig. 5). Skeletal pyroxene, more variable in size than the olivine (18.3 ± 13.1 μm), and large (55.7 ± 21 μm), subhedral plagioclase grains are also present in CaCO₃ vein (Fig. 5). We also observed a few small apatite crystals within the vein (not shown in Fig. 5) that most likely represent an alteration product. Porosity in this region ranges from 3 to 7%. When carbonate is present in the pore spaces, it often contains magnetite grains that are slightly larger (1–3 μm) than the magnetite in the BSE-dark matrix. Olivine in the BSE-light matrix is also larger (30.5 ± 14.9 μm) and more anhedral than olivine in the BSE-dark matrix. Despite textural differences, olivine grains in both the BSE-dark and BSE-light matrix contain submicron voids and oxides similar to those described by Brearley (2009) (Fig. 6).

**Bulk Composition of the Matrix**

**Rules for Discarding Points**

In previous studies of extremely fine-grained primitive nebular matrix material in CR and CV chondrites (Wasson and Rubin 2009; Hurt et al. 2012), analyses with extreme values were discarded to avoid the contributions of coarse nonnebular mineral grains. However, here our aim was to study the coarse, recrystallized matrix material, and even the finest grained material present in some interstices in the BSE-dark matrix appears to be recrystallized. Thus, we based our rules for discarding points solely on oxide totals and discarded all analyses with totals <85 wt% under the assumption that totals below this threshold represented voids rather than mineral phases. In general, areas in BSE-dark matrix had a higher proportion of discarded points (54–68%) than areas in the BSE-light matrix (33–38%). The number of discarded points is consistent with differences in porosity, which ranges from 10 to 14% in the BSE-dark matrix and from 3 to 7% in the BSE-light matrix. The lower porosity in the BSE-light matrix is an artifact of the interstitial calcium carbonate material. Thus, the porosity of the BSE-light matrix originally would have
likely been closer to that of the BSE-dark matrix prior to terrestrial alteration.

**Compositional Variability**

Insights into the compositional variability of the bulk matrix in NWA 5343 can be examined using scatter diagrams of major and minor oxides. Bulk compositions of BSE-light and BSE-dark matrix in eight representative grids are plotted on Fig. 7. Because the matrix of NWA 5343 is relatively coarse-grained, compositional trends are driven by mineralogy. As illustrated Figs. 7a and 7b, olivine is the dominant phase present, followed by high-Ca pyroxene. The effect of the interstitial CaCO$_3$ material on the bulk composition of the BSE-light matrix is illustrated in Fig. 7b by the negative correlation between SiO$_2$ and CaO. Data points with a higher CaO content reflect a higher proportion of CaCO$_3$ material in an individual
analysis (Fig. 7b). Some plagioclase is also present in the matrix, though it is a minor phase compared to olivine and pyroxene (Fig. 7c). Sulfur and NiO are both present at low abundances (<1 wt%), suggesting that pentlandite is not a prominent matrix phase. As illustrated in Fig. 7d, most bulk matrix compositions plot parallel to, but below, the pentlandite trend line, suggesting that there is an additional source of Ni in the matrix, most likely olivine.

Mean Compositions

Mean composition of each area reflect its dominant mineralogy. Because mineralogy is relatively homogenous throughout the sample, average compositions of most grids cannot be resolved from one another when standard deviations are considered. Grids L14-1 and L14-2 are the only exceptions, as they have mineralogies dominated by high-Ca pyroxene and CaCO₃ material rather than olivine. Mean compositions of all grids, plotted as MgO versus FeO, Al₂O₃ versus CaO, MnO versus Cr₂O₃, and NiO versus S, are provided in Figs. S1 and S2. Table 1 lists the mean composition of each grid in areas U12, K5, S6, L14, and I21, and the mean composition of the BSE-light and BSE-dark matrix. Because the pyroxene-dominated mineralogy of grids L14-1 and L14-2 is not representative of the overall matrix mineralogy, we have excluded these grids from the calculated mean of the BSE-light matrix. With the exception of CaO, Al₂O₃, and Cr₂O₃, the mean compositions of the BSE-dark and BSE-light matrix are nearly identical. The most significant difference is CaO content, which is 2.2× higher in the BSE-light matrix due to the presence of
the CaCO₃ weathering material in this region. Conversely, Al₂O₃ and Cr₂O₃ are higher in the BSE-dark matrix (1.2× and 0.66× higher, respectively), most likely due to compositional differences between high-Ca pyroxene in the BSE-dark matrix and the BSE-light matrix (see the Discussion section).

**Individual Mineral Phases**

Representative compositions of silicate minerals in the BSE-dark and BSE-light matrix are reported in Table 2. The Fa content of matrix olivine is plotted as a histogram in Fig. 8, and compositions of high-Ca pyroxenes are plotted in Fig. 9.

**Olivine**

Matrix olivine ranges in composition from Fa₃₀ to Fa₃₈ in the BSE-dark matrix and from Fa₃₅ to Fa₄₀ in the BSE-light matrix. The average composition of olivine is the same in both the matrix regions (Fa₃₆.₃ and Fa₃₆.₂, respectively). As is typical for CK chondrites, matrix olivine is nickel-rich (average of 0.31 wt% NiO in BSE-
NiO content of olivine is more homogeneous in the BSE-light matrix (0.24–0.48 wt%) than in the BSE-dark matrix (0.23–0.67 wt%). CaO is also slightly more homogenous in the BSE-light matrix. There is no discernible difference between average abundances and heterogeneity of MgO, SiO2, and MnO in the BSE-light and BSE-dark matrix. Compositions of matrix olivine in NWA 5343 are consistent with other CK3 chondrites we have analyzed in our previous work (Dunn et al. 2016b).

### Pyroxene

All pyroxene in the matrix of NWA 5343 is diopside (En32–54Wo46–50) (Fig. 9). This is consistent with previous studies of CK chondrites, in which diopside is the dominant, or only, pyroxene identified (e.g., Kallemeyn et al. 1991). The average composition of diopside is the same in both the matrix regions (En40Fs11Wo49) and is identical to that of the CK3 chondrite NWA 1628 (En41Fs10Wo49) (Brearley 2009). The Fs content of high-Ca pyroxene in NWA 5343 is also consistent with the CK4 chondrites Karoonda, Maralinga, and Dhofar 015 (Fs10–11) (Noguchi 1993; Ivanova et al. 2000). The heterogeneity of high-Ca pyroxene is the same in BSE-light and BSE-dark material (Fs6–18) and is similar to high-Ca pyroxene in the CK3 chondrite Watson 002 (Fs7–20) (Geiger et al. 1993). Also, like Watson 002, pyroxene in NWA 5343 is more heterogeneous than other CK chondrites.

<table>
<thead>
<tr>
<th>Points</th>
<th>Na2O</th>
<th>Al2O3</th>
<th>MgO</th>
<th>SiO2</th>
<th>NiO</th>
<th>Cr2O3</th>
<th>S</th>
<th>K2O</th>
<th>CaO</th>
<th>FeO</th>
<th>MnO</th>
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</tr>
</thead>
<tbody>
<tr>
<td>U12-1</td>
<td>0.05</td>
<td>0.19</td>
<td>0.10</td>
<td>0.40</td>
<td>0.30</td>
<td>0.16</td>
<td>0.02</td>
<td>2.78</td>
<td>30.62</td>
<td>0.13</td>
<td>88.37</td>
<td></td>
</tr>
<tr>
<td>BSE-dark</td>
<td>0.07</td>
<td>0.35</td>
<td>0.34</td>
<td>0.41</td>
<td>0.27</td>
<td>0.14</td>
<td>0.07</td>
<td>1.18</td>
<td>30.96</td>
<td>0.14</td>
<td>88.56</td>
<td></td>
</tr>
<tr>
<td>U12-2</td>
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<td>0.23</td>
<td>0.22</td>
<td>0.30</td>
<td>0.36</td>
<td>0.21</td>
<td>0.02</td>
<td>2.79</td>
<td>30.92</td>
<td>0.06</td>
<td>92.99</td>
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<tr>
<td>BSE-dark</td>
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<td>0.30</td>
<td>0.39</td>
<td>0.40</td>
<td>0.28</td>
<td>0.15</td>
<td>0.06</td>
<td>1.36</td>
<td>32.89</td>
<td>0.06</td>
<td>94.77</td>
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</tr>
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</table>

*Compositions of L14-1 and L14-2 are excluded from the average bulk composition of the BSE-light matrix, as both areas are unrepresentative of the overall matrix.*

Table 1. Average bulk compositions (wt%) and standard deviations (in italics) of grids in all five areas of the matrix.
olivine (Geiger et al. 1993). However, this is not unexpected, as olivine equilibrates faster than pyroxene during metamorphism (e.g., Huss et al. 1981).

Abundance and heterogeneity of minor elements in pyroxene is also similar in both regions of the matrix. Cr$_2$O$_3$ and NiO are typically below detectable limits, though a few grains contain measurable amounts (up to 0.13 wt% Cr$_2$O$_3$ and 0.26 wt% NiO). Na$_2$O is present at abundances from 0.03 to 0.59 wt%, with an average of 0.20 wt% in the BSE-dark and 0.11 wt% in the BSE-light. Published analyses of diopside in the CK3 chondrites are lacking. However, abundances of minor elements in high-Ca pyroxene are lower in NWA 5343 than in CK4 chondrites Karoonda and Maralinga (Noguchi 1993). The most remarkable difference is in Al$_2$O$_3$, which is present at average abundances of 1.66 and 1.83 wt% in Karoonda and Maralinga, respectively. The average abundance of Al$_2$O$_3$ in NWA 5343 is 0.58 wt% in the BSE-dark matrix and 0.27 wt% in the BSE-light matrix.

### Plagioclase

Plagioclase in NWA 5343 is Na-rich, with compositions of An$_{6.45}$Ab$_{54.91}$Or$_{1.4}$ in the BSE-dark and An$_{14.40}$Ab$_{55.84}$Or$_{1.2}$ in the BSE-light. This is consistent with matrix plagioclase in CK3 chondrites Watson 002 (An$_{7.50}$) (Geiger et al. 1993) and NWA 1628 (An$_{7.37}$) (Brearley 2009). Consistent with previous studies, plagioclase in NWA 5343 is enriched in FeO.

### Table 2. Representative compositions of olivine, high-Ca pyroxene, and plagioclase in the matrix.

<table>
<thead>
<tr>
<th></th>
<th>K5. 1</th>
<th>U12, 5</th>
<th>L14, 66</th>
<th>S6, 35</th>
<th>U12, 42</th>
<th>L14, 7</th>
<th>K5, 25</th>
<th>S6, 43</th>
<th>U12, 20</th>
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<tr>
<td>MgO</td>
<td>31.5</td>
<td>30.6</td>
<td>31.1</td>
<td>11.8</td>
<td>15.4</td>
<td>15.0</td>
<td>nm</td>
<td>nm</td>
<td>nm</td>
</tr>
<tr>
<td>Al$_2$O$_3$</td>
<td>0.02</td>
<td>0.26</td>
<td>&lt;0.01</td>
<td>0.09</td>
<td>0.90</td>
<td>0.76</td>
<td>23.6</td>
<td>22.8</td>
<td>25.7</td>
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<tr>
<td>SiO$_2$</td>
<td>36.6</td>
<td>35.9</td>
<td>36.5</td>
<td>52.6</td>
<td>52.7</td>
<td>53.0</td>
<td>61.1</td>
<td>62.6</td>
<td>58.7</td>
</tr>
<tr>
<td>Na$_2$O</td>
<td>nm</td>
<td>nm</td>
<td>nm</td>
<td>0.07</td>
<td>0.21</td>
<td>0.21</td>
<td>8.39</td>
<td>9.48</td>
<td>7.27</td>
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<tr>
<td>CaO</td>
<td>0.08</td>
<td>0.06</td>
<td>0.10</td>
<td>24.4</td>
<td>24.6</td>
<td>24.9</td>
<td>4.9</td>
<td>3.9</td>
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<tr>
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<td>nm</td>
<td>nm</td>
<td>nm</td>
<td>nm</td>
<td>nm</td>
<td>nm</td>
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<td>0.32</td>
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<tr>
<td>TiO$_2$</td>
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<td>0.01</td>
<td>0.03</td>
<td>&lt;0.01</td>
<td>0.31</td>
<td>0.57</td>
<td>nm</td>
<td>nm</td>
<td>nm</td>
</tr>
<tr>
<td>Cr$_2$O$_3$</td>
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<td>0.05</td>
<td>0.10</td>
<td>&lt;0.01</td>
<td>&lt;0.01</td>
<td>&lt;0.01</td>
<td>nm</td>
<td>nm</td>
<td>nm</td>
</tr>
<tr>
<td>MnO</td>
<td>0.11</td>
<td>0.25</td>
<td>0.30</td>
<td>0.78</td>
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<td>0.09</td>
<td>0.13</td>
<td>&lt;0.05</td>
<td>&lt;0.05</td>
</tr>
<tr>
<td>FeO</td>
<td>31.7</td>
<td>31.3</td>
<td>31.9</td>
<td>10.5</td>
<td>4.5</td>
<td>4.9</td>
<td>0.43</td>
<td>0.15</td>
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</tr>
<tr>
<td>Na$_2$O</td>
<td>nm</td>
<td>nm</td>
<td>nm</td>
<td>0.07</td>
<td>0.04</td>
<td>&lt;0.02</td>
<td>nm</td>
<td>nm</td>
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<tr>
<td>CaO</td>
<td>0.29</td>
<td>0.31</td>
<td>0.28</td>
<td>&lt;0.02</td>
<td>0.04</td>
<td>&lt;0.02</td>
<td>nm</td>
<td>nm</td>
<td>nm</td>
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<tr>
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</tr>
</tbody>
</table>

nm = not measured.

The number following the area name (i.e., K5, 1) indicates the analysis number.

**Fig. 8.** Histogram of olivine compositions in the BSE-light and BSE-dark matrix. The Fa content of matrix olivine ranges from 30.5 to 39.8 mol% Fa and peaks at 36 mol% Fa.

**Fig. 9.** Compositions of high-Ca pyroxene in the BSE-light and BSE-dark matrix of NWA 5343. All pyroxene in the matrix of NWA 5343 is diopside (En$_{32.46}$Wo$_{46.50}$).
Average abundances of FeO in plagioclase are 0.23 wt% in the BSE-dark matrix and 0.13 wt% in the BSE-light matrix; abundances range from 0.04 to 0.64 wt% in all matrix material. These abundances are not as high as those reported from the CK4 chondrite Maralinga (0.75–1.9 wt% FeO) (Keller et al. 1992). Abundances of MgO, NiO, TiO2, Cr2O3, and MnO are below detectable limits. The heterogeneity of plagioclase in NWA 5343 is consistent with other CK chondrites, and is thought to be the result of fractionation during shock melting (Kallemeyn et al. 1991; Rubin 1992).

### Oxides and Sulfides

Sulfides and oxides are present as accessory phases in the matrix of NWA 5343. Magnetite is the predominant oxide, though we also observed very minor chromite and hercynitic spinel. Sulfides are rare, reflecting the low sulfur content of the matrix (0.11 wt%). Although compositional trends indicate that minor pentlandite may be present, we did not encounter any sulfides of sufficient grain size to be analyzed in the matrix of NWA 5343. The sulfur that is present appears to be dispersed, along with oxides, as nm-sized inclusions in matrix olivine. Like other CK3 chondrites, magnetite in NWA 5343 is Cr-rich (2.3–4.1 wt% Cr2O3), Al-rich (2.8–5.0 wt% Al2O3), and contains high abundances of TiO2 (0.9–1.4 wt%) and NiO (0.16–0.22 wt%) (Dunn et al. 2016a). Magnetite in NWA 5343 plots in the CK3 chondrite field of our compositional diagrams for CV and CK chondrites (see figs. 4 and 5 in Dunn et al. 2016a).

### DISCUSSION

#### Comparison of Matrix and Whole-Rock Compositions

In Fig. 10, we plot Mg- and bulk-CK-normalized abundance ratios of eight elements in the BSE-dark and BSE-light matrix. Grids L14-1 and L14-2 are excluded from the average composition of the BSE-light region. The mean CK bulk composition was determined using bulk elemental abundances in two CK4 falls (Kobe and Karoonda) and five relatively unweathered CK4 chondrites (Oura et al. 2002; Huber et al. 2006; Greenwood et al. 2010). In NWA 5343, abundance ratios of Al, Cr, Ni, and Na are depleted in the matrix relative to the whole-rock, while abundance ratios of Fe and Mn are near unity. Relative to whole-rock, abundance ratios of Ca are significantly enriched in the BSE-light matrix and slightly depleted in the BSE-dark matrix. Conversely, abundance ratios of K are slightly depleted in the BSE-light matrix and slightly enriched in the BSE-dark matrix. However, if we average abundance ratios of K in the BSE-light and BSE-dark matrix regions, K plots near unity. These patterns of depletion in the matrix of NWA 5343 differ from patterns in other anhydrous carbonaceous chondrites, as matrix material is typically enriched in all elements except Ca and Cr relative to the whole-rock composition (e.g., Wasson and Rubin 2009).

There are two possible explanations for the depletion of Na in the matrix of NWA 5343; the first of which being volatile loss during impact heating (Wlotzka 1993; Wasson et al. 2013). There is substantial evidence that the CK chondrites have been fragmented by impacts (e.g., crushed chondrules), so Na loss would be expected. An alternative, and more likely, explanation is that this depletion is due to the absence of Na-bearing phases in the matrix. In the CK chondrites, whole-rock compositions are influenced by contributions from the matrix, chondrules, and CAIs. Because plagioclase in CAIs is the main contributor of Na to the bulk composition, we would expect the matrix, which contains very low abundances of plagioclase, to be depleted in Na relative to the whole-rock composition. The matrix depletion of Al can also be attributed to the low abundance of plagioclase in the matrix. CAIs also contain Cr-bearing phases, such as high-Ca pyroxene and spinel (Chaumard et al. 2014). Thus, the depletion of Cr in the matrix is also likely an artifact of the contribution of CAIs to the whole-rock composition.
The primary Ni-bearing phase in the CK chondrites is olivine, with magnetite being a secondary contributor. Because both phases are present in the matrix, chondrules, and to a lesser extent CAIs, the depletion of Ni in the matrix of NWA 5343 cannot be an artifact of other components' contribution to the bulk rock composition. However, depletion of Ni is characteristic of CK chondrite finds from hot deserts and has been attributed to breakdown of pentlandite during terrestrial weathering (Rubin and Huber 2005; Huber et al. 2006). Abundances of Ni in the matrix of NWA 5343 are consistent with Ni abundances in weathered hot desert finds (Huber et al. 2006) and correspond to a weathering index (wi) of wi-3 (significantly weathered) (Rubin and Huber 2005). Although we must use caution when comparing Ni abundances in NWA 5343, which represent only the matrix contribution, to whole-rock abundances measured by Huber et al. (2006), a visual estimate of the proportion of silicate minerals with brown staining confirms a weathering index of wi-3.

**Terrestrial Weathering**

Depletions of Ni and the brown staining of silicates, which forms when oxidized iron is mobilized (Rubin and Huber 2005), confirm that NWA 5343 experienced terrestrial weathering. Because abundances of Ni are the same in the BSE-dark and BSE-light regions (0.35 wt%), it is likely that the breakdown of pentlandite was homogenous throughout the sample. The similarity between abundances of Fe and Mn in the BSE-dark and BSE-light matrix also support this, as both elements are largely unaffected by weathering in the CK chondrites (Huber et al. 2006). However, in addition to the characteristic weathering observed in CK chondrites, the BSE-light region of the matrix has also been affected by a calcium carbonate weathering product, which fills the pore space in this region. This carbonate weathering product is responsible for the Ca content of the BSE-light matrix, which is 2.2× higher than the BSE-dark matrix. Huber et al. (2006) also observed elevated Ca in hot desert finds, which they attributed to terrestrial weathering.

The source of the interstitial CaCO$_3$ material in the BSE-light matrix is a vein in region L14 (Fig. 5). This is not the first occurrence in carbonate veins in a CK chondrite, as Keller et al. (1992) also observed fine-grained carbonate veins in Maralinga, an anomalous CK4 chondrite found in the desert of Australia. Because Maralinga was extensively degraded and covered by patches of carbonate, Keller et al. (1992) attributed these veins to terrestrial weathering. Carbonate veins have been observed in CV chondrites Vigarano, Leoville, and NWA 1456 (Abreu and Brearley 2005; Kereszturi et al. 2015). Although Kereszturi et al. (2015) argued that veins in NWA 1456, which were filled with interlayered carbonate and Fe-Ni oxihydroxides, formed under varying oxidizing conditions on the parent body, Abreu and Brearley (2005) suggested that Fe-oxides associated with carbonate were produced during terrestrial weathering of metal and sulfides. Abreu and Brearley (2005) also argued that carbonate veins in Vigarano were terrestrial in origin, due in large part to their association with a carbonate layer on the surface of the Vigarano specimen. Additional support for a terrestrial origin of the carbonate in NWA 5343 is the large extent of carbonate material. The vein itself is 150–300 μm wide, and carbonate material infiltrates into ~170 cm$^2$ of the total sample area. This extent of infiltration would require a significant amount of fluid, which would be unlikely on the CK chondrite parent body, as much of the fluid present would be driven off during metamorphism.

The presence of a well-developed CaCO$_3$ vein in the matrix of NWA 5343 provides us with an opportunity to examine the potential effects of terrestrial weathering on silicate mineral compositions. Although diffusion of fluids into silicate minerals at surface temperature and pressures is unlikely, if diffusion did occur, we would expect silicate minerals in the BSE-light matrix to have higher CaO contents than their counterparts in the BSE-dark matrix. Although we did observe a higher mean CaO content of olivine in the BSE-light matrix than in the BSE-dark matrix (0.12 wt% versus 0.09 wt%), values for both are consistent with matrix olivine in other CK3 chondrite that we have analyzed (0.10–0.19 wt% CaO) (Dunn et al. 2016b). Therefore, the fluid does not appear to have influenced the composition of olivine. This also appears to be true for high-Ca pyroxene. Although the Wo content of high-Ca pyroxene in NWA 5343 (Wo$_{46.50}$) is higher than some CK4 chondrites (Wo$_{42.47}$) (Noguchi 1993; Ivanova et al. 2000), there is no discernible difference between the composition of high-Ca pyroxene in BSE-dark and BSE-light material (22.2–25.4 wt% CaO). Calcium content of plagioclase is also the same in both regions and is consistent with previously measured CK chondrites. The only mineralogical changes that can be attributed to terrestrial weathering are the precipitation of carbonate and (possibly) apatite from the Ca-bearing fluid.

**Thermal Metamorphism**

**Petrologic Subtype**

The most easily recognizable effects of thermal metamorphism are textural changes, such as recrystallization of matrix material (i.e., blurring of chondrules) (Dodd 1969; Huss et al. 1978, 1981).
During metamorphism, opaque matrix material recrystallizes, becoming more coarse-grained, less porous, and less friable and translucent (Dodd 1969; Huss et al. 1978, 1981). Although Huss et al. (1978, 1981) suggested that matrix material begins to recrystallize at petrologic subtype 3.6 conditions, recent transmission electron microscopy (TEM)-based studies suggest that recrystallization begins much earlier (e.g., Dobrică and Brearley 2011). Thus, it is difficult to estimate the lower limit of metamorphism in NWA 5343 based on the extent of matrix recrystallization. However, because recrystallization is mostly complete by petrologic subtype 3.8 or 3.9 conditions (Huss et al. 1978, 1981), the extent of recrystallization in NWA 5343 suggests that it is less than petrologic subtype 3.8.

The most pronounced change that occurs during thermal metamorphism is the chemical equilibration of silicate minerals due to diffusive exchange of cations (e.g., Fe$^{2+}$, Mg$^{2+}$, Cr$^{3+}$, Mn$^{2+}$, and Ca$^{2+}$) between chondrite silicate minerals due to diffusive exchange of cations (e.g., Dobrică and Brearley 2011). Because pyroxene equilibrates more slowly during metamorphism, the composition and homogeneity of olivine is the most commonly used indicator of metamorphic grade (Dodd and Van Schmus 1967; Huss et al. 1978, 1981). We can use the ordinary chondrites, which were metamorphosed on onion-shell asteroids (Treiloff et al. 2003), to help us quantify the extent of metamorphism in the CK chondrites. Because both groups have different precursor materials and reached equilibration at different compositions, we cannot directly compare olivine compositions. However, we can compare the heterogeneity of olivine, as we would expect olivine to equilibrate at roughly the same rate during thermal metamorphism. As illustrated in a histogram of matrix olivine compositions (Fig. 8), the Fa content of matrix olivine in NWA 5343 ranges from 30.5 to 39.8 with a mean at 36 mol%. If we compare this histogram to histograms of matrix olivine in higher grade ordinary chondrites (subtypes 3.6–3.8), it most closely resembles ordinary chondrites Mezo-Madras (L3.7) and Parnallee (LL3.6) in terms of distribution (shape) and compositional range (Huss et al. 1981; Matsunami et al. 1990). Comparison of matrix olivine in NWA 5343 to olivine in CO chondrites yields similar results, as matrix olivine in NWA 5343 most closely resembles matrix olivine in Warrenton (CO 3.6) (Brearley and Jones 1998). Although a detailed examination of chondrule olivine in the CK chondrites is beyond the scope of this study, the composition and homogeneity of chondrule olivine in NWA 5343 (Fa$_{15.9}^{3.6}$(L3.7)) is also consistent with CO chondrites of petrologic type 3.6 or higher (Scott and Jones 1990).

In addition to silicate minerals, the bulk composition of the matrix is also sensitive to the effects of metamorphism (Huss et al. 1981). The FeO/(FeO+MgO) ratio of the matrix decreases during metamorphism, approaching the FeO/(FeO + MgO) ratio of the bulk sample when equilibration is reached. Thus, the F/FM$_{\text{matrix}}$ to F/FM$_{\text{bulk}}$ ratio for an equilibrated chondrite is 1.0. The FeO/(FeO+MgO) ratio of NWA 5343 matrix is 0.578. This value falls between the CO chondrites ALHA 77003 (3.6) and Isna (3.8) (Scott and Jones 1990). Since we have not measured the bulk composition of NWA 5343, we cannot directly address how close to equilibrium the matrix is to the bulk. However, using the bulk FeO/(FeO+MgO) ratios of the CK4 finds Kobe and Karoonda (Oura et al. 2002; Greenwood et al. 2010) as a proxy for the bulk composition, the F/FM$_{\text{matrix}}$ to F/FM$_{\text{bulk}}$ ratio of NWA 5343 is 1.05–1.08. These values are closest to ordinary chondrites of petrologic subtypes 3.4–3.9, and are most consistent with Bremervörde (H3.9) (Huss et al. 1981). Overall, the homogeneity of the bulk matrix and silicate minerals suggests that NWA 5343 is petrologic subtype 3.6–3.7.

Potential Differences in Metamorphism Between the BSE-Dark and BSE-Light Matrix

Because grain size of silicate minerals increases during metamorphism, the observation that olivine grains appear to be larger and more uniform in size in the BSE-light matrix than in the BSE-dark matrix may suggest that the BSE-light matrix is more metamorphosed. Chaumard et al. (2009) observed that tabular olivine grains found at lower petrologic types became more globular at higher petrologic types. So, the more anhedral nature of grains in the BSE-light matrix may also indicate a higher degree of metamorphism. However, Chaumard et al. (2009) suggested this change occurred between petrologic types 3.8 and 3.9. Because NWA 5343 is no higher than petrologic type 3.7, it may be that this change is more gradational, beginning at lower petrologic types and becoming more evident at higher petrologic types. An alternative explanation may be that the apparent textural differences between the two regions are the result of transient heating during impacts rather than thermal metamorphism. As there is substantial evidence that the CK chondrites have been shocked and fragmented by impacts, such heating events would be expected.

If the BSE-light matrix is more metamorphosed than the BSE-dark matrix, we would also expect olivine and pyroxene in the BSE-light matrix to be more chemically homogeneous and Fe-rich than in the BSE-dark matrix. Although the average composition of olivine is the same in both the regions (Fa$_{36}$), olivine in the BSE-light matrix is more homogeneous, spanning a
smaller range in Fa content ($\text{Fa}_{35-40}$) than olivine in the BSE-dark matrix ($\text{Fa}_{30-38}$). The average composition of diopside is the same in the both the BSE-dark and BSE-light matrix ($\text{En}_{40}\text{Fs}_{11}\text{Wo}_{49}$). However, unlike olivine, the heterogeneity of high-Ca pyroxene is the same in both regions of matrix. This may not be of significance though, as pyroxene equilibrates slower than olivine. Heterogeneity of plagioclase, which is attributed to fractionation during shock rather than thermal metamorphism, cannot be used as an indicator of petrologic type. Thus, there is minor mineralogical evidence suggesting that the BSE-light matrix is more metamorphosed than the BSE-light matrix, though this would be difficult to confirm.

**Relationship Between Silicate Mineral Equilibration and Texture**

Though silicate minerals seem to be chemically equilibrated to the same extent throughout the sample, there are recognizable differences in grain size and shape. The relationship between grain size and petrologic type has already been established for the equilibrated CK chondrites (Kallemeyn et al. 1991), and both Chaumard et al. (2009) and Wasson et al. (2013) observed an increase in the average grain size from petrologic subtype 3.8 to type 4. Wasson et al. (2013) measured an average grain size of 11 µm in NWA 1559 (CK3) and 35 µm in NWA 5035 (CK4), while Chaumard et al. (2009) measured an average grain size of 16 µm in NWA 1559 and 65 µm in TNZ 057 (CK4). This may suggest that grain size and shape are still sensitive to metamorphism at petrologic subtypes where silicate mineral equilibration is nearly complete (e.g., $>3.7$). However, a more detailed examination of matrices in CK3 chondrites would be necessary in order to fully quantify the extent of this textural change during the first stages of metamorphism.

**Possible CV–CK Metamorphic Sequence**

If the CV and CK chondrites are derived from the same parent body, then the CV chondrites, which are less than petrologic subtype 3.6 (Guimon et al. 1995; Bonal et al. 2006), must be the precursors to the more metamorphosed CK chondrites. The CV chondrites are a highly complex group, with three subgroups (CV$_{\text{red}}$, CV$_{\text{OXA}}$, and CV$_{\text{OXB}}$) characterized by different oxidation states and types of secondary alteration. The Bali-type CVs (CV$_{\text{OXB}}$) are an unlikely precursor, as they have heavier oxygen isotope compositions than the CK chondrites (Greenwood et al. 2010) and often have shock-induced petrofabrics (Rubin 2012) that are absent from the CKs. Greenwood et al. (2010) suggested that the reduced CV chondrites are a good isotopic match for CK chondrite precursor material. If this is the case, two possible metamorphic sequences exist, i.e., (1) CV$_{\text{red}} \rightarrow$ CV$_{\text{OXA}} \rightarrow$ CK or (2) CV$_{\text{red}} \rightarrow$ CK. Abreu and Brearley (2011) argued that the CV$_{\text{OXA}}$ chondrites could not have formed from thermal processing of CV$_{\text{red}}$ material because of the complex, fluid-assisted growth required to form matrix olivine in the CV$_{\text{OXA}}$ chondrites. Thus, the most likely scenario would be that CV$_{\text{red}}$ chondrites are a direct precursor to the CK chondrites.

To examine CV$_{\text{red}}$ chondrites as a possible precursor material for the CKs, we compared the mean composition of the BSE-dark matrix in NWA 5343 to the average matrix composition in Vigarano (CV$_{\text{red}}$) (Hurt et al. 2012). We elected to use the BSE-dark matrix in this comparison because the bulk composition of the BSE-light reflects the contribution of Ca from the carbonate weathering product. Magnesium- and Vigarano-normalized abundance ratios of 10 elements in the matrix of NWA 5343 are plotted in Fig. 11. If the CV and CK chondrites are genetically related, then matrix material in both groups should have nearly identical starting compositions, and any differences between the two matrices must be attributable to secondary alteration process (i.e., impact heating, aqueous alteration, or thermal metamorphism). Compared to Vigarano, the matrix of NWA 5343 is significantly depleted in Fe, Mn, Na, Al, and S, and slightly less depleted in Cr, Si, and K (Fig. 11). The only element that is enriched in the matrix of NWA 5343 is Ca, which is 2.8× higher than matrix in Vigarano. Because we are using the BSE-dark in our comparison with Vigarano, the Ca enrichment in NWA 5343 cannot be attributed to CaCO$_3$ weathering material.

NWA 5343 is clearly more metamorphosed than Vigarano, as evidenced by the chemical composition, heterogeneity, and grain size of olivine in Vigarano (Abreu and Brearley 2011). This is confirmed by the abundance of Fe in the matrix of NWA 5343, which is depleted relative to Vigarano. We would expect the matrix of NWA 5343 to have a lower Fe content than Vigarano, as iron diffuses from the matrix into chondrules during metamorphism (e.g., Huss et al. 1981; Scott and Jones 1990). Calcium behaves in the opposite manner (diffusing from chondrules to matrix), so the enrichment of Ca in the matrix of NWA 5343 is likely a result of the destabilization of chondrule mesostasis during metamorphism. However, the magnitude of the enrichment seems to argue against metamorphism as the sole source. The depletion of S in the matrix of NWA 5343 could be an effect of thermal metamorphism or terrestrial weathering. Both processes have been shown to result in a decrease in sulfur
Because Vigarano is a fall, we would expect it to have higher S than a hot desert find such as NWA 5343. The depletion of Na in the matrix of NWA 5343 is probably due to impact heating and shock, which is indicated by the presence of fragmented matrix. It is not clear what secondary process could be responsible for the depletion of Mn and Al, but the remaining differences between the matrices of Vigarano and NWA 5343 can be explained by secondary effects, suggesting that it is at least possible that NWA 5343 and Vigarano are genetically linked.

Although it is certainly possible that the matrix composition of NWA 5343 could be derived by secondary processing of a Vigarano-like precursor, this alone does not provide conclusive evidence for a genetic relationship between the CVred and CK chondrites. In addition to the arguments outlined by Dunn et al. (2016a) and Yin et al. (2017), porosity and texture of matrix olivine are also hard to reconcile with a continuous metamorphic sequence. Because porosity decreases during metamorphism, a continuous metamorphic sequence would imply that the CK chondrites should have lower porosities than the CVred chondrites. Actual porosity measurements of the CVred and CK chondrites are contrary to this, as porosity of the CVred chondrites is much lower than porosity of the CK chondrites (average of 3.6% and 17.8%, respectively) (Macke et al. 2011). However, the low porosity of the CVred chondrites could be a result of shock flattening. In addition, matrix olivine in the CVred chondrites, Allende and ALH 84028 (CV3), exhibits a “dusty” texture similar to matrix olivine in the CK chondrites; however, this texture is absent from CVred chondrite, Vigarano (Brearley 2009; Abreu and Brearley 2011). Abreu and Brearley (2011) argued that the absence of this texture required a different formation mechanism for olivine in the CVred chondrites, thus invalidating the argument that CVOX chondrites could be derived from thermal processing of the CVred chondrites. Because matrix olivine in the CK chondrites appears to have formed via the same process as olivine in the CVOX chondrites, this argument then applies to the CK chondrites as well.

**SUMMARY**

Based on the chemical heterogeneity of chondrule and matrix olivine, NWA 5343 is the least metamorphosed CK chondrite reported so far. In an effort to better constrain the lower limit of metamorphism in the CK chondrites, we performed a detailed analysis of matrix material in NWA 5343, including characterization of the texture and bulk composition and analyses of individual silicate minerals. To evaluate the possibility of a metamorphic sequence within the CV and CK chondrites, we also compared the bulk matrix composition of NWA 5343 to that of the reduced CV chondrite Vigarano. Our study of matrix in NWA 5343 yields the following conclusions.

1. Compared to bulk compositions of CK4 chondrites, the matrix of NWA 5343 is depleted in Al, Cr, Ni, and Na. The depletion of Na, Al, and Cr is likely an artifact of the contribution of CAIs to the bulk composition, while the depletion of Ni is the result of terrestrial weathering. The enrichment of Ca in the BSE-light matrix is due to the presence of interstitial carbonate.

2. The CaCO3 material filling the pore space in the BSE-light matrix is a product of terrestrial weathering. Infiltration of a Ca-bearing, terrestrial fluid does not appear to have directly influenced the composition of silicate minerals in the matrix. The only mineralogical changes that can be attributed to terrestrial weathering are the precipitation of carbonate and (possibly) apatite from the Ca-bearing fluid.

3. The extent of matrix recrystallization, heterogeneity of matrix olivine, and bulk composition of the matrix suggest that NWA 5343 is petrologic subtype 3.6 or 3.7.

4. Though silicate minerals seem to be equilibrated to roughly the same extent throughout the sample, there are recognizable differences in grain size and shape. These textural differences may be the result of transient heating events during impacts, which
would be likely on the CK chondrite parent body. The difference between compositional equilibration and texture may also suggest that grain size and shape is still sensitive to metamorphism at petrologic subtypes where silicate mineral equilibration is nearly complete (e.g., >3.7).

5. Although the matrix composition of NWA 5343 could be derived by secondary processing of a Vigarano-like precursor, porosity and texture of matrix olivine are hard to reconcile with a continuous metamorphic sequence. Porosity is lower in the CVred chondrites than in the CK chondrites, and matrix olivine in the CKs appears to require a different formation mechanism than matrix olivine in the CVred chondrites.

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REFERENCES


**SUPPORTING INFORMATION**

Additional supporting information may be found in the online version of this article:

**Fig S1.** BSE images of the five regions examined in this study. Areas that were analyzed for bulk composition are indicated on each area by 150 × 150 μm boxes. Areas K5, S6, and U12 are porous matrix, while I21 and L14 are weathered matrix. Scale bar is 500 μm.

**Fig S2.** Average bulk compositions of each of the grid areas.