## Noble Gases of 4 Aggregate Samples Allocated as the 4<sup>th</sup> AO Ryugu Samples Collected by the Hayabusa2 Spacecraft

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Hayabusa2 arrived at the C-type asteroid 162173 Ryugu in June 2018, successfully collected surface materials from two touchdown (TD) sites, and returned delivering ~5.4 g of regolith materials to Earth on December 6, 2020. Samples stored in Chamber A were collected from the 1<sup>st</sup> TD site on February 21, 2019 [1]. Samples in Chamber C were collected proximal to an artificial crater by the 2nd TD on July 11, 2019 [2], perhaps collecting some of ejecta materials from the crater.

Our studies on the Hayabusa2 samples are based on the measurement of those nuclides produced in asteroidal surface materials by both solar (SCR) and galactic cosmic rays (GCR). Cosmic-ray-produced (cosmogenic) nuclides are used to determine the duration and nature of the exposure of materials to energetic particles [3]. For Hayabusa2 samples, there are several specific questions we aim to address: (1) are the Chamber C samples ejecta deposits from the artificial crater, (2) if so, what is the original depth of each recovered sample grain in the Ryugu regolith, and (3) what is the average surface exposure time, mixing rate of regolith materials, and erosion/escape rate of materials from the Ryugu's surface? We are continuing to investigate and utilize cosmogenic radionuclides (<sup>10</sup>Be, <sup>26</sup>Al, <sup>36</sup>Cl, and <sup>41</sup>Ca) and noble gases in Ryugu samples.

We present here the noble gas isotopic compositions of 4 aggregate samples, A0490, A0492, C0372, and C0377 (Table 1), allocated as the 4<sup>th</sup> AO, and compare these with those of the 1<sup>st</sup> and 2<sup>nd</sup> AO samples [4]. Cosmogenic radionuclide measurements are in progress. Noble gases were measured with a modified-VG5400 mass spectrometer at the Korea Polar Research Institute (KOPRI). This noble gas system was used to measure the Itokawa samples, as well as other meteorites, at the University of Tokyo [5]; this system was moved to KOPRI in 2015. Noble gases were extracted from each sample using a miniature furnace specially designed for small samples. The furnace consists of a small Mo-crucible inserted within a Ta-tube. The Ta-tube is heated by applying current to a W-heater, which in turn is controlled by a computer program monitoring the heating temperature with a W-Re thermocouple. In this work, we aimed to measure noble gases, including those weakly trapped on by employing precise stepwise pyrolysis. The samples were installed in a glass-made sample holder that was set on the furnace, then kept at room temperature under ultra-high vacuum condition for about one month. To extract noble gases from weakly bound sites the first step was 200°C. The heating schedule then proceeded from 400–1300 in 100°C each step, 1500°C, and 1700°C (total 13 steps). A 300°C heating step was applied to C0372 (total 14 steps). The liberated noble gases from each sample at each heating step were purified and separated into 5 fractions, i.e., He, Ne, Ar, Kr, and Xe; isotope abundances and ratios were obtained for each fraction.

The concentrations of <sup>4</sup>He, <sup>20</sup>Ne, <sup>36</sup>Ar, <sup>84</sup>Kr, and <sup>132</sup>Xe, and the isotope ratios for He, Ne, and Ar are presented in Table 1. Data of two aggregate samples, 4 grain samples as well as Orgueil CI chondrite reported at the Hayabusa Symposium 2023 [4] are also included in the Table 1 for comparison. The aggregate samples A0490, A0492, C0372, and C0377 contained solar-wind implanted He and Ne. The concentrations of <sup>4</sup>He and <sup>20</sup>Ne are  $(8.9-39.5)\times10^{-4}$  and  $(1.2-3.6)\times10^{-5}$  cm<sup>3</sup>STP/g, respectively. These high concentrations are similar to those of 2 aggregate samples, A0221 and C0212, measured previously. These are likely trapped in fine grained particles present in the aggregate samples, similar to lunar fines and interplanetary dust particles (IDPs), which also have a large surface/volume ratio. The lowest temperature extraction at 200°C released relatively small concentrations of <sup>4</sup>He and <sup>20</sup>Ne compared to the bulk of the release occurring in the temperature range of 400–800°C. These samples were kept at room temperature and not preheated so the lack of noble gases at the low-temperature sites indicates that weakly bound solar gases residing on the upper surfaces of the Ryugu samples were lost on the Ryugu's surface, perhaps by the solar heating. In the 400 to 800°C heating steps He and Ne composed mainly of solar gases with <sup>3</sup>He/<sup>4</sup>He of (3–4)×10<sup>-4</sup> and <sup>20</sup>Ne/<sup>22</sup>Ne of 12.6–13.4 [6] were observed. Smaller amounts of He and Ne, composed of cosmogenic and primordial (Q, P3, and HL) components, were released at the higher heating temperatures ( $\geq$  900°C) [6]. <sup>36</sup>Ar showed sharp release peaks at 800°C, followed by a gentle peak for Q-Ar at 1200°C. Xe showed typical release profile for carbonaceous chondrites, i.e., a small peak for HL-Xe at 800 and 900°C, and a major peak for Q-Xe at 1200°C. The <sup>36</sup>Ar/<sup>132</sup>Xe versus <sup>84</sup>Kr/<sup>132</sup>Xe indicates that heavy noble gases are mostly composed of Q-component at high temperatures with a small contribution of solar gases released at lower temperatures from 300 to 800°C.

Isotopic ratios of <sup>3</sup>He/<sup>4</sup>He were highest,  $(5.7-6.8)\times10^{-4}$  at the lowest temperature of 200°C, then gradually decrease to solar He (4.6×10<sup>-4</sup>) and finally to primordial values of  $\leq 2\times10^{-4}$  for Q and/or P3, at 1300 and 1500°C. The total concentration of

cosmogenic <sup>3</sup>He for each sample is difficult to calculate because of the difficulty in evaluating the trapped <sup>3</sup>He/<sup>4</sup>He at each step. The almost negligible contribution of cosmogenic <sup>36,38</sup>Ar to the trapped Ar components, such as SW, Q etc. with variable <sup>38</sup>Ar/<sup>36</sup>Ar ratios, makes estimation of cosmogenic <sup>38</sup>Ar concentrations extremely difficult.

Accordingly, cosmogenic Ne is the most reliable stable noble gas indicator for estimation of the cosmic-ray exposure ages. We calculate the concentration of cosmogenic <sup>21</sup>Ne in the samples assuming that the trapped Ne is a mixture of solar-Ne [7] and Q- or air-Ne [6]. Shifts in the isotopic ratio off this trapped mixing line represent cosmogenic production. Assuming a superposition of cosmogenic and trapped components it is possible to calculate the cosmogenic <sup>21</sup>Ne amount for each temperature step; these are then summed for all heating steps from 200 to 1700°C. This method was applied to the aggregate samples A0221 and C0212 reported in 2023 [4]. Similar values of cosmogenic <sup>21</sup>Ne ranging (5.23–6.94)×10<sup>-9</sup> cm<sup>3</sup>STP/g, and cosmic-ray exposure ages in range from 5.7 to 10.7 Myr for all the aggregate samples were obtained (Table 1). The <sup>21</sup>Ne production rate (P<sub>21</sub>) was calculated for a body having a  $2\pi$  geometry with Ryugu's chemical composition using the MCNP Code System [8].

 ${}^{38}$ Ar/ ${}^{36}$ Ar was as low as  $\approx 0.182$  at 200–600°C, suggesting solar Ar, but abruptly increased to 0.186–0.190 at 700°C, followed by a sharp decrease to 0.184–0.185, and then gradually increased to 0.188 indicating Q-Ar in the range of 1000–1500°C. The  ${}^{130}$ Xe/ ${}^{132}$ Xe versus  ${}^{136}$ Xe/ ${}^{132}$ Xe shows that terrestrial atmospheric Xe adsorbed on the samples was released at the low heating temperatures of 200–400°C, then small amounts of solar-Xe at 500–700°C, HL-Xe at 800–900°C, and Q-Xe at  $\geq 1000°$ C. The sharp release of  ${}^{36,36}$ Ar with low  ${}^{38}$ Ar/ ${}^{36}$ Ar at 800°C noted above coincide with the release of HL-Xe, though  ${}^{38}$ Ar/ ${}^{36}$ Ar = 0.227 for HL-Ar [6] is much higher than the value of 0.184–0.185 observed in this work.

| Table 1. Concentrations and isotopic ratios of noble gases <sup>1)</sup> in 6 aggregate and 4 grain Ryugu samples, and Orgueil CI chondrite. |             |            |                 |                      |                  |                  |                   |                                  |             |             |           |  |  |                         |                               |           |
|--|-------------|------------|-----------------|----------------------|------------------|------------------|-------------------|----------------------------------|-------------|-------------|-----------|--|--|-------------------------|-------------------------------|-----------|
| Sample   | Mass        | Heating    | <sup>4</sup> He | <sup>20</sup> Ne     | <sup>36</sup> Ar | <sup>84</sup> Kr | <sup>132</sup> Xe | <sup>3</sup> He/ <sup>4</sup> He | 20. 22. 22. | 21          | 38 . 36 . | 40 . 36 .                              | <sup>21</sup> Ne <sub>cos</sub>            | P <sub>21</sub>         | T <sub>21</sub> <sup>6)</sup> | Domostra  |
|  | mg          | steps      |                 | n <sup>3</sup> STP/g | 5                |                  | 10-4              | 'Ne/ Ne                          | Ne/ Ne      | Ar/**Ar     | Ar/**Ar   | 10 <sup>-9</sup> cm <sup>3</sup> STP/g | 10 <sup>-9</sup> cm <sup>3</sup> STP/g/Myr | Myr                     | Rentarks                      |           |
| Aggregate  |             |            |                 |                      |                  |                  |                   |                                  |             |             |           |  |  |                         |                               |           |
| A0490  | 1.786       | 13         | 1473000         | 23700                | 3040             | 13.9             | 15.8              | 4.16                             | 12.83       | 0.0355      | 0.1857    | 0.66                                   | 6.60                                       | 1.00-1.15 <sup>3)</sup> | 5.7-6.6                       | This work |
| A0492  | 0.885       | 13         | 888000          | 12300                | 2290             | 17.4             | 19.0              | 4.35                             | 12.76       | 0.0385      | 0.1861    | 4.39                                   | 6.52                                       | 1.00-1.15 <sup>3)</sup> | 5.7-6.5                       | This work |
| C0372  | 2.406       | 14         | 1710000         | 16600                | 2420             | 13.4             | 15.4              | 4.23                             | 12.75       | 0.0358      | 0.1856    | 1.19                                   | 5.23                                       | 0.65-0.92 3)            | 5.7-8.0                       | This work |
| C0377  | 2.328       | 13         | 3950000         | 35700                | 4100             | 13.1             | 14.3              | 4.20                             | 13.09       | 0.0347      | 0.1848    | 0.66                                   | 6.94                                       | 0.65-0.92 3)            | 7.5–10.7                      | This work |
| A0221  | 1.661       | 5          | 1490000         | 20400                | 3430             | 16.9             | 18.5              | 3.54                             | 12.85       | 0.0362      | 0.1842    | 0.49                                   | 6.91                                       | 0.87-1.08 4)            | 6.4–7.9                       | [4]       |
| C0212  | 0.823       | 5          | 932000          | 15200                | 2720             | 16.1             | 18.7              | 3.89                             | 12.95       | 0.0371      | 0.1849    | 0.43                                   | 6.04                                       | 0.77-0.88 4)            | 6.9–7.8                       | [4]       |
| Grain  |             |            |                 |                      |                  |                  |                   |                                  |             |             |           |  |  |                         |                               |           |
| A0130  | 0.209       | 3          | 219000          | 1230                 | 1600             | 15.9             | 19.3              | 3.84                             | 10.54       | 0.0654      | 0.1858    | 1.79                                   | 4.41                                       | 1.00-1.12 4)            | 3.9-4.4                       | [4]       |
| C0012  | 2.218       | 5          | 97700           | 472                  | 1530             | 15.9             | 19.6              | 3.59                             | 8.27        | 0.0978      | 0.1861    | 0.42                                   | 4.06                                       | 0.77-0.88 4)            | 4.6-5.3                       | [4]       |
| C0162  | 0.189       | 3          | 118000          | 706                  | 1430             | 15.3             | 17.7              | 4.46                             | 8.68        | 0.0984      | 0.1867    | 1.34                                   | 5.83                                       | 0.72-0.92 4)            | 6.3-8.1                       | [4]       |
| C0182  | 0.277       | 3          | 93800           | 432                  | 1370             | 20.5             | 17.0              | 3.80                             | 8.09        | 0.1280      | 0.1876    | 1.18                                   | 5.45                                       | 0.65-0.72 4)            | 7.6-8.4                       | [4]       |
| Orgueil(CI)  | 1.721       | 3          | 672000          | 1130                 | 1260             | 13.0             | 15.7              | 3.51                             | 10.08       | 0.1099      | 0.1856    | 2.37                                   | 9.37                                       | 2.22 5)                 | 4.22                          | [4]       |
| <sup>1)</sup> Isotopic ratios of Kr and Xe are not presented.  |             |            |                 |                      |                  |                  |                   |                                  |             |             |           |  |  |                         |                               |           |
| <sup>2)</sup> Heating temperatures are from 200 to 1700°C.   |             |            |                 |                      |                  |                  |                   |                                  |             |             |           |  |  |                         |                               |           |
| 3) Depths of   | A0490 an    | d 0492 are | assumed         | to be $> 8$          | 80 g/cm          | $n^2$ and t      | hose of           | C0372 and                        | 0377 are a  | ssumed to b | e 100-180 | g/cm <sup>2</sup> .                    |  |                         |                               |           |
| 4) Each dept   | h was esti  | mated bas  | ed on preli     | ninary m             | easure           | ments            | of cosm           | ogenic radi                      | onuclides [ | 9].         |           |  |  |                         |                               |           |
| <sup>5)</sup> Prodction  | rate in 4π- | -geometry  | for CI cho      | ndrites [            | 10].             |                  |                   |                                  |             |             |           |  |  |                         |                               |           |
| 6 21   |             | 1          |                 |                      |                  |                  |                   | $\mathbf{D} \rightarrow (10)$    | 1 21        |             |           | 0()                                    |  |                         |                               |           |

 $^{6) 21}$ Ne exposure ages do not include uncertainties in the production rate (P<sub>21</sub>) (~±10 %) and  $^{21}$ Ne measurements (±5 %).

*Acknowledgment:* We thank Hayabusa2 project team, especially the curation members. We also thank MNHN-Paris for providing Orgueil sample.

## References

- [1] Arakawa M. et al. 2020. Science 368, 67-71
- [2] Tsuda Y. et al. 2020. Acta Astronautica 171, 42-54
- [3] Reedy R. C. et al. 1983. Annu. Rev. Nucl. Part. Sci. 33, 505–537.
- [4] Nagao K. et al. 2023. Abstract S31-02\_Nagao.pdf, 2023 Hayabusa Symp.
- [5] Nagao K. et al. 2011. Science 26, 1128–1131.
- [6] Ott U. 2002. Noble gases in Geochem. and Cosmochem., Rev. in Mineral. Geochem. 47, 71-100.
- [7] Heber V. S. et al. 2009 Geochim. Cosmochim. Acta 73, 7414–7432.
- [8] Masarik J. and Reedy R. C. (1994) Geochim. Cosmochim. Acta. 58, 5307-5317.
- [9] Nishiizumi K. et al. 2023. Abstract S31-01\_Nishiizumi.pdf, 2023 Hayabusa Symp.
- [10] Eugster O. 1988. Earth Planet Sci. Lett. 52, 1649-1662.