

## Cosmogenic neon in grains separated from individual chondrules: Evidence of precompaction exposure in chondrules

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**Abstract**—Neon was measured in 39 individual olivine (or olivine-rich) grains separated from individual chondrules from Dhajala, Bjurböle, Chainpur, Murchison, and Parsa chondrites with spallation-produced <sup>21</sup>Ne the result of interaction of energetic particle irradiation. The apparent <sup>21</sup>Ne cosmic ray exposure (CRE) ages of most grains are similar to those of the matrix with the exception of three grains from Dhajala and single grains from Bjurböle and Chainpur, which show excesses, reflecting exposure to energetic particles prior to final compaction of the object. Among these five grains, one from chondrule BJ2A5 of Bjurböle shows an apparent excess exposure age of approximately 20 Ma and the other four from Dhajala and Chainpur have apparent excesses, described as an “age,” from 2 to 17 Ma. The precompaction irradiation effects of grains from chondrules do not appear to be different from the effects seen in olivine grains extracted from the matrix of CM chondrites. As was the case for the matrix grains, there appears to be insufficient time for this precompaction irradiation by the contemporary particle sources. The apparent variations within single chondrules appear to constrain precompaction irradiation effects to irradiation by lower energy solar particles, rather than galactic cosmic rays, supporting the conclusion derived from the precompaction irradiation effects in CM matrix grains, but for totally different reasons. This observation is consistent with Chandra X-Ray Observatory data for young low-mass stars, which suggest that our own Sun may have been 10<sup>5</sup> times more active in an early naked T-Tauri phase (Feigelson et al. 2002).

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### INTRODUCTION

Energetic particles currently bombarding our solar system consist of two main components: solar cosmic rays (SCR) and galactic cosmic rays (GCR). The Sun emits a steady stream of low-energy (KeV/amu) solar wind ions with sporadic SCR flares of higher energy (MeV/amu), whereas the extra-solar GCR component consists of much higher energy ions (GeV/amu). With energy difference comes a difference in range. The range of SCR ions in solids is less than a centimeter but GCR ions, with their fully developed secondary cascades, can penetrate meters of the material. These energetic ions, mostly protons, energetic secondary neutrons, produce

cosmogenic nuclides (e.g., <sup>3</sup>He, <sup>10</sup>Be, <sup>14</sup>C, <sup>21</sup>Ne, <sup>38</sup>Ar, <sup>53</sup>Mn, etc.) by spallation reactions. Some individual olivine grains from the matrix of CM chondrites (Murchison, Murray, Cold Bokkeveld), H chondrites (Weston and Fayetteville), and grain size separates from the howardite Kapoeta contain significantly more cosmogenic Ne than could have been produced during their conventional cosmic ray exposure ages (the time they were exposed as meter-sized objects). This led to the conclusion that either these grains were individually irradiated for much longer times than given by their exposure ages, or there was a much higher energetic particle flux in the early solar system when they were irradiated on the surfaces of their parent bodies (Caffee

et al. 1987; Wieler et al. 1989, 2000; Hohenberg et al. 1990; Rao et al. 1997; also see review by Woolum and Hohenberg 1993).

For some of these objects, the “gas-rich” chondrites like Kapoeta, extended regolith durations are reflected by their large solar gas contents, so both alternatives may be possible in these cases. However, for others, like the CM chondrites, there is not sufficient time for the observed concentrations of spallation Ne to be due to irradiation in the parent body regolith by the contemporary cosmic ray suite because some of these grains would require such exposures to be improbably long (Caffee et al. 1988; Hohenberg et al. 1990; Woolum and Hohenberg 1993). Those authors reported that track-rich olivine grains (meaning they were exposed directly on the surface to solar flares) from all of these meteorites seem to contain excess spallation Ne. For Murchison 17 of 41, and for Murray 8 of 10, of the olivine grains with solar flare tracks have large apparent precompaction ages of more than 20 times their conventional cosmic ray exposure ages, ages that are at least 20 Ma (up to >145 Ma, the highest apparent precompaction exposure duration observed to date). These effects are totally absent for grains devoid of solar flare tracks whose individual exposure ages are all consistent with the conventional cosmic ray exposure age of their parent object. These very high concentrations of precompaction cosmogenic Ne must be due to either long exposures to the conventional particle suite or enhanced particles sources in the early solar system. The CM chondrite observations strongly suggested the latter (e.g., Caffee et al. 1988; Hohenberg et al. 1990; Woolum and Hohenberg 1993). Those results led us to undertake this study of other meteorite components, in particular, grains within individual chondrules.

Chondrules, the major constituent of chondrites, may provide similar records of precompaction exposures since their formation clearly predates incorporation into the parent meteorite. Have these experienced the same precompaction effects as the matrix grains, or are they different? Some studies on chondrules do indicate excess of cosmogenic noble gases that may be attributed to precompaction exposure, but statistics can play an important role here. The individual matrix grain studies allowed preselection of grains that contained solar flare tracks by etching to make the tracks visible, thus grains that were exposed on or near the regolith surface. These accounted for only approximately 1 in at least 20 olivine grains from the Murchison matrix (Hohenberg et al. 1990) and those with the larger precompaction effects are even rarer. Without this preselection, the statistics are much less favorable for finding precompaction exposures. If the grain studies of the CM chondrites are used as a guide, and if the statistics are the same for chondrules as for olivine

grains, only about 1 in 20 randomly selected chondrules should show evidence of precompaction exposure. Records of precompaction exposures of chondrules do exist, some with and some without the aid of solar flare tracks. Caffee et al. (1987) find that chondrules with solar flare heavy ion tracks from the H chondrite Weston show apparent precompaction exposures of up to 19 Ma. Polnau et al. (1999) report a precompaction exposure of approximately 1 Ma in a randomly chosen chondrule from the H6 chondrite ALH 76008, based on the cosmogenic noble gases  $^3\text{He}$ ,  $^{21}\text{Ne}$ , and  $^{38}\text{Ar}$ . A similar study on a bulk collection of chondrules and matrix samples of eight ordinary chondrites, including Bjurböle, also reports evidence of some precompaction exposure for chondrules (in the range of 0.1–3 Ma, Polnau et al. 2001), an effect clearly diluted by the bulk nature of the measurement. More recently, similar results for at least three chondrules from Dhajala and Bjurböle are reported (Eugster et al. 2007; Das and Murty 2009). Higher apparent CRE ages (up to 22 Ma) have been reported for six (out of 35) individual Murchison chondrules (Roth et al. 2011). Beyersdorf-Kuis et al. (2012) report slightly higher apparent CRE ages for El Djouf 001 and Vigarano chondrules relative to their respective matrix materials. Huber et al. (2012) found that Murchison chondrules embedded in a clastic matrix have CRE ages up to 25 Ma, while the one chondrule from a primary rock fragment has a CRE age similar to that of the matrix (approximately 1 Ma).

Detailed studies of  $^{207}\text{Pb}$ – $^{206}\text{Pb}$  ages (Amelin et al. 2002; Amelin and Krot 2007), and the constraints derived from the extinct radionuclides  $^{26}\text{Al}$ ,  $^{53}\text{Mn}$ , and  $^{129}\text{I}$ – $^{129}\text{Xe}$  ages (Rudraswami et al. 2008; Villeneuve et al. 2009; Nyquist et al. 2009; see reviews by Hohenberg and Pravdivtseva 2008; Krot et al. 2009), have demonstrated that primary chondrule formation is constrained to 2–3 Ma, immediately after the formation of CAIs. While these studies, by themselves, do not constrain the duration of precompaction irradiation, cosmogenic noble gas records of it could not predate chondrule formation, nor could it effectively extend beyond meteorite formation (compaction into the final object). Thus, it must have occurred quite early, either in space or in the parent body regolith, and limits on the time available for such irradiations must come from constraints on the compaction time of the host object. What is unknown is how much gas remained in the nebula at that time. Cuzzi and Alexander (2006) argue for the existence of considerable nebular gas, which can effectively shield constituents from energetic particles. However, a proto-star is quite active magnetically and with particle emissions, and an ionized solar nebula couples tightly to the solar magnetic field spiraling outward, rapidly clearing the inner solar system of gas (evidenced by the

gas giants further out). Contraction of low-mass stars onto the main sequence is characterized by an early T-Tauri phase, and then a later “naked” T-Tauri phase in which much of the nebular gas has been cleared from the inner solar system, with energetic particle emission still elevated and potentially able to account for early precompaction irradiation effects (Feigelson et al. 2002).

In this work, we report the concentration of cosmogenic (spallation-produced)  $^{21}\text{Ne}$  in chemically characterized grains extracted from chondrules separated from the Dhajala, Bjurböle, Chainpur, Parsa, and Murchison chondrites (Das et al. 2009, 2010) and identify the contributions from precompaction exposures of these grains to energetic particle irradiation.

## SAMPLES AND EXPERIMENTAL METHODS

### Samples and Their Chemical Characterization

The samples of Murchison (CM2), Dhajala (H3.8), Chainpur (LL3.4), Bjurböle (L/LL4), and Parsa (EH3) meteorites were selected from the meteorite collection at the Physical Research Laboratory, India. The starting source material used to extract all samples for this investigation was small, at most a few grams, therefore the shielding conditions for cosmic ray exposure were uniform within each sample. Accordingly, for the same target chemistry, differences between cosmogenic  $^{21}\text{Ne}$  concentrations will reflect differences in cosmic ray exposure. The samples were gently crushed and disaggregation was completed by high-powered ultrasonic agitation. Chondrules exceeding approximately 1 mm in diameter were handpicked using an optical microscope. The individual separated chondrules were then carefully broken. Fragments that were mostly olivine were selected, visually identified by optical microscopy, and those fragments were mounted on an electron microscope stub using double-sided carbon tape. These grains, chiefly olivine, have some adhering mesostasis, which cannot be removed so EDX spectra were obtained for each fragment using the JEOL 540 SEM, with its Tracor Northern 5400 X-ray system, at Washington University, to confirm the mineralogy and to determine the specific spallation target chemistry for each fragment. To obtain a representative elemental composition of each grain, the EDX spectra were measured on the largest face. The chondrule grains were then removed from the stub and weighed using a Mettler Toledo XP2U microbalance. The elemental compositions of individual grains are listed in Table 1 where it can be seen that olivine compositions dominate so we will refer to these olivine rich fragments as “olivine grains.” The chondrule grains were then loaded in 2 mm deep blind holes in an aluminum laser extraction stub for analysis in a high

transmission ion-counting mass spectrometer, similar to the one described by Hohenberg (1980). Unfortunately, due to the manner in which the chondrules were crushed to extract the grains, location information of each grain within the chondrule could not be preserved.

### Ne Analyses

The laser extraction cell was degassed at 150 °C for 2 days to minimize noble gas blank, typically  $8 \times 10^{-15} \text{ cm}^3\text{STP } ^{21}\text{Ne}$ . The noble gases were extracted using a CW Nd-YAG laser where the grains were completely melted, forming spherical droplets under coaxial microscope observation. The released gases were sequentially exposed to two SAES Ti-Zr getters held at 275 °C and then exposed to a freshly deposited Ti-film for final cleaning leaving only the noble gases. He and Ne were separated by freezing Ar, Kr, and Xe on activated charcoal at -178 °C (LN temperature). To minimize  $^{40}\text{Ar}^{++}$  interference for mass 20 from any remaining or liberated Ar, the mass-spectrometer remained connected to a second activated charcoal finger kept at -178 °C. Details of this micro-analytical laser extraction technique are described by Nichols et al. (1995).

### Blanks, Interference Corrections, and Calibrations

Gases admitted into the mass spectrometer still contain traces of contaminant species and memory gases are generated during analyses. The production of  $^{40}\text{Ar}^{++}$  and  $^{44}\text{CO}_2^{++}$  in the source region of the mass spectrometer interferes with the neon isotopes at  $m/e = 20$  and 22. An SAES NP10 getter is installed in the mass spectrometer volume to minimize interferences from various species during analyses. Correction factors are determined from Ne-free blanks, and were measured to be:  $\text{HF}^+/\text{F}^+ = 0.0045 \pm 0.0005$ ,  $\text{H}_2^{18}\text{O}^+/\text{H}_2\text{O}^+ = 0.0022 \pm 0.0001$ ,  $40^{++}/40^+ = 0.0360 \pm 0.0004$ ,  $\text{CO}_2^{++}/\text{CO}^+ = 0.0020 \pm 0.0004$ , and  $\text{H}_2^+/\text{HD}^+ = 0.00085 \pm 0.00003$ , but are not significant for this work.

An automated pneumatically controlled gas pipette containing  $8.56 \times 10^{-10} \text{ cm}^3\text{STP}$  of atmospheric neon was used to determine Ne sensitivity and instrumental mass discrimination. Fractionation was avoided during the filling of calibration volumes with an atmospheric sample by using a capillary inlet system, which assures viscous (rather than molecular) flow conditions. This pipette was calibrated using the LP-6 biotite standard (Charbit et al. 1998), which contains  $(11.580 \pm 0.006) \times 10^{14}$  atoms of  $^{40}\text{Ar}$  per gram, and remains homogeneous at the mg-level and calibrations were independently verified (to 1.5%) by inter-laboratory pipette exchanges with ETH, Zurich. Pipette standards were typically run

Table 1. Target chemistry relevant for the production of neon in individual chondrule grains.

Sample	Mg	Al	Si	Fe	Na	S	K	Ca	Cr	Mn
<b>Dhajala (H3.8)</b>										
Chondrule DH2										
DH2A1	18.6	n.d.	12.1	2.8	n.d.	n.d.	0.04	n.d.	n.d.	0.2
Chondrule DH3										
DH3A11	13.7	0.1	9.3	2.0	0.1	n.d.	n.d.	0.09	n.d.	0.1
DH3A13	16.9	0.9	15.7	3.3	3.1	0.3	0.2	1.1	1.3	-
DH3A22	16.5	0.2	12.1	2.5	0.6	0.1	0.1	0.2	0.1	0.2
Matrix <sup>a</sup>										
Dhajala	21.1	1.06	11.0	41.1	0.54	2.0	0.11	1.26	0.37	0.41
<b>Bjurböle (L/LL4)</b>										
Chondrule BJ0										
BJ01	11.7	0.9	10.6	2.3	1.1	n.d.	0.1	0.3	n.d.	0.1
BJ06	11.7	1.4	11.0	10.3	2.7	n.d.	0.1	0.9	0.2	0.5
Chondrule BJ1										
BJ1A1	13.1	3.4	25.5	2.3	7.7	0.1	0.2	1.3	0.4	0.2
BJ1A2	16.4	2.5	30.1	3.7	5.3	0.1	0.2	0.7	0.6	0.3
BJ1B1	18.6	1.2	30.8	4.9	1.6	0.1	0.1	0.6	0.5	0.5
BJ1B2	15.8	2.7	30.2	3.6	7.1	0.3	0.2	1.6	0.5	0.3
BJ1B3	14.6	3.4	28.6	3.0	7.8	0.4	0.2	1.2	0.7	0.3
Chondrule BJ2										
BJ2A1	15.9	2.8	30.2	3.6	7.2	0.2	0.2	1.5	0.4	0.2
BJ2A2	16.5	n.d.	23.1	4.1	0.1	n.d.	n.d.	0.2	0.3	0.3
BJ2A5	13.0	0.4	16.6	3.1	0.4	-	n.d.	0.3	n.d.	0.1
BJ2B8	16.6	0.1	16.7	4.5	0.5	0.1	n.d.	0.3	0.2	0.3
Matrix <sup>b</sup>										
Bjurböle	17.8	1.25	18.9	18.6	n.d.	2.2	0.08	1.05	0.39	0.31
<b>Chainpur (LL3.4)</b>										
Chondrule CH2										
CH2A2	18.7	0.1	13.5	3.7	0.3	n.d.	n.d.	-	0.1	0.2
Chondrule CH3										
CH3A1	14.6	-	17.7	2.7	n.d.	n.d.	n.d.	0.6	0.2	n.d.
CH3A2	11.1	0.5	13.2	1.7	n.d.	1.1	n.d.	0.7	0.1	n.d.
CH3A3	13.0	1.2	15.3	2.0	n.d.	n.d.	n.d.	0.5	0.1	n.d.
CH3A4	12.4	0.6	14.9	1.7	n.d.	n.d.	n.d.	0.7	0.1	n.d.
CH3A6	12.6	0.1	14.4	2.3	n.d.	2.7	n.d.	0.4	0.1	n.d.
CH3A7	10.7	1.3	10.1	2.4	n.d.	2.6	n.d.	0.4	0.1	n.d.
Chondrule CH5										
CH5A2	14.1	3.3	19.1	1.2	3.2	1.6	0.2	1.3	0.3	0.2
CH5A3	16.1	0.2	9.5	0.7	-	2.4	n.d.	0.1	n.d.	0.1
Chondrule CH7										
CH7A1	24.1	1.4	23.0	5.3	4.5	0.2	0.0	0.6	0.1	0.2
CH7A2	16.2	1.5	17.0	4.5	8.9	0.0	0.1	0.6	0.3	0.2
CH7A7	19.7	1.6	25.3	5.6	6.7	n.d.	1.7	2.5	0.3	0.3
Matrix <sup>c</sup>										
Chainpur (100–200 µm)	13.1	0.7	16.2	30	n.d.	n.d.	n.d.	0.6	n.d.	n.d.
<b>Murchison (CM2)</b>										
Chondrule MURSPH										
MURSPH	11.1	0.1	11.2	0.9	n.d.	4.5	n.d.	0.2	n.d.	n.d.
Chondrule CH1										
MURCH1	13.0	0.9	11.1	0.4	1.1	3.9	0.1	0.5	0.2	n.d.
Chondrule CH2										
MURCH2	18.3	0.2	10.2	0.5	n.d.	n.d.	n.d.	0.1	0.1	n.d.
Matrix grains										
MUROL1	26.0	13.4	11.4	1.3	n.d.	1.2	n.d.	0.2	0.4	0.9

Table 1. *Continued.* Target chemistry relevant for the production of neon in individual chondrule grains.

Sample	Mg	Al	Si	Fe	Na	S	K	Ca	Cr	Mn
MUROL2	15.2	n.d.	11.6	3.5	n.d.	3.7	n.d.	0.1	0.1	0.1
MUROL4	12.7	n.d.	8.6	2.3	n.d.	5.5	n.d.	0.1	n.d.	n.d.
<b>Parsa (EH3)</b>										
Chondrule PR1										
PR1A12	22.9	6.3	45.9	1.0	7.2	n.d.	0.1	2.5	0.4	0.2
PR1A3	13.4	8.9	35.3	4.4	12.1	n.d.	0.1	3.1	0.9	0.1
PR1A4	26.6	3.3	37.6	5.1	11.3	n.d.	0.3	1.1	0.5	0.6
PR1A5	18.7	1.7	28.1	0.7	3.6	n.d.	-	0.5	0.3	n.d.
Bulk <sup>c</sup>										
Bulk	10.8	0.78	16.7	2.77	n.d.	n.d.	n.d.	0.8	n.d.	n.d.

<sup>a</sup>Eugster et al. (2007); <sup>b</sup>Polnau et al. (2001); <sup>c</sup>Das and Murty (2009). Uncertainties are determined by averaging the  $1\sigma$  errors of three measurements for each grain and are propagated into the cosmogenic production rates for  $^{21}\text{Ne}$ . Mg and Si are the primary targets for  $^{21}\text{Ne}$  production and n.d. = not determined so, for grains DH3A21 and CH5A1, the chemical compositions of DH3A13 and CH5A2 are respectively used to calculate  $^{21}\text{Ne}$  production rates.

before and after each set of sample analyses and, over the course of these measurements, Ne sensitivities remained constant within 5% and instrumental isotopic mass discrimination remained linear at  $3.0 \pm 0.1\%/amu$ . Procedural blanks were run before, during, and after each set of sample analyses. The  $^{21}\text{Ne}$  blank is particularly low and free of any interference. Typically  $8 \times 10^{-15} \text{ cm}^3\text{STP } ^{21}\text{Ne}$  is observed, sufficiently low for the measurement of small quantities of spallation-produced Ne in each sample.

## RESULTS

Our mass spectrometer does not have sufficient resolution to resolve HD from  $^3\text{He}$ , and there is a significant H background, due to hot hydrogen cleaning of hydrocarbons when this instrument was constructed, so precise  $^3\text{He}$  measurements were only possible for Chainpur with its higher CRE age. However, it has a very low background, and there are no isobaric interferences at mass 21, so the  $^{21}\text{Ne}$  measurement in these samples is both precise and free of any significant corrections. It is also dominated by spallation in these samples so the quantity of spallation-produced  $^{21}\text{Ne}$  is essentially independent of the other Ne isotopes, which may have significant corrections, and any instrumental effects or interferences. The Ne isotopic ratios, relative to  $^{21}\text{Ne}$ , and the concentrations of spallation-produced  $^{21}\text{Ne}$ , are shown in Table 2. Corrections are negligibly small for  $^{21}\text{Ne}$ , a few percent for  $^{20}\text{Ne}$ , and up to 20 percent for  $^{22}\text{Ne}$ , and essentially all uncertainties in the ratios are due to uncertainties in  $^{20}\text{Ne}$  and  $^{22}\text{Ne}$ . The sample with the largest trapped Ne component is BJ2A1 from Bjurböle, but even here, the contribution of trapped to the measured  $^{21}\text{Ne}$  in BJ2A1 is still less than 1%. Therefore, any uncertainties at masses 20 and 22 due to interference corrections have totally negligible effects on

the cosmogenic  $^{21}\text{Ne}$  in these chondrule grains, which ranges from 0.7 to  $15 \times 10^{-8} \text{ cm}^3\text{STPg}^{-1}$  (Table 2).

### Production Rates for Cosmogenic $^{21}\text{Ne}$ and the Apparent CRE Ages

The most sensitive shielding indicator for meteorites is the cosmogenic  $^{22}\text{Ne}/^{21}\text{Ne}$  ratio, which ranges from 1.05 to 1.25 (Wieler 2002). For these measurements, however, the cosmogenic  $^{21}\text{Ne}$  is much more precise than the inferred cosmogenic  $^{22}\text{Ne}$ , and the corresponding cosmogenic  $^{22}\text{Ne}/^{21}\text{Ne}$  ratios not well enough defined to constrain the shielding depth. However, if we assume *maximum* production rate, which occurs for a cosmogenic  $^{22}\text{Ne}/^{21}\text{Ne}$  ratio of about 1.08, (which is consistent with our data within its precision), the inferred apparent cosmic ray exposure ages will be *minimum* values. Since the target chemistry of each grain varies from the bulk sample, we must calculate the specific  $^{21}\text{Ne}$  production rate for each grain (Eugster and Michel 1995) to obtain the minimum apparent exposure age. Table 2 presents the  $^{21}\text{Ne}$  production rates ( $P_{21}$ ) and the equivalent apparent minimum CRE ages ( $T_{21}$ ) for each of the chondrule grains.

### Comparison of Apparent CRE Ages of Chondrules Grains With the Respective Matrix

If grains extracted from chondrules have higher apparent CRE ages than their corresponding matrix, they must have been exposed to energetic particles before formation of the object (precompaction exposure). The CRE ages of the matrices of these meteorites are well known, but they can also be calculated in this work using their bulk compositions and their cosmogenic  $^{21}\text{Ne}$  abundances (Polnau et al. 2001; Eugster et al. 2007; Das and Murty 2009). This calculation may be better suited

Table 2. Mass, cosmogenic  $^{21}\text{Ne}$ , production rates of  $^{21}\text{Ne}$  ( $P_{21}$ ), apparent  $^{21}\text{Ne}$  exposure ages ( $T_{21}$ ), and apparent precompaction times. Spallation-produced  $^{21}\text{Ne}$  concentrations are in  $10^{-8}\text{cm}^3\text{STPg}^{-1}$ . Production rates are in  $10^{-8}\text{cm}^3\text{STPg}^{-1}\text{Ma}^{-1}$ .

Sample	Mass ( $\mu\text{g}$ )	$^{21}\text{Ne}_c^a$	$^{20}\text{Ne}/^{21}\text{Ne}$	$^{22}\text{Ne}/^{21}\text{Ne}$	$P_{21}$	$T_{21}(\text{Ma})$	$T_{\text{pre}}^b(\text{Ma})$
<b>Dhajala (H3.8)</b>							
Chondrule DH2							
DH2A1	16.5	$2.74 \pm 0.11$	$1.27 \pm 0.33$	$0.97 \pm 0.24$	0.405	$6.76 \pm 1.05$	$3.08 \pm 1.20$
Chondrule DH3							
DH3A11	7.7	$2.40 \pm 0.14$	$0.97 \pm 0.91$	$0.80 \pm 0.51$	0.301	$7.98 \pm 1.29$	$4.30 \pm 1.41$
DH3A13	9.0	$2.18 \pm 0.12$	$1.34 \pm 0.76$	$0.84 \pm 0.51$	0.395	$5.52 \pm 0.88$	$1.84 \pm 1.05$
DH3A21*	2.9	$3.83 \pm 0.26$	$1.28 \pm 1.42$	$0.74 \pm 0.86$	0.395	$9.70 \pm 1.59$	$6.02 \pm 1.69$
DH3A22	5.2	$2.63 \pm 0.21$	$0.53 \pm 1.30$	$0.91 \pm 0.66$	0.367	$7.17 \pm 1.22$	$3.49 \pm 1.35$
Matrix <sup>c</sup>							
Matrix	20800	$1.74 \pm 0.09$	$1.00 \pm 0.01$	$1.12 \pm 0.01$	0.473	$3.68 \pm 0.58$	–
<b>Bjurböle (L/LL4)</b>							
Chondrule BJ0							
BJ01	3.7	$4.60 \pm 0.20$	$1.11 \pm 1.02$	$1.02 \pm 2.69$	0.273	$16.85 \pm 2.63$	$7.64 \pm 2.99$
BJ06	1.4	$6.08 \pm 0.53$	$2.82 \pm 2.12$	$0.73 \pm 5.28$	0.280	$21.75 \pm 3.77$	$12.54 \pm 4.03$
Chondrule BJ1							
BJ1A1	10.1	$4.03 \pm 0.15$	$0.96 \pm 0.37$	$0.97 \pm 1.21$	0.375	$10.76 \pm 1.66$	$1.55 \pm 2.18$
BJ1A2	12.3	$4.27 \pm 0.18$	$1.24 \pm 0.29$	$0.86 \pm 0.94$	0.449	$9.51 \pm 1.48$	$0.30 \pm 2.05$
BJ1B1	13.5	$5.10 \pm 0.19$	$0.91 \pm 0.22$	$0.99 \pm 0.73$	0.487	$10.47 \pm 1.62$	$1.26 \pm 2.15$
BJ1B2	14.6	$4.41 \pm 0.23$	$0.77 \pm 0.25$	$1.03 \pm 0.77$	0.442	$9.97 \pm 1.58$	$0.76 \pm 2.12$
BJ1B3	24.9	$5.01 \pm 0.18$	$0.93 \pm 0.12$	$1.09 \pm 0.40$	0.416	$12.03 \pm 1.86$	$2.82 \pm 2.34$
Chondrule BJ2							
BJ2A1	12.8	$3.93 \pm 0.17$	$3.76 \pm 0.36$	$1.06 \pm 0.91$	0.443	$8.87 \pm 1.39$	$-0.34 \pm 1.99$
BJ2A2	3.1	$3.10 \pm 0.12$	$2.34 \pm 1.88$	$1.77 \pm 4.79$	0.406	$7.64 \pm 1.18$	$-1.54 \pm 1.85$
BJ2A5	6.5	$9.34 \pm 0.40$	$0.94 \pm 0.28$	$0.95 \pm 0.76$	0.316	$29.56 \pm 4.60$	$20.35 \pm 4.81$
BJ2B8	7.1	$4.95 \pm 0.21$	$2.04 \pm 0.50$	$0.93 \pm 1.31$	0.386	$12.84 \pm 2.00$	$3.63 \pm 2.45$
Matrix <sup>d</sup>							
Matrix	30000	$4.01 \pm 0.13$	$0.81 \pm 0.01$	$1.07 \pm 0.02$	0.435	$9.21 \pm 1.42$	–
<b>Chainpur (LL3.4)</b>							
Chondrule CH2							
CH2A2	3.3	$9.98 \pm 0.56$	$0.58 \pm 0.50$	$0.90 \pm 1.50$	0.414	$24.11 \pm 3.86$	$-1.64 \pm 6.05$
Chondrule CH3							
CH3A1	26.0	$9.68 \pm 0.27$	$1.00 \pm 0.06$	$1.09 \pm 0.20$	0.351	$27.60 \pm 4.21$	$1.85 \pm 6.28$
CH3A2	11.1	$8.45 \pm 0.30$	$0.98 \pm 0.16$	$1.01 \pm 0.53$	0.269	$31.43 \pm 4.84$	$5.68 \pm 6.72$
CH3A3	9.4	$8.86 \pm 0.32$	$1.20 \pm 0.18$	$1.12 \pm 0.60$	0.318	$27.87 \pm 4.30$	$2.12 \pm 6.34$
CH3A4	16.2	$9.52 \pm 0.25$	$1.00 \pm 0.10$	$1.11 \pm 0.33$	0.301	$31.63 \pm 4.82$	$5.88 \pm 6.70$
CH3A6	8.4	$9.00 \pm 0.31$	$1.06 \pm 0.20$	$1.11 \pm 0.66$	0.300	$30.04 \pm 4.62$	$4.29 \pm 6.56$
CH3A7	9.8	$9.48 \pm 0.42$	$0.97 \pm 0.17$	$1.11 \pm 0.54$	0.255	$37.12 \pm 5.80$	$11.37 \pm 7.44$
Chondrule CH5							
CH5A1 <sup>#</sup>	12.2	$12.34 \pm 0.39$	$1.05 \pm 0.11$	$1.07 \pm 0.31$	0.370	$33.29 \pm 5.11$	$7.54 \pm 6.92$
CH5A2	19.7	$12.70 \pm 0.30$	$0.98 \pm 0.07$	$1.07 \pm 0.19$	0.370	$34.32 \pm 5.21$	$8.57 \pm 6.99$
CH5A3	2.1	$15.00 \pm 0.88$	$1.42 \pm 0.56$	$0.91 \pm 1.45$	0.347	$43.14 \pm 6.95$	$17.39 \pm 8.37$
Chondrule CH7							
CH7A1	18.7	$11.97 \pm 0.30$	$1.26 \pm 0.08$	$1.06 \pm 0.21$	0.564	$21.23 \pm 3.23$	$-4.52 \pm 5.67$
CH7A2	3.6	$11.10 \pm 0.51$	$1.11 \pm 0.44$	$0.95 \pm 1.15$	0.389	$28.53 \pm 4.48$	$2.78 \pm 6.46$
CH7A7	6.3	$8.69 \pm 0.35$	$1.31 \pm 0.31$	$1.11 \pm 0.84$	0.492	$17.66 \pm 2.74$	$-8.09 \pm 5.41$
Matrix <sup>e</sup>							
Matrix (100–200 $\mu\text{m}$ )	18450	$8.42 \pm 0.85$	$2.38 \pm 0.12$	$1.26 \pm 0.06$	0.327	$25.75 \pm 4.66$	–
<b>Murchison (CM2)</b>							
MURSPH							
MURSPH (chondrule)	55.7	$0.68 \pm 0.03$	$0.82 \pm 0.39$	$1.01 \pm 0.33$	0.259	$2.64 \pm 0.42$	$1.79 \pm 0.46$

Table 2. *Continued.* Mass, cosmogenic  $^{21}\text{Ne}$ , production rates of  $^{21}\text{Ne}$  ( $P_{21}$ ), apparent  $^{21}\text{Ne}$  exposure ages ( $T_{21}$ ), and apparent precompaction times. Spallation-produced  $^{21}\text{Ne}$  concentrations are in  $10^{-8}\text{cm}^3\text{STPg}^{-1}$ . Production rates are in  $10^{-8}\text{cm}^3\text{STPg}^{-1}\text{Ma}^{-1}$ .

Sample	Mass ( $\mu\text{g}$ )	$^{21}\text{Ne}_c^a$	$^{20}\text{Ne}/^{21}\text{Ne}$	$^{22}\text{Ne}/^{21}\text{Ne}$	$P_{21}$	$T_{21}(\text{Ma})$	$T_{\text{pre}}^b(\text{Ma})$
Chondrule CH1							
MURCH1	234.0	$0.76 \pm 0.02$	$1.29 \pm 0.85$	$1.11 \pm 0.09$	0.302	$2.50 \pm 0.38$	$1.65 \pm 0.42$
Chondrule CH2							
MURCH2	159.0	$0.73 \pm 0.02$	$1.74 \pm 0.13$	$1.08 \pm 0.13$	0.395	$1.86 \pm 0.28$	$1.01 \pm 0.33$
Matrix olivine							
MUROL1	74.2	$0.45 \pm 0.02$	$2.55 \pm 0.45$	$1.05 \pm 0.33$	0.574	$0.80 \pm 0.12$	–
MUROL2	92.3	$0.58 \pm 0.03$	$1.20 \pm 0.28$	$0.99 \pm 0.22$	0.339	$1.73 \pm 0.27$	–
MUROL4	124.6	$0.25 \pm 0.01$	$1.77 \pm 0.49$	$1.07 \pm 0.40$	0.279	$0.90 \pm 0.14$	–
<b>Parsa (EH3)</b>							
Chondrule PR1							
PR1A12	2.9	$8.80 \pm 0.51$	$0.68 \pm 0.64$	$1.04 \pm 1.94$	0.672	$13.09 \pm 2.11$	$-3.91 \pm 3.31$
PR1A3	2.6	$14.10 \pm 0.71$	$0.71 \pm 0.43$	$0.91 \pm 1.35$	0.494	$28.54 \pm 4.52$	$11.54 \pm 5.94$
PR1A4	3.3	$10.00 \pm 0.56$	$0.72 \pm 0.49$	$0.93 \pm 1.50$	0.683	$14.61 \pm 2.34$	$-2.39 \pm 3.46$
PR1A5	2.9	$10.33 \pm 0.51$	$0.64 \pm 0.53$	$0.92 \pm 1.64$	0.483	$21.39 \pm 3.38$	$4.39 \pm 4.23$

<sup>a</sup>Cosmogenic  $^{21}\text{Ne}$  is found by subtracting the nominal  $^{21}\text{Ne}$  blank from the measured  $^{21}\text{Ne}$ . Corrections for trapped or atmospheric  $^{21}\text{Ne}$  are insignificant.

<sup>b</sup> $T_{\text{pre}}$  represents excess of  $^{21}\text{Ne}$  CRE ages in chondrule grains above that of respective matrix. To calculate  $T_{\text{pre}}$  for Parsa chondrules,  $T_{21}$  for Parsa matrix is considered 17 Ma, as reported by Bhandari et al. (1980). Uncertainties in apparent ages are primarily due to the propagated errors of target chemistry.

<sup>c</sup>Eugster et al. (2007).

<sup>d</sup>Polnau et al. (2001).

<sup>e</sup>Das and Murty (2009).

Table 3. Conventional CRE ages as reported in the literature for matrix and bulk samples.

Samples	Weight (mg)	$(^3\text{He}/^{21}\text{Ne})_c$	$(^{22}\text{Ne}/^{21}\text{Ne})_c$	CRE ages (Ma)			Source
				$T_3$	$T_{21}$	$T_{38}$	
Dhajala matrix	20.8	$4.31 \pm 0.27$	$1.09 \pm 0.02$	$5.0 \pm 0.6$	$3.8 \pm 0.6$	$3.8 \pm 1.5$	[1]
Bjurböle matrix	19.8	$3.44 \pm 0.16$	$1.06 \pm 0.02$	$8.4 \pm 0.4$	$8.0 \pm 1.2$	$7.9 \pm 1.7$	[2]
Chainpur matrix (100–200 $\mu\text{m}$ )	18.4	$2.06 \pm 0.39$	$1.09 \pm 0.02$	$10.8 \pm 1.5$	$25.9 \pm 2.7$	$23.5 \pm 3.3$	[3]

[1] Eugster et al. (2007); [2] Polnau et al. (2001); [3] Das and Murty (2009).

for comparison with these grains since the shielding assumptions (maximum production rates), target-specific production rates, and the computations will be the same for the grains and the matrix reference. However, the literature values for the CRE ages of the matrix are given for comparison in Table 3, the target chemistries in Table 1, and the computed CRE ages from production rates and chemistry in Table 2. As can be seen, the computed and literature values for the CRE ages are very similar, but there are some differences. We adopt CRE age of 17 Ma for Parsa matrix as reported by Bhandari et al. (1980). Three olivine grains from Murchison matrix were analyzed. Two of them had consistent exposure ages of 0.80 and 0.90 Ma, which we take as the matrix CRE age (a little lower than the accepted value), and grain MUROL2 had a higher CRE age of 1.73 Ma, which may represent some precompaction exposure of matrix olivine grains in Murchison similar to that observed by Hohenberg et al. (1990).

To detect statistically significant precompaction exposures of chondrule grains, we calculate the difference between the apparent CRE ages of the grain and the conventional CRE age of the matrix with compounded errors:

$$T_{\text{pre}} \pm \sigma_{\text{pre}} = (T_{\text{g}} - T_{\text{m}}) \pm \sqrt{\sigma_{\text{g}}^2 + \sigma_{\text{m}}^2} \quad (1)$$

where  $T_{\text{m}}$  and  $T_{\text{g}}$  are  $^{21}\text{Ne}$  cosmic ray exposure ages of the matrix and the corresponding grain and  $\sigma_{\text{m}}$  and  $\sigma_{\text{g}}$  are the associated errors.

### Apparent CRE Ages

#### Dhajala

In this work, we obtain a CRE age for Dhajala matrix of  $3.68 \pm 0.58$  (Table 2), essentially the same as Eugster et al. (2007) who report an exposure age of  $3.8 \pm 0.6$  Ma. The apparent CRE ages of olivine grains

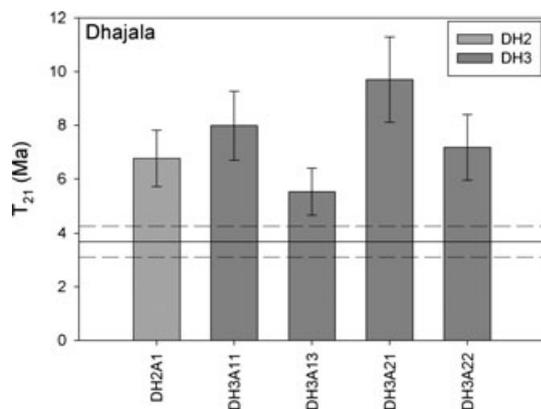


Fig. 1. Apparent  $^{21}\text{Ne}$  cosmic ray exposure ages for individual olivine grains extracted from Dhajala chondrules. The horizontal line shows the conventional  $^{21}\text{Ne}$  CRE age, with parallel dashed lines representing the error extremes ( $3.7 \pm 0.6$  Ma). As can be seen, grains DH2A1, DH3A11, DH3A21, and DH3A22 show CRE age higher by more than  $3\sigma$  compared with the conventional matrix CRE age, calculated using  $^{21}\text{Ne}$  concentrations and chemical composition reported by Eugster et al. (2007). All error bars are  $1\sigma$ .

extracted from two Dhajala chondrules DH3 and DH2, grains DH3A11, DH3A13, DH3A21, DH3A22 and DH2A1 are  $7.98 \pm 1.29$  Ma,  $5.52 \pm 0.88$ ,  $9.70 \pm 1.59$ ,  $7.17 \pm 1.22$ , and  $6.76 \pm 1.05$ , respectively, all larger than the CRE age of the Dhajala matrix, shown as horizontal lines in Fig. 1. Comparisons of these with the exposure age of Dhajala matrix, both our own value and that of Eugster et al. (2007), gives us confidence that these grains do indeed have significantly different exposures than the matrix, all larger, indicating precompaction exposures to energetic particles. Grain DH3A21 has the highest apparent exposure age of all the grains, a factor of two higher than that of the matrix, and three of the four other grains show apparent CRE ages that are higher by at least  $2\sigma$ . This is consistent with Eugster et al. (2007), and Das and Murty (2009), who reported apparent exposure ages for Dhajala chondrules that are longer than its conventional exposure age.

What is new here is that different apparent CRE ages have been observed for different grains separated from the same chondrule (DH3), and the apparent precompaction exposure age of DH3A21 is higher by a factor of 3.3 than that of DH3A13 ( $6.02 \pm 1.69$  Ma versus  $1.84 \pm 1.05$  Ma). What is unknown here is the relative position of these two grains within the chondrule. However, it should be noted that the host chondrule was more than 1 mm in diameter and the range of 10 MeV solar flares (typical of current solar energetic particles) is about 0.5 mm, so variability of exposures within a single chondrule does place restrictions on the particle energy for the precompaction irradiation, as will be discussed later.

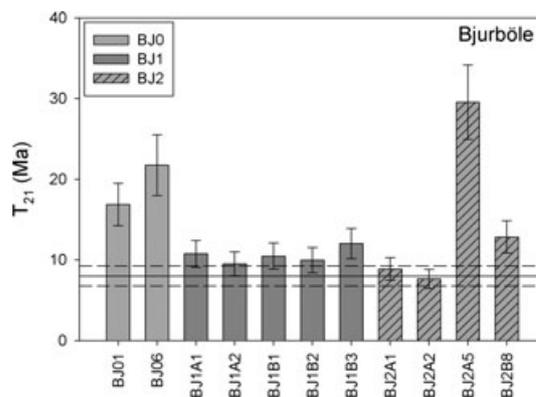


Fig. 2. Apparent  $^{21}\text{Ne}$  cosmic ray exposure ages for individual olivine grains extracted from Bjurböle chondrules. The horizontal line shows the conventional  $^{21}\text{Ne}$  CRE age, with parallel dashed lines representing the error extremes (i.e.,  $9.2 \pm 1.4$  Ma). Three grains BJ01, BJ06, and BJ2A5 show exposure ages higher by more than  $4\sigma$  compared with that of the matrix. Grain BJ2A5 shows highest apparent precompaction duration (at least 14 Ma), while three other grains from the same chondrule show little or no precompaction effects. The CRE age of Bjurböle matrix is calculated by using  $^{21}\text{Ne}$  concentrations and chemical composition reported by Polnau et al. (2001). All error bars are  $1\sigma$ .

### Bjurböle

Using measured cosmogenic  $^{21}\text{Ne}$  concentration, we obtain a CRE age of  $9.21 \pm 1.42$  Ma for Bjurböle matrix, the same within uncertainty of the  $8.01 \pm 1.23$  Ma found by Polnau et al. (2001). Out of eleven grains from three chondrules, three grains (BJ01 and BJ06 from chondrule BJ0 and BJ2A5 from chondrule BJ2) show apparent CRE ages higher by more than  $3\sigma$  compared with that of matrix, shown as horizontal lines in Fig. 2. For grain BJ2A5 the apparent CRE age is higher by  $4\sigma$ , representing minimum precompaction exposure of more than 20 Ma, twice that of the matrix, and among the highest of all the grains analyzed in this study. Polnau et al. (2001) reported  $^{21}\text{Ne}$  CRE ages for a group of chondrules from Bjurböle larger by  $>2\sigma$  compared with that of the matrix, thus also resolving an apparent precompaction irradiation, but diluted by the collective sampling (multiple chondrules). Das and Murty (2009) also reported a higher apparent CRE age for one of the Bjurböle chondrules studied by them.

We also observe that, like Dhajala, grains from the same chondrule in Bjurböle have had different energetic particle exposures. Grain BJ2A5 has an apparent precompaction CRE age of 20 Ma, whereas most of the other olivine grains in chondrule BJ2 have exposures consistent with that of the matrix (Table 2 and Fig. 2). Also, as stated earlier for Dhajala, variations of exposure

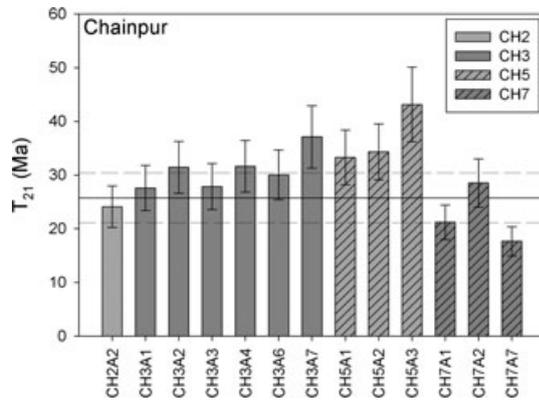


Fig. 3. Apparent  $^{21}\text{Ne}$  cosmic ray exposure ages for individual olivine grains extracted from Chainpur chondrules. The horizontal line shows the conventional  $^{21}\text{Ne}$  CRE age, with parallel dashed lines representing the error extremes ( $25.8 \pm 4.7$  Ma). CH5A3 shows a minimum apparent exposure age that is higher by at least  $3\sigma$  than the conventional CRE age. The CRE age of Chainpur matrix is calculated by using  $^{21}\text{Ne}$  concentrations and chemical composition reported by Das and Murty (2009). All error bars are  $1\sigma$ .

within a given chondrule can provide constraints for the irradiation energy.

#### Chainpur

Thirteen grains from four different chondrules from Chainpur were analyzed in this study. Among these, only CH5A3 shows an apparent CRE age that is larger than that of the matrix by more than  $2\sigma$  (Fig. 3). This excess spallation  $^{21}\text{Ne}$  corresponds to a minimum apparent precompaction exposure age of 17 Ma. The rest of the grains had CRE ages that within errors were more or less consistent with that of the matrix, but, given the higher conventional CRE age of Chainpur, it is more difficult to resolve precompaction exposures for this meteorite.

#### Murchison

We analyzed three olivine grains (MUROL1, MUROL2, and MUROL4) from the matrix of Murchison, one whole single chondrule (MURSPH) and two grains from two additional chondrules. The horizontal line in Fig. 4, based upon the average of three olivine grains from the matrix with the lowest apparent CRE ages, the two lowest values from this work (0.9 and 0.8 Ma), and the lowest value of an olivine matrix grain (1.10 Ma) reported by Roth et al. (2011). This average represents the best estimate (0.9 Ma) of the conventional CRE age of Murchison for this work, a value somewhat lower than that assumed by Hohenberg et al. (1990). We take the lowest value for the conventional CRE age here because it is impossible to have an apparent CRE age lower than the conventional CRE age, and because, in the work of Hohenberg et al. (1990), with very large

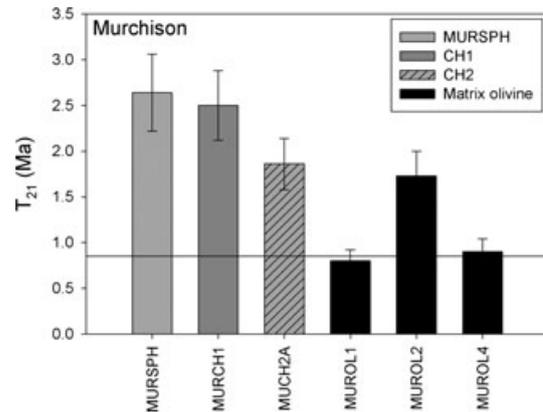


Fig. 4. Apparent  $^{21}\text{Ne}$  cosmic ray exposure ages for individual olivine grains extracted from Murchison chondrules and three olivine matrix grains. The horizontal line at approximately 0.9 Ma represents the average of the two lowest olivine matrix grain ages, the two lowest values from this work (0.9 and 0.8 Ma), and the lowest value of an olivine matrix grain (1.10 Ma) reported by Roth et al. (2011). Hohenberg et al. (1990) reported precompaction irradiation effects in Murchison olivines, so the lowest values may best represent the conventional CRE exposure. All error bars are  $1\sigma$ .

precompaction exposures observed in some grains, the difference between a conventional CRE age of 0.9 and 1.7 Ma is not very important.

Four of the six grains from Murchison show excess  $^{21}\text{Ne}$  from precompaction irradiations of a few Ma. Higher apparent precompaction exposure ages (up to 20 Ma) for Murchison chondrules have been recently reported (Roth et al. 2011; Huber et al. 2012). Hohenberg et al. (1990) observed large precompaction exposure effects for olivine grains extracted from Murchison matrix, requiring apparent CRE exposures of up to 145 Ma. In that study, precompaction irradiation effects were limited to the grains that contained solar flare heavy ion tracks, confirming their near-surface exposure in the regolith. The Murchison samples in our work consisted of only 3 chondrules whereas the Hohenberg et al. (1990) study started with several thousand olivine grains and selected approximately 40 with solar flare tracks and a comparable number without them. Given the limited statistics of our study, it is not surprising that we did not observe extremely long precompaction exposures.

#### Parsa

We used the reported CRE age of 17 Ma (Bhandari et al. 1980), the dashed horizontal line in Fig 5, as an estimate of the conventional CRE age of Parsa. While the data of Das and Murty (2009) would suggest a somewhat larger value, it would be greater than the apparent CRE ages of three of the four grains removed from the PR1 Parsa chondrule in this study. Although

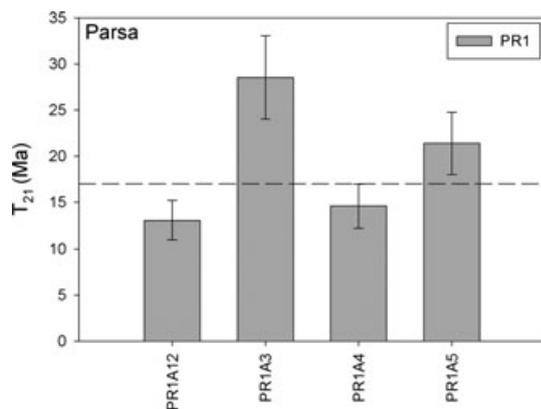


Fig. 5. Apparent  $^{21}\text{Ne}$  cosmic ray exposure ages for olivine grains extracted from a Parsa chondrule. The dashed horizontal line shows a reported  $^{21}\text{Ne}$  exposure age for Parsa of 17 Ma (Bhandari et al. 1980), but this value is not known with certainty. All error bars are  $1\sigma$ .

no evidence for large precompaction exposures is apparent for Parsa, there may be indications for smaller variations in this data, and in that of Das and Murty (2009), suggesting that some precompaction effects may exist for this meteorite.

### Precompaction Exposures

It is apparent that most of the grains within chondrules in this study, and most olivine grains removed from the matrix of the CM chondrites (Hohenberg et al. 1990), have exposure records identical with that expected if all were simultaneously irradiated after compaction of the final object. However, some olivine grains, both from chondrules and from the matrix, show excess amounts of spallation-produced  $^{21}\text{Ne}$  above that which could have been produced while collectively packaged. Eight grains (DH2A1, DH3A11, DH3A21, and DH3A22 from Dhajala; BJ01, BJ06, and BJ2A5 from Bjurböle; and CH5A3 from Chainpur) have had significantly higher exposure to energetic particles than reflected by their conventional CRE ages. These are not experimental artifacts.

While there could have been systematic gas losses from the fine-grained matrix material, yielding lower apparent CRE ages than their actual exposures, this is not a serious possibility given the broad data base for measured  $^{21}\text{Ne}$  and, in many cases  $^3\text{He}$ , concentrations. The more mobile  $^3\text{He}$  has not shown significant losses from either bulk matrix material, olivine grains in the matrix, or chondrules. Figure 6 shows  $^3\text{He}/^{21}\text{Ne}$  vs.  $^{22}\text{Ne}/^{21}\text{Ne}$  ratios for Dhajala, Bjurböle, and Chainpur matrix samples which plot within  $\pm 15\%$  of the “BERN line” (Eberhardt et al. 1965) indicating little or no  $^3\text{He}$

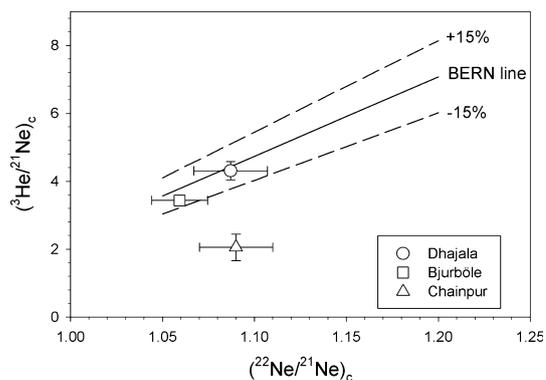


Fig. 6.  $^3\text{He}/^{21}\text{Ne}_c$  versus  $^{22}\text{Ne}/^{21}\text{Ne}_c$  for Dhajala, Bjurböle, and Chainpur matrix. A trend line with  $\pm 15\%$  error, referred to as the BERN line (Eberhardt et al. 1965), is shown, which represents a relationship between cosmogenic  $^3\text{He}/^{21}\text{Ne}_c$  and  $^{22}\text{Ne}/^{21}\text{Ne}_c$  in undisturbed ordinary chondrites. Samples with  $^3\text{He}$  losses would fall below the line, as perhaps seen for the Chainpur matrix, but no loss of  $^3\text{He}$  is anticipated in case of Dhajala and Bjurböle matrix samples.

losses, and certainly no  $^{21}\text{Ne}$  losses, and CRE ages (Table 3) are similar whether they are derived from  $^3\text{He}$ ,  $^{21}\text{Ne}$ , or  $^{38}\text{Ar}$  (Polnau et al. 2001; Eugster et al. 2007).

If the production rates were estimated incorrectly, due to either shielding or chemical composition differences, exposure comparisons may be compromised. However, the target chemistry was measured individually for each grain and production rates were based on these measurements. If differences were due to such an error, the apparent  $^{21}\text{Ne}$  excess (or deficit) might be expected to correlate with Mg, the major spallation target, but Fig. 7 shows this not to be the case. This is also borne out by the measurements of individual olivines in bulk Murchison and Murray most of which have apparent CRE ages that cluster around the conventional CRE values, but with some individual grains of identical composition that have much larger amounts of spallation-produced  $^{21}\text{Ne}$  (Hohenberg et al. 1990). We thus must conclude that grains with excess spallation-produced  $^{21}\text{Ne}$  were individually exposed to galactic cosmic rays, to solar cosmic rays (solar flares), or to some other energetic particles prior to compaction into the final object.

## DISCUSSION

### Precompaction Exposure of Chondrules in Nongas-Rich Meteorites

In general, the minimum apparent CRE ages for individual grains taken from chondrules are similar to those of the host matrix material and reflect spallation production during their conventional cosmic ray

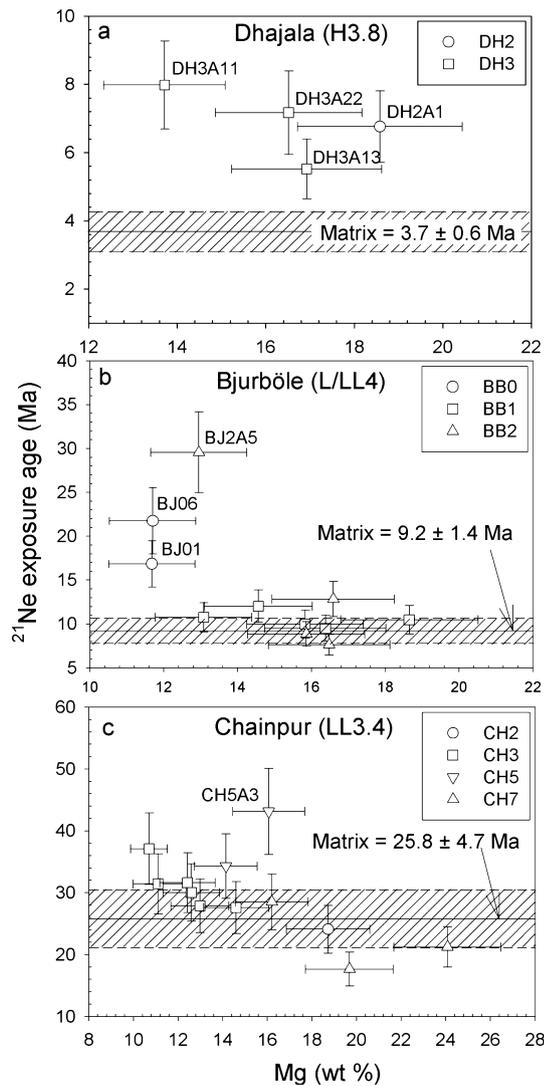


Fig. 7. CRE ages based on  $^{21}\text{Ne}$  are compared with Mg content in the chondrule grains separated from Dhajala, Bjurböle, and Chainpur. Mg is the major target element for production of  $^{21}\text{Ne}$  so the determined concentration of Mg could have a significant effect on the computed CRE age. However, there is no relationship between observed cosmogenic  $^{21}\text{Ne}$  excesses and Mg content, so any incorrect assessment of target chemistry does not seem to be responsible for these excesses.

exposure ages. Spallation-produced noble gases in excess of those which could have been produced during this collective exposure period must have been the result of energetic particle irradiation prior to compaction of the final object. While precompaction irradiation effects have been shown in olivines extracted from the matrix of the CM chondrites Murchison and Murray (cf. Hohenberg et al. 1990; Woolum and Hohenberg 1993), this study demonstrates that this effect also exists within grains extracted from individual chondrules. In addition,

others have reported precompaction irradiation effects in whole chondrules from a variety of meteorites.

Polnau et al. (1999, 2001) reported precompaction exposure durations of approximately 1 Ma for a large chondrule fragment from the H6 chondrite ALH76008 (from cosmogenic  $^3\text{He}$ ,  $^{21}\text{Ne}$ , and  $^{38}\text{Ar}$ ), and approximately 3 Ma for a bulk collection of chondrules from the Bjurböle chondrite (although the effect is clearly diluted by treating them collectively). More recently, precompaction exposures of up to 2–4 Ma were reported for Bjurböle and Dhajala chondrules (Eugster et al. 2007; Das and Murty 2009). A cursory look at the statistics would suggest that the fraction of grains or chondrules that show such precompaction irradiation effects may be similar.

Extensive precompaction irradiation by energetic particles for matrix olivine grains and chondrules from Murchison and Murray suggested that these constituents were exposed to energetic particles while residing on the parent body surface (Hohenberg et al. 1990; Woolum and Hohenberg 1993; Roth et al. 2011; Huber et al. 2012). Hohenberg et al. (1990) and Woolum and Hohenberg (1993) concluded that this irradiation must have occurred during a period in which the particle flux was enhanced, higher than supported by the present SCR or GCR (with the secondary cascade) environment. Therefore, there are two questions that need to be answered: Can the present nominal particle flux account for the observed irradiation effects within the time constraints placed on it by the known solar system chronology and where did the irradiation take place, in the parent body regolith or in the nebula?

The relative ages of chondrules and CAIs from various meteorites derived from numerous studies of short-lived radionuclides (e.g.,  $^{26}\text{Al}$ - $^{26}\text{Mg}$ ,  $^{53}\text{Mn}$ - $^{53}\text{Cr}$ , and  $^{129}\text{I}$ - $^{129}\text{Xe}$ ), as well as their respective Pb-Pb ages, constrain chondrule formation to within 2–3 Ma of CAI formation (Amelin et al. 2002; Bizzarro et al. 2004; Kita et al. 2005; Hohenberg and Pravdivtseva 2008; Rudraswami et al. 2008; Krot et al. 2009). Some relict chondrules have even been observed within CAIs (Krot et al. 2004), also suggesting nearly contemporary formation of chondrules and CAIs. The most rigorous time constraint we have between CAI formation and final meteorite compaction applies to the CI chondrite Orgueil and, in that case, the final meteorite must also have formed shortly after CAI formation (< 10 Ma), as evidenced by the Sr isotopes in carbonates observed as veins found in Orgueil, which clearly cannot predate the meteorite (Macdougall et al. 1984). However, there is much evidence (DuFresne and Anders 1962; Macdougall and Kothari 1976; Richardson 1978; Kerridge and Bunch 1979) that limits the duration to less than the 300 Ma for Murchison and Murray matrix olivines

(Hohenberg et al. 1990; Woolum and Hohenberg 1993) and <40 Ma for some of the precompaction effects reported here.

The required precompaction exposure times are essentially the same for both of the present sources of irradiation, the contemporary SCR flux or the contemporary GCR flux, as the *maximum* production rates are almost the same whether irradiated in near-surface material by SCR particles or by fully developed secondary GCR cascades at the optimum shielding depth (Hohenberg et al. 1990). This leaves insufficient time for precompaction spallation effects to occur either in the nebula or within the regolith of the parent body if these effects are due to normal (contemporary) solar flare (SCR) or galactic cosmic ray (GCR) effects. With the contemporary particle environment, there is simply not enough time for these precompaction irradiation effects to be produced.

Wieler et al. (1989, 2000) propose that the contemporary GCR flux can account for these precompaction effects, but these authors base their argument on the gas-rich meteorites Kapoeta and Fayetteville. However, those gas-rich meteorites do not play any role whatsoever in the conclusions reached by Hohenberg et al. (1990), Woolum and Hohenberg (1993), and others. For the reader, the term “gas-rich” is used for meteorites, like Kapoeta and Fayetteville that have had extensive regolith histories, as evidenced by their large amounts of light noble gases implanted in these by the solar wind. In contrast, carbonaceous meteorites, like the CM chondrites Murchison and Murray, contain large amounts of “planetary” heavy noble gases, carried by “phase-Q” that is not related to the light noble gases implanted by the solar wind in gas-rich meteorites. Kapoeta and Fayetteville were simply the first meteorites studied for precompaction irradiation and, although many grains from gas-rich meteorites have had an extensive precompaction irradiation, there is no time constraint on the duration of regolith exposure, like for lunar samples, so those meteorites cannot be used as an argument for or against an enhanced early particle flux. They may well have sufficient time in the regolith for the observed spallation effects to be produced by normal GRC effects, as argued by Roth et al. (2011) and Wieler et al. (2000).

In addition, Roth et al. (2011) discuss the relationship between solar flare track density and the quantity of spallation-produced noble gases in olivines from the gas-rich meteorites in which the “solar flare track densities are much lower than that expected for concurrent solar flare production of cosmogenic  $^{21}\text{Ne}$ .” This is not relevant as gas-rich meteorites do argue for the solar flare case, one way or the other, enhanced or not enhanced. The presence or absence of solar flare

heavy ion tracks in the single grain studies was used only as an indicator for whether the grain had been exposed at the top of the regolith or not. They were often not counted, just noted as “minimum track densities” (Hohenberg et al. 1990) in an effort to see if spallation-produced  $^{21}\text{Ne}$  was enhanced in those grains that had spent some time at the very surface of the parent body regolith. It was found that no olivine grains without solar flare tracks in Murchison and Murray contained precompaction spallation  $^{21}\text{Ne}$ , they all had the amount that would be produced during their conventional cosmic ray exposure age, and many grains that had solar heavy ion tracks also had large amounts of precompaction spallation  $^{21}\text{Ne}$  (Hohenberg et al. 1990; Woolum and Hohenberg 1993).

It should be noted that over 3000 original grains were examined in the studies summarized by Hohenberg et al. (1990), far too many to be individually measured in the mass spectrometer. The purpose of those studies was to look for precompaction  $^{21}\text{Ne}$ , so a representative subset of grains with no solar-flare tracks could be compared with a subset containing observable solar flare tracks. The statistics of a starting population of more than 3000 provides a more definitive study that is made with a dozen or so. That is why those earlier studies may be more pertinent in deciphering the details of precompaction spallation in these objects than the randomly selected chondrules and chondrule parts reported in later studies.

#### **Different Irradiations Within Single Chondrules and Implications**

Our data have demonstrated that precompaction effects exist not only within the matrix and individual chondrules, but within different grains of a single chondrule as well. Grain BJ2A5 of the BJ2 chondrule from Bjurböle chondrite has an apparent excess CRE of approximately 20 Ma compared with other grains from the same chondrule. While the “age” difference is clearly not meaningful as it is a model age based upon the nominal shielding during the conventional cosmic ray exposure, it clearly shows that one part of the chondrule received a different particle dose than another. Similarly, grains from the DH3 chondrule from Dhajala show varying doses of equivalent CRE “ages” of 1.8, 3.1, 3.5, 4.3, and 6.0 Ma, and those from Chainpur range from 0 to 17 Ma.

The variation in apparent precompaction exposure ages of individual grains from within the same chondrule may indicate that the spallation-produced  $^{21}\text{Ne}$  from the precompaction irradiation was attenuated on a scale similar to that of the chondrule size. This suggests that the precompaction exposure effects in the chondrules,

and the grains within them, may be due to irradiation by particles with energies lower than those of primary galactic cosmic rays which, with their secondary cascade, do not attenuate over this distance. The olivine grains we studied were obtained from chondrules larger than 1 mm in size. Using the SRIM (The Stopping and Range of Ions in Matter) software package (Ziegler 2004), we find the projected range of 10 MeV protons to be approximately 0.5 mm in a Mg-Al-Si target having density of 3.3 g/cm<sup>3</sup>. As these distance scales are similar, chondrules exposed to particles of this energy range would have spallation effects, from the proton or secondary neutron, that attenuate quickly with the depth, even within a single chondrule. The outer layers may receive a greater dose and would contain more spallation-produced <sup>21</sup>Ne from precompaction irradiation by lower energy particles than those deeper inside. It is unfortunate that we do not know the specific location of the grains within a given chondrule. This is because the chondrule had to be fragmented to search for olivine grains and the grain location was lost during this process. Variation in production rate across an individual chondrule, observed in this work, constrains the energy of the irradiation, suggesting that an enhanced solar, rather than the more energetic GCR irradiation produced these effects. This same conclusion was reached by Hohenberg et al. (1990) and Woolum and Hohenberg (1993), but, for an entirely different reason, reviewed in the last paragraph.

Roth et al. (2011) suggest that the olivine grains recovered from Murchison and Murray matrices were once part of chondrules and received their precompaction irradiation as chondrules within the parent body regolith. Like Hohenberg et al. (1990) and Woolum and Hohenberg (1993), these authors argue against an irradiation in the nebula for there is simply not enough time, and they point out that not all chondrules are irradiated. However, if the precompaction irradiation occurred as chondrules in the regolith, and the olivine grains from the Murchison and Murray matrix were once parts of chondrules as suggested by Roth et al. (2011), we would not observe that different olivine grains within a single chondrule have different doses, an argument that rules out GCR irradiation at the same time. Moreover, there is simply insufficient time for the observed precompaction irradiation by galactic cosmic rays *regardless of anything else*. Even at the optimum depth in the regolith (for maximum production), it would take more than 300 Ma of GCR exposure to produce the observed <sup>21</sup>Ne in some of these grains, clearly too long to wait for the compaction of Murchison and Murray (Hohenberg et al. 1990; Woolum and Hohenberg 1993).

To summarize: (1) Precompaction irradiation effects in meteorites most likely occurred in the regolith of their

parent bodies. (2) For at least the CM meteorites, there is insufficient time for the precompaction irradiation effects to be caused by the contemporary suite of energetic particles in the solar system, whether SCR or GCR. An enhanced source of energetic particles in the early solar system seems to be required. Before this work, it was concluded that an enhanced irradiation by an early active sun was most likely a conclusion based upon the argument that, at  $4.56 \times 10^9$  years ago, the galaxy was pretty much the same as it is today, but the Sun at that time was probably transitioning from an early active (T-Tauri) stage to the main sequence. After removal of most nebular gas from the inner solar system, the “naked T-Tauri” stage (Feigelson and Kriss 1989; Bary et al. 2002), energetic particles from the intense flares can get through. This work further adds support with the observation of different production rates in different parts of a chondrule, restricting the range and constraining the energy of the irradiation to be lower, more consistent with solar flares than galactic cosmic rays. In further confirmation, Feigelson et al. (2002) report that Chandra X-Ray Observatory data for young stars suggest that the naked T-Tauri phase of our own Sun may have been accompanied by solar flares 10<sup>5</sup> times more intense than current activity, sufficient to produce the observed precompaction <sup>21</sup>Ne seen in these grains.

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