# $^{81}$Kr-Kr COSMIC RAY EXPOSURE AGE OF THE PUERTO LAPICE (AND OTHER) EUCRITES

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**Introduction:** The HED group of meteorites is linked to the asteroid Vesta through similarities in the IR and visible spectra [1]. It is considered the most probable parent body because its location at 2.36 AU (semi-major axis) is near the 3:1 orbital resonance with Jupiter (the Kirkwood gap is at 2.5 AU), and this seems to provide a mechanism for relatively quick (<100 My) delivery to Earth of material ejected by impacts. This hypothesis is supported by the recent discovery of the Vestoids – several small (<10 km in diameter) main belt asteroids with apparent HED compositions at 2.31-2.45 AU [2]. They are thought to have been ejected from Vesta by collisions with other bodies, some of them gaining high enough velocities to reach resonance orbits from which, simulations show [3], it would take ~ 1 My to become an Earth crosser.

Several recent findings indicate that some meteorites classed as eucrites do not originate from the same parent body as the majority of the HED group. Among these "anomalous eucrites", Pasamonte and Ibitira show reproducible oxygen isotope signatures different from those of the rest of the "eucrites" [4]. Bunburra Rockhole (BR) – a recent recovery by the Desert Fireball Network from the Nullarbor desert in Australia [5] – is heterogeneous in oxygen isotopes among fine-, medium- and coarse-grained lithologies. This indicates that, unlike Vesta [6], the BR parent body experienced less early stage melting and did not completely equilibrate with respect to oxygen isotopes.

Another valuable piece of information can be obtained by determining the time between fragmentation of the meteoroid from a parent body and its arrival on Earth. Cosmic ray induced spallation reactions with Sr, Y and Zr produce $^{81}$Kr ($T_{1/2}$=2.29x10$^{5}$ y) and several stable isotopes – this forms the basis of $^{81}$Kr-Kr exposure age dating [7]. The production rate of krypton isotopes depends on both chemistry and cosmic ray flux, but after several half lives the concentration of $^{81}$Kr achieves an equilibrium between production and radioactive decay allowing the (unknown) production rate of krypton isotopes to be inferred from the (well known) decay rate. Stable cosmogenic isotopes accumulate for the duration of the exposure, so once the production rate is known the duration of exposure to cosmic rays can be calculated.

In part to extent the range of application of the $^{81}$Kr-Kr system, we have recently developed RIMSKI – Resonance Ionization Mass Spectrometer for Krypton Isotopes [8]. In this work we present further results from systematic application of RIMSKI to $^{81}$Kr-Kr analysis of eucrites. Our goals are to demonstrate that RIMSKI can obtain $^{81}$Kr-Kr ages consistent with previous analyses, but from samples smaller by two orders of magnitude, to extend the dataset of eucritic falls with known $^{81}$Kr-Kr ages and to re-examine the impact history of the parent body/bodies. In this work we expand our previously reported dataset with a Kr-Kr age for the recent eucrite fall Puerto Lapice [9], and revisit the assignation of normal eucrites to $^{81}$Kr-Kr age clusters.

**Experimental technique:** The development and current configuration of RIMSKI are described in detail elsewhere [8]. Briefly, krypton is extracted from samples by continuous wave laser step-heating (1064 nm). The gas is admitted into the mass spectrometer, where atoms continuously condense on a cold spot; this cold spot is maintained in the back plate of the Wiley-McLaren ion source by a cold stage held at 75K by a liquid helium cryogenerator. Accumulated krypton atoms are released from the cold spot by a pulsed 1064 nm laser with a duty-cycle of 10 Hz; ionization takes place in the plume. A laser system consisting of one Q-switched Nd:YAG laser pumping three tunable dye lasers is used to generate the wavelengths needed for ionization. Two one-resonant transitions are employed. The first step requires light with a wavelength of 116.5 nm, which is produced by four-wave sum-frequency mixing (two 252.5 and one 1507.1 photons) in a Xe-Ar mixture. Then transition at 558.1 nm followed by ionization at 1064 nm. Ions are accelerated into the flight tube and detected using a pair of MCPs.

The precision of ~1% for major isotope ratios have been demonstrated by multiple analyses of air aliquots containing ~10$^{6}$ total krypton atoms. Analyses are reproducible to this level through a day's analyses, allowing calibration for linear and non-linear mass discrimination by sample-standard bracketing. Developed apparatus has a low blank (~5800 total Kr atoms with atmospheric composition) and highly sensitive (<1000 atoms).

**CRE ages and their clusters:** All meteorites except Bunburra Rockhole and Puerto Lapice were provided by the Natural History Museum, London. Bunburra Rockhole was obtained from the sample curator of the Desert Fireball Network and the Puerto Lapice was provided by the Universitat Politecnica de Catalunya. Determined CRE ages are summarized in the Table 1. Typically ~6 grains (1-4 mg each) of each eucrite were used for analysis. The resulted CRE ages are the weighted means of individual analyses.

$T_{81}$, the $^{81}$Kr-Kr CRE age (in My) is calculated using the following equations [9]:

$$T_{81}\,(\mathrm{My}) = \tau_{81}\left(\frac{P_{81}}{P_{83}}\right)\left(\frac{^{83}Kr}{^{81}Kr}\right)_c ,$$

$$\frac{P_{81}}{P_{83}} = \frac{0.95}{2}\left[\left(\frac{^{80}Kr}{^{83}Kr}\right)_c + \left(\frac{^{82}Kr}{^{83}Kr}\right)_c\right] ,$$

where the subscript "$c$" refers to the cosmogenic composition, $\tau_{81}$ = 0.330 My is the mean lifetime of $^{81}$Kr and $P_{81}/P_{83}$ is the ratio of the production rates of $^{83}$Kr and $^{81}$Kr.

We compare our data with literature CRE ages (where available) in Table 1 Because the $^{81}$Kr half-live has been recently revised from 2.13x10$^{5}$ y to 2.29x10$^{5}$ y [11], for the purpose of this comparison literature ages derived using the old half-live have been scaled to the new half life. The ages and literature values determined in this work agree within experimental uncertainties. Experimental uncertainty is controlled predominantly by the uncertainty in the $^{81}$Kr measurement. Our uncertainties vary over 4.2-8.6% with an average value of 6.2% and are comparable to those reported by other groups for analyses of x100 larger samples.

The CRE ages of eucrites appear to define clusters. We used the Akaike information criterion (AIC) [12] to determine the number of clusters and assign ages. The models are characterised by the number of originating impacts, $n$, which is allowed to range between 1 and 10. We first define a $\chi^2$ statistic $\chi_n^2 = \sum_i \left( \frac{T_{81,i} - T_n}{\sigma_i} \right)^2_{\min}$ Where the subscript "min" indicates that only the impact in the model with the highest probability of producing each meteorite has been taken into consideration; in effect the finite probability that it was produced from each of the other impacts in the model has been neglected. For each model labeled by $n$, the number of originating impacts it considers, we first find the set of $n$ impact ages that minimize $\chi^2$. We then calculate the AIC appropriate for the case where the number of data does not greatly exceed the number of parameters in the models under consideration, which is conventionally labeled $\mathrm{AIC_c}$:

$$\mathrm{AIC_c} = \chi^2_{n,\min} + 2n + \frac{2n(n+1)}{i-n+1}$$

here $n$ and $i$ are the numbers of parameters (originating impact times) and meteorites respectively and $\chi^2_{n,\min}$ is the minimum $\chi^2$ for the model with $n$ parameters. The best model is that which minimizes $\mathrm{AIC_c}$. For models where the number of data is very much higher than the maximum number of free parameters, the final term approaches zero and $\mathrm{AIC_c}$ approaches AIC = $\chi^2_{n,\min}$ +2$n$.

To determine the maximum likelihood set of $n$ timings of originating impacts for each model an initial set of $n$ timings randomly spaced between 0 and 60 Myr was selected. The solver™ routine of MS Excel™ was then used to find the local minimum $\chi^2$ accessible from these initial conditions. We adopted the set that minimized $\chi^2$ over a series of iterations of this process, each starting from a different randomly selected initial set of timings; the number of iterations was increased for larger values of $n$. This process was itself repeated twice and found the same optimum set of initial timings for each model with $n$ < 8, suggesting this approach is robust to the presence of local minima that are not global minima. The $\chi^2$ corresponding to this set of timings was then used with $n$ to calculate $\mathrm{AIC_c}$ for the model.

A Monte Carlo approach was used to estimate the uncertainties on the timings of originating impacts. Twenty simulated datasets were generated from the original data by choosing a modeled CRE age for each meteorite at random from a normal distribution based on its age and associated error. Macibini and Serra de Mage (due to high uncertainties) and the anomalous eucrites have not been included in the calculations. The calculated timings of four impact events (n=6 model) are essentially similar to those of Shukolukov and Begeman: 10.6± 0.4 My, 14.4± 0.6 My, 21.7± 0.4 My, 37.8± 0.6 My, with the addition of a fifth at 25.4 ± 0.4 My. This new cluster is supported by our measurements of Lakangaon (25.3± 1.1 My, determined for the first time), Puerto Lapice (22.12±1.13) and Bereba (25.0±1.1 My); the Bereba value is similar to that of Miura (25.5± 0.6 My) [13] and Freundel (24.9± 2.3 My) [14] but smaller than reported by Shukolukov and Begeman (26.1± 2.2 My) [15].

| | $T_{81}$ Our data | $T_{81}$ literature | |
|---|---|---|---|
| Bereba | 25.0±2.1 | 27.2±2.3 | [15] |
| | | 24.9±2.3 | [16] |
| | | 25.5±0.6 | [13] |
| Binda | | 21.9±1.7 | [13] |
| Bouvante | 7.9±0.5 | 7.0±0.4 | [14] |
| Bunburra Rockhole | 27.6±1.4 | | |
| Cachari | | 9.4±1.1 | [15] |
| Caldera | | 14.8±1.5 | [15] |
| Camel Donga | | 41.5±2.8 | [15] |
| | | 38.1±1.5 | [13] |
| Chervony Kut | | 37.9±1.5 | [15] |
| Jonzac | | 37.7±1.5 | [15] |
| Juvinas | 11.9±0.5 | 11.0±0.4 | [15] |
| | | 9.95±0.7 | [16] |
| | | 11.2 ±0.9 | [13] |
| Ibitira | 13.3±0.7 | 13.4±2.2 | [15] |
| Lakangaon | 25.3±1.1 | | |
| Macibini | | 50.0±4.2 | [7] |
| Moor County | | 10.1±0.3 | [15] |
| Millbillielle | 22.1±1.8 | 21.5±3 | [10] |
| | | 21.7±0.8 | [13] |
| Padvarnikai | 14.5±0.9 | 15.2±2.1 | [15] |
| Pasamonte | 7.1±0.6 | 7.7±0.5 | [15] |
| Pomozdino | | 22.5±0.7 | [15] |
| | | 8.6±0.6 | [16] |
| **Puerto Lapice** | **22.12±1.13** | | |
| Stannern | 38.9±2.8 | 36.4±2.1 | [14] |
| | | 41.0±1.3 | [16] |
| | | 36.6±0.7 | [13] |
| Serra de Mage | | 30.2±8.3 | [15] |
| Sioux County | | 21.5±1.4 | [15] |
| Vetluga | | 23.9±1.9 | [15] |

Table.1 $^{81}$Kr-Kr ages of eucrites determined with RIMSKI and available literature values. Due to high efficiency of the laser resonance ionization scheme the RIMSKI is capable of analyzing x100 smaller samples (1-4 mg).

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