COSMIC RAY EXPOSURE HISTORY OF INDIVIDUAL CHONDRULES FROM ALLEGAN H5 ORDINARY CHONDRITE PROBED BY ^{81}Kr-Kr CHRONOMETER

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Introduction: Radioisotope chronometers e.g. Pb-Pb, ^{26}Al/^{27}Al or ^{53}Mn/^{55}Mn indicate chondrules found in ordinary chondrites were formed ~2 Ma after CAIs. Whereas some models propose they originated in the solar nebula as small objects, others consider formation on the surface of asteroids and protoplanets as a result of collisions [1]. If chondrules were formed in the solar nebula and later accreted they are expected to contain cosmogenic isotopes produced prior to accretion, though the concentrations may have changed due to diffusion loss during metamorphism. After breakout from the parent asteroid induced by a collision the meteoroids again become subject to cosmic ray exposure. The ratio of cosmogenic stable isotope to cosmogenic radioactive isotopes produced during meteoroid transit to Earth (e.g. ^{81}Kr/^{stable}Kr) can be used for calculation of its integrated exposure history (apparent CRE age). Thus the CRE ages of individual chondrules and meteoritic matrix can constrain the extent of prexposure (including the time of accretion before chondrules). The CRE ages of chondrules with cosmogenic gases acquired before their accretion would be longer than those of matrix. In turn, variations between the ages of individual chondrules may point to a different formation mechanism.

Previous works on H-chondrites occasionally reported differences in concentrations of cosmogenic isotopes of bulk samples [2, 3]. Polnau et al. [4] studied mutiple chondrules (total weight of each sample 20-50 mg) separated from eight chondrites of a higher petrologic type using ^{3}He, ^{21}Ne, ^{38}Ar chronometer and reported a small excess in favor of chondrules. The same group reported the CRE age determined for one individual chondrule of ALH76008 H6 chondrite [5]. This meteorite has the age of 1.72±0.11 Ma. The ^{3}He, ^{21}Ne, ^{38}Ar CRE ages of the chondrule are higher by 31%, 67% and 55% respectively. These methods however require calculation of concentrations of target elements and associated production rates. The ^{81}Kr-Kr method, in contrast, is independent of shielding and calculation of production rates [6]. Theoretical calculations for H chondrites using excitation functions of nuclear reactions, cross sections and primary and secondary particle spectrum show the method is also not susceptible to change in concentration of target elements (for Rb, Sr, Y, Zr concentrations of 0.1-10 of those of usual H-chondrites the variations in the ^{81}Kr-Kr CRE ages <3‰) [7].

The main limitation of ^{81}Kr-Kr method has been the low concentration of ^{81}Kr in meteorites. The method has been applied mainly to eucrites (~1-4x10^{3} of ^{81}Kr atoms/mg) having the highest concentrations of target elements. In contrast, a single ~10 mg chondrule contains at most a few hundred ^{81}Kr atoms. We have developed a resonance ionization mass spectrometer (RIMSKI) capable of determining Kr isotope ratios at this level [8]. It combines a laser resonance ionization ion source with a high transmission time-of-flight mass spectrometer and a cryogenic sample concentrator. The method has been established using eucrite samples of <5mg (x100 improvement in sample size) and allows isotope ratios determination with comparable precision [9]. Recently, sensitivity has been improved still further (Fig. 1). Here for the first time we present the ^{81}Kr-Kr CRE age data for individual chondrules and matrix of Allegan H5 chondrite.

Technique, samples and ^{81}Kr-Kr chronometer: Laser heating is used for extraction of krypton from samples into the mass spectrometer volume. Atoms continuously condense on a cold spot in the ion source, and are repeatedly released by laser heating. They are ionized in the evaporation plume using three tunable dye lasers and one frequency tripled Q-switched Nd:YAG laser. Two resonant transitions are employed (at 116.5 nm, produced by four-wave frequency mixing in Xe, and 558.1 nm) and are followed by single photon ionization at 1064 nm. Multiple analyses of air aliquots and <5 mg samples of meteorites, both containing ~10^{6} total krypton atoms, show precision ~1% for major isotope ratios. They are reproducible to this level through a day's analyses, allowing calibration for linear and non-linear mass discrimination by sample-standard bracketing.

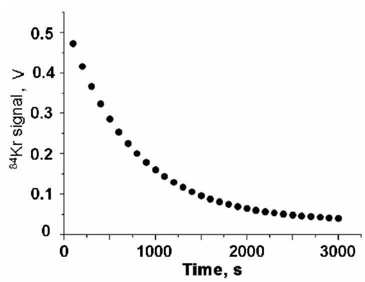

Fig. 1 ^{84}Kr pump out curve: the Kr signal decreases as ionized atoms are removed from the ion source and implanted into the detector The fast signal decrease indicates efficient ionization of atoms.

In this pilot study we studied chondrules previously separated from the Allegan H5 chondrite (Allegan USNM215, provided by Smithsonian's National Museum of Natural History, Washington DC) by gentle crushing and polished under the optical microscope to remove the matrix. ~10 chondrules of <10 mg have been separated for analysis (here we present our first measurements of 4). The matrix powder was collected using 25 µm sieve. In principle, the collected matrix is not guaranteed to be free of crushed chundrules, but indeed this material can be regarded as a chondrule-depleted.

T_{81}, the ^{81}Kr-Kr CRE age (in My) is calculated using the following equations [10]:

$$T_{81}(\text{My}) = \tau_{81}\left(\frac{P_{81}}{P_{83}}\right)\left(\frac{^{83}Kr}{^{81}Kr}\right)_c, \quad \frac{P_{81}}{P_{83}} = 1.262\,\frac{^{78}Kr}{^{83}Kr} + 0.381,$$

where the subscript "*c*" refers to the cosmogenic composition, τ_{81} = 0.330 My is the mean lifetime of ^{81}Kr (the half-live has been recently revised from 2.13x10^{5} a to 2.29x10^{5} a [10]) and P_{81}/P_{83} is the ratio of the production rates of ^{83}Kr and ^{81}Kr. Experimental uncertainty is controlled predominantly by the uncertainty in the ^{81}Kr measurement.

Results and discussion: The calculated ^{81}Kr-Kr ages for chondrules and matrix are presented in the table 1 and Fig. 2 a). Several heating steps have been performed for both the chondrules and matrix; for each step the ages were determined. A typical TOF spectrum of a chondrule is presented in Fig.2 b). The estimated content of this isotope is only a few hundred atoms. Some interference with benzene C_6H_6 observed at mass 78. This compound presumably comes from the oil vapours of the turbo pumps. It is condensed on the cold finger throughout the day and negligible at the beginning of the day linearly increasing to ~10^4 atoms Kr equivalent after 3 hours of operation. As air aliquots have been measured in-between the samples, it was possible to estimate the amount of hydrocarbon and substract it from the 78 peak. The bromine content is low in this chondrite so we also calculated the ages using production rates as:

$$\frac{P_{81}}{P_{83}} = 0.95\frac{^{80}Kr + ^{82}Kr}{2\,^{83}Kr} \quad [6]$$

and found them identical within uncertainties.

On the Fig. 2 a) the average age for bulk meteorite is represented by the horizontal line. The determined age of the chondrules and those of bulk meteorite are identical within uncertainties which are on average 20%. The calculated average values for each chondrule (open dots) correlates even better with the bulk number. The situation is the same with the matrix ages which are also correspond to the bulk data. Their uncertainties are higher due to higher content of trapped krypton.

Table 1. CRE ages of Allegan H5 chondrules and its matrix.

Chondrules		
Weight	Laser power	CRE age, Ma
10 mg	5 W	4.18±0.86
	11 W	5.17±0.56
	16 W	5.01±1.61
		average: 4.89±0.45
6 mg	4 W	5.33±1.18
	12 W	8.16±2.87
		average: 5.74±1.09
7 mg	8W	7.41±1.22
9 mg	7 W	4.08±0.91
	10 W	7.84±2.21
	17 W	8.32±1.57
		average: 5.45±0.74
		Average chondrles: **5.30±0.35**
Matrix		
Weight	Laser Power	CRE age, Ma
9 mg	9 W	6.49±4.05
	13 W	7.50±2.25
10 mg	8 W	6.40±4.09
	14 W	6.92±2.79
5 mg	14 W	6.62±2.14
		Average matrix: **7.04±1.49**

We calculate the weighted averages for all the heating steps of the chondrules (5.30±0.35 Ma) and matrix (7.04±1.49 Ma) and compare them to the bulk age (6.09±0.93 Ma) [7] on Fig. 2c). From this figure it is clearly seen that these numbers are identical within experimental uncertainties. It means either the Allegan lost its gases as a result of igneous processes or collisions (but precompaction gases were detected in ALH76008 H6 which is even higher petrologic type [5]) or its chondrules were accreted too rapidly (<1 Ma), beyond the ^{81}Kr-Kr method resolution.

We have established a reliable ^{81}Kr-Kr chronometer for individual chondrules detection and in future will extend the data set focusing on the samples of lower petrologic type having small CRE ages.

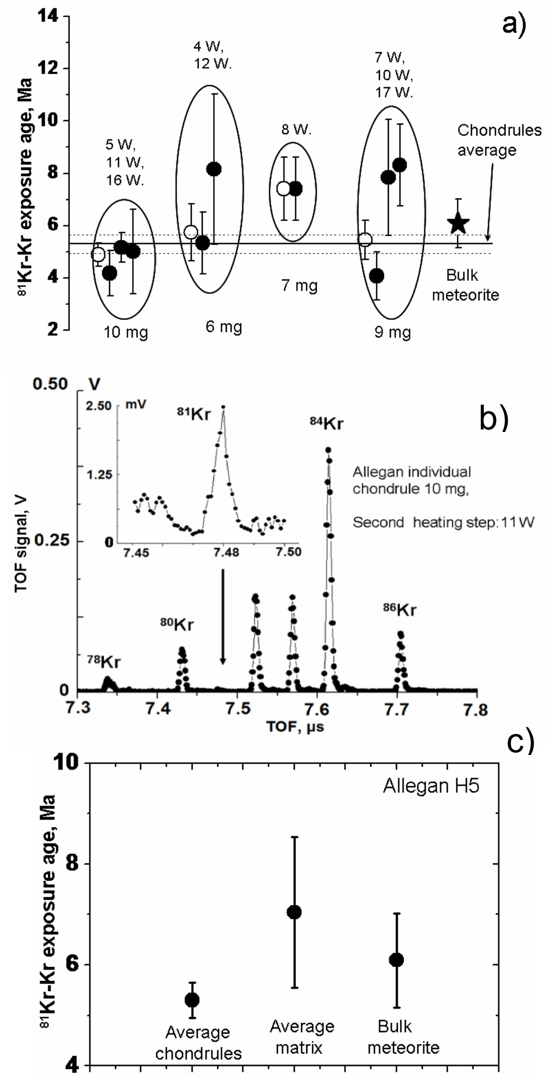

Fig.2 a) ^{81}Kr-Kr ages determined for 4 individual chondrules from Allegan H5 chondrite (filled dots). The lase g have been used for gas extraction. The laser power used at each step is indicated. The open dots are the average age calculated for each chondrule. b) TOF spectrum collected for 10 mg chondrule during the second heating step (11W). The inset is enlarged ^{81}Kr region (<1000 atoms) c) Fig.2 Comparison between ^{81}Kr-Kr exposure ages of chondrules, matrix and bulk [7] of the Allegan H5 ordinary chondrite.

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References: [1] Boss A.P. (1996) A concise guide to chondrule formation models. In: Chondrules and the Protoplanetary Disk, Cambridge University Press, London, 257-263. [2] Vogt S. et al. (1990) Rev. Geophysics 28, 253-275. [3] Herzog G.F. (1997) Meteoritics 32, 413. [4] E. Polnau et al., (2001) GCA 65, 11, 1849 [5] E. Polnau et al. (1998) GCA 63, 6, 925 [6] Marti K. (1967) Phys Rev. Lett., **18**, 264-266. [7] I. Leya (2004) Antarct. Meteorite Res., 17, 185-199, 2004. [8] I.Strashnov et al. (2011) JAAS 26, 9, 1763. [9] I.Strashnov (2012) LPSC abstracts.