SPACE SCIENCES

Ryugu's nucleosynthetic heritage from the outskirts of the Solar System

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Little is known about the origin of the spectral diversity of asteroids and what it says about conditions in the protoplanetary disk. Here, we show that samples returned from Cb-type asteroid Ryugu have Fe isotopic anomalies indistinguishable from Ivuna-type (CI) chondrites, which are distinct from all other carbonaceous chondrites. Iron isotopes, therefore, demonstrate that Ryugu and CI chondrites formed in a reservoir that was different from the source regions of other carbonaceous asteroids. Growth and migration of the giant planets destabilized nearby planetesimals and ejected some inward to be implanted into the Main Belt. In this framework, most carbonaceous chondrites may have originated from regions around the birthplaces of Jupiter and Saturn, while the distinct isotopic composition of CI chondrites and Ryugu may reflect their formation further away in the disk, owing their presence in the inner Solar System to excitation by Uranus and Neptune.

INTRODUCTION

Main Belt asteroids show great compositional diversity (*1*), ranging from metallic objects that are remnants of differentiated planetesimals (*2*) to carbon-rich objects with comet-like dust-ejection activity (*3*). The original formation locations of these diverse objects are unknown. Meteorites are remnants of planetesimals and protoplanets that formed at various heliocentric distances within the first few million years after the birth of the Solar System. They, therefore, provide invaluable insights into the early evolution of the Solar System and the building blocks of the terrestrial planets. Most meteorites are fragments of Main Belt asteroids, but direct asteroid-meteorite links are scarce (*1*). Establishing such links is important as it provides clues on the relationship and formation locations of meteorite parent bodies, asteroids, and other small bodies in the Solar System. Cb-type asteroid (162173) Ryugu is a near-Earth object (NEO) that most likely originated from the inner Main Belt (*4*). Chemical and mineralogical analyses of Ryugu samples returned to Earth by Japan Aerospace Exploration Agency's (JAXA) Hayabusa2 mission (*5*) show that they share chemical and mineralogical characteristics with Ivuna-type carbonaceous (CI) chondrites (*6*). The latter is the only group of meteorites containing most nonvolatile elements in proportions nearly equal to those measured in the solar photosphere (*7*). The physical and chemical similarities of Ryugu and CI chondrites are, however, not diagnostic of a shared heritage because the low-temperature conditions required to explain their solar-like chemical compositions could have been widespread in the outer Solar System disk.

To better constrain Ryugu's nucleosynthetic heritage, we measured the Fe isotopic compositions of four Ryugu samples collected during the first and second touchdown (*5*), 11 different carbonaceous chondrites from five different groups (CI, CM, CV, CO, and CR), and two ungrouped chondrites. Materials formed in the Solar System display variations in the isotopic compositions of some elements that stem from the heterogeneous distribution and processing of highly anomalous presolar materials in the protosolar nebula (*8*–*11*). Such anomalies cannot easily be modified by physicochemical processes on planetary bodies and, therefore, provide lasting isotopic fingerprints of the regions where planetary bodies formed (*12*–*14*). For several elements, meteorites display a dichotomy in their isotopic anomalies between non-carbonaceous (NC) and carbonaceous (CC) meteorite groups (*13*). The origin of this dichotomy could have involved the physical separation between inner and outer Solar System reservoirs by Jupiter (*13*, *15*, *16*) and planetesimal formation at distinct locations in an evolving protoplanetary disk (*17*–*21*). Although isotopic analyses of Ti and Cr show that Ryugu's building blocks formed in the CC reservoir and support a possible kinship between Ryugu and CI chondrites (*6*), an unambiguous genetic link to a specific carbonaceous chondrite group could not be established because the Ti and Cr isotopic anomalies of Ryugu overlap with several carbonaceous chondrites and achondrites (Fig. 1A). Carbonaceous chondrites also display mass-independent variations in O isotopes that correlate with Ti and Cr isotopic anomalies (*13*). Ryugu and CI chondrites have similar O isotopic anomalies and represent an endmember composition for the CC cluster in O-Cr and O-Ti spaces (Fig. 1, B and C) (*6*). Those results support the view that CI chondrites and Ryugu formed within the CC reservoir, but because of correlations between O, Cr, and Ti anomalies (Fig. 1), there is substantial redundancy in evidence presented thus far and further work is needed to better understand the isotopic architecture of the outer Solar System.

The Fe isotopic composition of CI chondrites (*22*) is clearly distinct from all other carbonaceous meteorites (*22*–*24*), where all CC meteorites except CI chondrites show significant excesses in ^{54}Fe. Iron isotopes, therefore, provide a diagnostic tool to evaluate if Ryugu has the same distinct nucleosynthetic heritage as CI chondrites.

RESULTS

Ryugu samples and all carbonaceous chondrites display limited mass-dependent isotopic variation relative to the terrestrial standard IRMM-524a (Table 1), meaning that all isotopic anomalies reported here are real and not artifacts from the internal normalization scheme used to correct natural and instrumental isotopic fractionation (*14*). Mass-independent Fe isotopic analyses confirm that most carbonaceous chondrites display variable excesses of ~+15 to ~+40 in μ^{54}Fe but no resolvable variations in μ^{58}Fe (see Table 1 for a definition of the μ-notation) (Table 1 and Fig. 2). By contrast, all three CI chondrites analyzed in this study are distinct from other carbonaceous chondrites, defining an average μ^{54}Fe = +3 ± 2, consistent with previous measurements (Fig. 2 and fig. S1) (*22*). The four Ryugu samples (A0106 and A0106-A0107 collected during the first touchdown from the surface; C0107 and C0108 collected during the second touchdown and possibly sampling material from the subsurface) define an average μ^{54}Fe value of +1 ± 4, which is indistinguishable from the composition of CI chondrites but distinct from all other carbonaceous chondrites (Fig. 2). The μ^{54}Fe difference of Ryugu and CI chondrites compared to all other carbonaceous chondrites cannot be due to prolonged exposure to cosmic rays in space because (i) CI chondrites have low cosmic ray exposure ages (*25*) and (ii) cosmogenic effects would induce a positive shift in μ^{54}Fe that would correlate with a negative shift in μ^{58}Fe (*24*, *26*), which is not observed (Table 1 and fig. S2). Thus, the distinct μ^{54}Fe values of CI chondrites and Ryugu represent the nucleosynthetic heritage of their formation reservoir in the protosolar nebula. Examination of μ^{54}Fe-μ^{50}Ti (Fig. 3) and μ^{54}Fe-μ^{54}Cr (fig. S3) isotopic anomalies shows that Ryugu and CI chondrites form a compositional cluster that is distinct from the NC and CC fields defined by other meteorites. Thus, whereas the isotopic anomalies of Ti, Cr, and O in CI chondrites (*10*, *11*, *27*–*31*) and Ryugu (*6*) tie them to the CC reservoir, the Fe isotopic data reveal that CI chondrites and Ryugu formed in a reservoir that is rarely sampled by meteorites.

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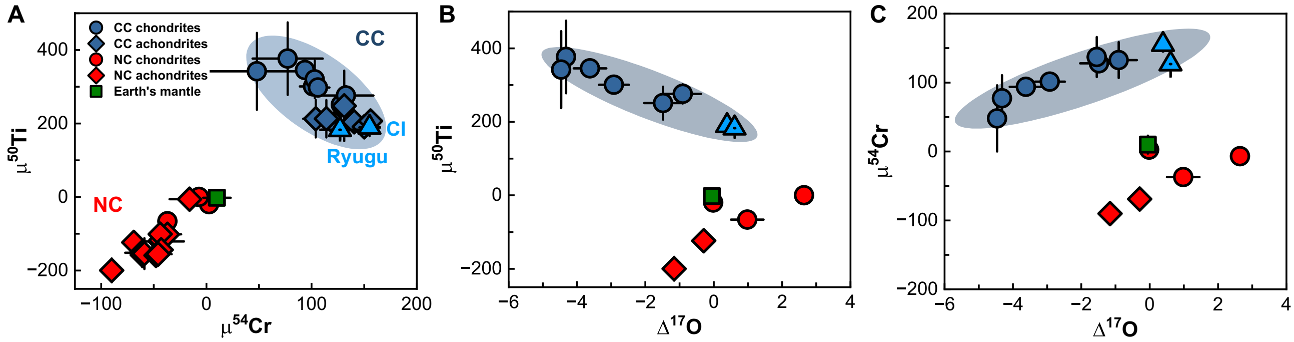

Fig. 1. Previously published isotopic anomalies of Ti, Cr, and O in Ryugu and other Solar System materials. Plots of μ^{50}Ti versus μ^{54}Cr (**A**), μ^{50}Ti versus Δ^{17}O (**B**), and μ^{54}Cr versus Δ^{17}O (**C**). In these diagrams, Ryugu and CI represent an endmember to the CC array. Data for Ryugu are from Yokoyama *et al.* (*6*). The average Ti and Cr isotopic compositions of NC and CC meteorite groups and Earth's mantle are from the data compilation of Burkhardt *et al.* (*64*) and O isotopic data from the compilation of Dauphas (*65*). Uncertainties for individual groups are the 95% confidence interval of the mean. If not visible, error bars are smaller than symbols.

DISCUSSION

Recent models tie the distinct isotopic characteristics of meteorites to planetesimal formation at different locations and/or at different times in an isotopically heterogeneous protoplanetary disk (*9*, *15*, *17*, *18*, *20*, *21*). The main driver of the isotopic heterogeneity could be a change in the composition of infalling material during collapse of the parental molecular cloud core of the Solar System (*14*, *32*, *33*) or, alternatively, unmixing of presolar carriers by disk processes (*9*, *10*, *19*). Irrespective of the origin of the heterogeneity, it requires the existence of large-scale isotopic heterogeneities throughout the disk. The Fe isotopic compositions of CI chondrites and Ryugu are similar to Earth's mantle and NC meteorites, while isotopic anomalies in other elements are similar to CC meteorites (Figs. 1 and 3). This suggests that CI chondrites and Ryugu derive from an isotopic reservoir that has a distinctive nucleosynthetic heritage from NC and CC meteorites. While the Fe isotopic characteristics of CI chondrites could be explained by an inner Solar System origin, the isotopic anomalies of Ti, Cr, and Mo clearly tie CI chondrites to the CC reservoir and the outer Solar System (*10*–*12*, *16*, *27*). The unfractionated chemical compositions and volatile element-rich nature of CI chondrites and Ryugu support the view that they formed beyond the snow line, mostly from material that experienced minimal thermal processing. The heavy hydrogen and nitrogen isotopic compositions of Ryugu are also consistent with an outer Solar System origin (*34*). The findings that CI chondrites and Ryugu share the same nucleosynthetic heritage (Fig. 3) and have close mineralogical, chemical, and isotopic characteristics (*6*, *34*), therefore, suggest that these objects formed contemporaneously and were co-located in the same outer Solar System reservoir. It is even possible, although not required by the data, that CI chondrites and the NEO Ryugu originally derived from the same precursor object, which was fragmented by collision during its residence in the inner Main Belt (*4*, *35*).

The distinct isotopic heritage of CI chondrites and Ryugu is unlikely to reflect a temporal change in the isotopic composition of the CC reservoir because (i) CI chondrites and Ryugu are distinct from other carbonaceous chondrites and intermediate compositions are missing (Fig. 2), and (ii) CI chondrites have similar inferred accretion ages as other carbonaceous chondrites [~2.5 to 4 million years (Myr) after condensation of refractory inclusions] (*36*). Thus, the distinctive isotopic heritage of CI chondrites and Ryugu is most likely caused by spatial separation of their source region from the CC reservoir or, as discussed below, bias in the implantation of planetesimals from distinct heliocentric distances into the Main Belt.

Cb-type asteroids like Ryugu represent ~10 to 20% of all C-type asteroids (*37*, *38*), suggesting that a substantial portion of Main Belt asteroids formed in the same outer Solar System reservoir where CI chondrites and Ryugu formed. Planetesimals from a large range of heliocentric distances could have been implanted into the inner Solar System during the growth and migration of the giant planets within the protoplanetary disk, before dissipation of nebular gas (*39*). Planetesimals from the outer Solar System could have also been implanted during subsequent instabilities in the orbits of Uranus and Neptune after dissipation of nebular gas (*40*–*43*), but the efficiency of this process is low and early implantation of C-type asteroids in the Main Belt while nebular gas was still present is more likely. The planetesimals scattered by the growth and migration of the giant planets were originally on highly eccentric orbits, but they experienced a strong headwind from nebular gas that circularized their orbits, leading to their trapping in the Main Belt. Simulations of the growth and migration of the giant planets show that most outer Solar System planetesimals implanted in the Main Belt originated from the formation region of the gas giant planets Jupiter and Saturn [e.g., ~4 to 12 astronomical units (AU)], but some could have come from further away in the formation region of ice giant planets Uranus and Neptune (e.g., 13 to 25 AU) (*39*). The dynamical process of orbital excitation and circularization introduces strong biases in the original orbital radii of the planetesimals that are eventually implanted in the Main Belt (fig. S4). In that context, most CC meteorites could have come from the birth region of Jupiter and Saturn, while the distinctive Fe isotopic heritage (Fig. 3) and primitive chemical characteristics of CI chondrites and Ryugu (*6*) could be explained if they were implanted into the Main Belt from a reservoir that was located further outside, possibly in the vicinity of the birthplaces of Uranus and Neptune (Fig. 4 and fig. S4). If correct, CI chondrites and Ryugu would possibly share a common heritage with Oort cloud comets (*44*, *45*).

A common source region for the parent bodies of CI chondrites/Ryugu and Oort cloud comets would need to be reconciled with their present-day distinct chemical and physical characteristics (*6*, *34*, *46*). Deuterium/hydrogen (D/H) ratios of water in carbonaceous chondrites

Table 1. Iron isotopic compositions of Ryugu samples and carbonaceous chondrites. Calculated mass-independent (μ^{54}Fe and μ^{58}Fe) and mass-dependent (δ^{56}Fe) Fe isotopic compositions in Ryugu samples, carbonaceous chondrites, and geostandards. Uncertainties of individual samples are 95% confidence intervals of the mean of *N* standard-sample-standard bracketing analyses. Uncertainties on group averages are the 95% confidence intervals of the mean. The isotopic anomalies are calculated by internal normalization to ^{57}Fe/^{56}Fe = 0.023095 and expressed in μ-notation (*22, 24*) defined as the parts-per-million deviation of the internally normalized ^{5x}Fe/^{56}Fe ratio in the sample relative to bracketing measurements of IRMM-524a. The mass-dependent Fe isotopic variations are calculated by sample-standard bracketing and given as δ-notation defined parts-per-thousand deviation of the ^{56}Fe/^{54}Fe ratio of the sample relative to the IRMM-524a standard solution.

Sample	*N*	μ^{54}Fe	μ^{58}Fe	δ^{56}Fe
Ryugu				
A0106	30	−2 ± 5	14 ± 14	−0.01 ± 0.02
A0106-A0107	30	2 ± 5	10 ± 10	−0.01 ± 0.02
C0107	25	4 ± 4	15 ± 17	−0.02 ± 0.02
C0108	25	2 ± 5	11 ± 7	0.01 ± 0.02
Average Ryugu		1 ± 4	13 ± 4	−0.01 ± 0.02
CI chondrites				
Orgueil-1	30	2 ± 5	12 ± 8	−0.02 ± 0.02
Orgueil-2	30	3 ± 5	2 ± 11	0.01 ± 0.02
Orgueil-3	14	4 ± 9	3 ± 12	0.00 ± 0.02
Orgueil-4	30	3 ± 5	12 ± 8	0.06 ± 0.02
Ivuna	30	1 ± 4	16 ± 11	0.00 ± 0.02
Alais	28	6 ± 6	17 ± 12	−0.02 ± 0.02
Average CI		3 ± 2	10 ± 6	0.00 ± 0.02
CM chondrites				
Murchison-1	14	23 ± 5	6 ± 7	0.00 ± 0.02
Murchison-2	15	18 ± 6	2 ± 9	0.01 ± 0.02
Mighei	14	27 ± 8	7 ± 9	−0.01 ± 0.02
Average CM		24 ± 7	6 ± 11	0.00 ± 0.02
CV chondrites				
Allende-1	15	29 ± 4	2 ± 9	−0.02 ± 0.02
Allende-2	15	32 ± 6	2 ± 9	−0.05 ± 0.02
Allende-3	15	21 ± 7	3 ± 12	0.08 ± 0.02
Vigarano	15	29 ± 5	−7 ± 13	0.02 ± 0.02
Average CV		28 ± 6	0 ± 6	0.01 ± 0.08
CO chondrites				
Ornans	15	21 ± 5	0 ± 8	−0.03 ± 0.02
Lance	15	18 ± 5	1 ± 10	0.01 ± 0.02
CR chondrites				
Acfer 139	14	38 ± 8	9 ± 11	−0.11 ± 0.02
GRA 06100	15	39 ± 5	9 ± 6	0.00 ± 0.02
Ungrouped chondrites				
Tagish Lake-1	15	18 ± 7	2 ± 14	−0.03 ± 0.02
Tagish Lake-2	15	15 ± 6	2 ± 22	0.05 ± 0.02
Tarda	30	24 ± 8	3 ± 12	−0.01 ± 0.02
Geostandards				
BHVO-2-1	15	1 ± 7	−9 ± 8	0.09 ± 0.02
BHVO-2-2	15	1 ± 7	−1 ± 11	0.06 ± 0.02
AGV-2	30	2 ± 4	1 ± 6	0.08 ± 0.02
Average geostandards		1 ± 1	−3 ± 14	0.09 ± 0.06

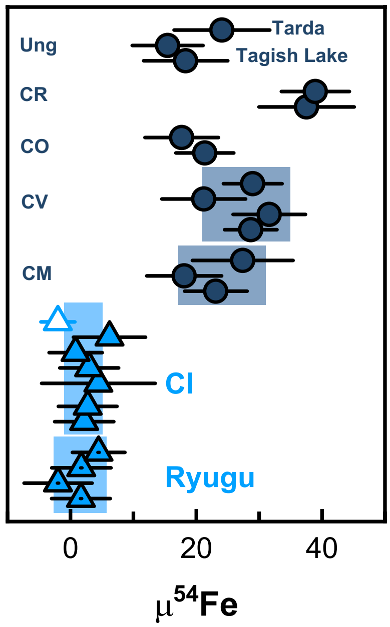

Fig. 2. Fe isotopic anomalies of returned samples from Cb-type asteroid (162173) Ryugu and carbonaceous chondrites (Table 1). Ryugu samples and CI chondrites have identical μ^{54}Fe values, which are distinct from all other carbonaceous chondrite groups [CM, CV, CO, CR, and Ung (ungrouped)]. The open triangle is the CI chondrite average from Schiller *et al.* (*22*).

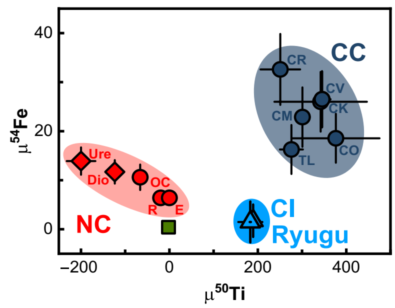

Fig. 3. Isotopic anomalies of Fe and Ti in Solar System materials. Ryugu samples and CI chondrites have identical μ^{54}Fe and μ^{50}Ti values that are distinct from other meteorites. Red circles correspond to NC chondrite groups (E, enstatite; R, rumuruti; OC, ordinary chondrites), red diamonds to NC achondrites (Ure, ureilites; Dio, diogenites), and blue circles to CC chondrite groups (TL, Tagish Lake). The green square is Earth's mantle. The average composition of Ryugu and CI chondrites is shown as triangles. Average compositions of meteorite groups and Earth's mantle are calculated using data from this study (Table 1) and the data compilation of Burkhardt *et al.* (*64*) (table S1). If not visible, error bars are smaller than symbols.

(*47*) and Ryugu (*34*) are lower than those of Oort cloud comets and overlap partially with Jupiter-family comets (*48*). Simulations of ice transport in the nebula predict a spatially and temporally complex evolution of water D/H ratio in the nebula (*49*). Furthermore, the water D/H ratio of active comets and asteroids might have been modified by water sublimation (*50*, *51*) and water-rock reactions (*52*) during their lifetimes. Therefore, the present-day water D/H ratio of ice in comets and rock-bound water in carbonaceous chondrites provides little insights into formation locations. CI chondrites and Ryugu show evidence for extensive aqueous alteration as late as ~5 Myr after the birth of the Solar System (*6*, *53*, *54*). Water responsible for this aqueous alteration would have presumably been accreted as ice, with melting caused by decay of ^{26}Al. Such melting could have been hampered in ice-rich comets if much of radioactive heat from ^{26}Al was consumed by ice sublimation rather than melting (*55*). While most dust grains captured in the coma of comet 81P/Wild2 were anhydrous (*46*), Berger *et al.* (*56*) found evidence for low-temperature aqueous activity in 81P/Wild2 under conditions akin to those inferred for CI chondrites. The Deep Impact mission also found signatures of carbonates, phyllosilicates, sulfides, water gas, and ice in the ejecta of comet 9P/Tempel, which is consistent with extensive aqueous alteration (*57*). While these observations support the presence of aqueous activity on extant comets, a comparison between icy planetesimals that were scattered inward and outward by the growth and migration of the giant planets is difficult because they would have experienced very different thermal histories. The planetesimals scattered inward would have been put on eccentric orbits with low perihelion, well inside the snow line, where ice could have been sublimated and the more volatile compounds could have been lost. For a ~100-km planetesimal, the time scale for damping eccentricity through gas interaction is on the order of several tens of thousands of years, which is in the order of the expected lifetime of short-period comets (*58*). Thus, the rock/ice ratio of ice-rich planetesimals formed around Uranus and Neptune may have increased substantially by the time these planetesimals were implanted in the Main Belt. Such processing could also have affected other characteristics of the icy planetesimals, notably their inventories of organics, mineralogical compositions, and physical properties. Further transformations would have taken place due to collisions during residence in the Main Belt (*35*, *59*). Consequently, although the Ryugu asteroid and Oort cloud comets may have been born in the same region of the protoplanetary disk, they would have rapidly diverged in their chemical evolution after being scattered inward and outward by the ice giant planets.

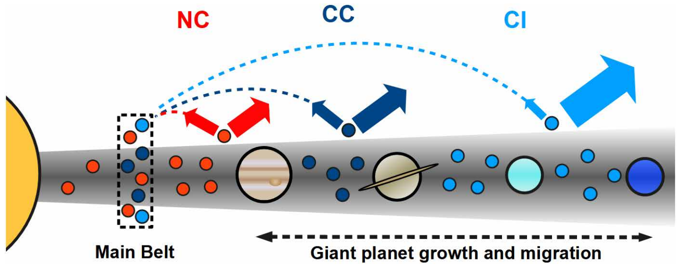

Fig. 4. Schematic of the possible source region of Cb-type asteroids and CI chondrites. Planetesimals formed in different regions of the protoplanetary disk. Volatile-poor planetesimals (red circles) formed in the inner region, while volatile-rich planetesimals (blue circles) formed beyond Jupiter's orbit. The growth and migration of the gas and ice giant planets implanted some of the planetesimals into the Main Belt (small arrows), while the majority of planetesimals were transported outward or ejected from the disk (large arrows) (*39*). A plausible explanation for the distinct Fe nucleosynthetic heritage and primitive chemical composition of CI chondrites and Ryugu is that they were implanted in the Main Belt by excitation from Uranus and Neptune (filled bright blue circles), while other CCs formed in more internal regions near Jupiter and Saturn (filled dark blue circles) (fig. S4). The icy planetesimals that were formed around Uranus and Neptune and were ejected outward went to populate the Oort cloud (*44*, *45*). CI chondrites and Ryugu may thus share some parentage with long-period comets. Such a scenario could explain the trichotomy between NC, CC, and CI for nucleosynthetic anomalies (Fig. 3).

In our model, Cb-type asteroids formed in a reservoir that was located at the outskirts of the planetary accretion region and were possibly implanted into the Main Belt due to excitation by Uranus and Neptune (Fig. 4). The outer extent of this isotopic reservoir is unknown. The main source region of Kuiper Belt objects (KBOs) was likely the trans-Neptunian disk (>20 to 25 AU) (*60*). These KBOs are out of reach for sample return missions, but possible trans-Neptunian objects 203 Pompeja and 269 Justitia were recently found in the Main Belt (*61*). Measuring the Fe and Ti isotopic compositions of these objects would provide important new insights into the isotopic architecture of the early Solar System and help evaluate the extent of the CI reservoir.

MATERIALS AND METHODS

Samples, preparation, and chemical purification

The Hayabusa2 spacecraft returned a total of ~5 g of material from Cb-type (162173) asteroid Ryugu. Surface samples were collected in Chamber A (~3 g) during the first touchdown and subsurface samples in Chamber C (~2 g) during the second touchdown (*5*, *62*). Two subsamples from Chamber A (A0106 and A0106-A0107) and two from Chamber C (C0107 and C0108) were digested for isotopic analysis (Table 1). Sample masses were 14.2, 23.88, 14.20, 12.90, and 22.24 mg, respectively. For samples A0106 and C0107, soluble organic matter was separated before digestion by acids. Sample A0108 was analyzed by x-ray fluorescence before digestion. Approximately 20 to 25 mg of six carbonaceous chondrite powders (Table 1: Orgueil-4, Alais, Murchison-2, Allende-3, Tagish Lake-2, and Tarda) were digested and processed together with the Hayabusa2 samples. Sample digestion for these samples was conducted at the Tokyo Institute of Technology. Powder aliquots were digested using mixtures of HF-HNO_3-HCl-H_2O_2 on hot plate and under ultrasonic agitation. Approximately 80% of the solutions was taken for sequential separation of several elements for isotopic analysis. We measured 20 additional carbonaceous chondrite samples and three terrestrial geostandards (Table 1) to provide some context for interpreting Ryugu's results. Some samples were digested for this study, while others are elution cuts from previous studies focused on elements other than Fe. The masses digested, original masses homogenized, and details on the processing history for each sample are summarized in table S2. The first step in the chemical processing of Fe in the four Ryugu samples and six accompanying carbonaceous chondrites was conducted at the Tokyo Institute of Technology and involved (i) separation of major elements, including Fe, from Zn, Pb, and highly siderophile elements using anion exchange chromatography (AG-1X8); (ii) separation of Fe and U from remaining major elements using AG-1X8; and (iii) separation of U from Fe using Eichrom UTEVA resin. A 20% aliquot of the Fe solution was then purified from remaining traces of Cr, Ni, Co, and Cu at the University of Chicago using an established protocol (*24*). Approximately 0.5 to 1 mg of Fe were loaded in 0.25 ml of 10 M HCl onto 10.5-cm-long perfluoralkoxy (PFA) teflon columns (0.62 cm inner diameter) filled with 3-ml precleaned AG1-X8 (200 to 400 mesh) anion resin. Matrix elements were eluted in 5 ml of 10 M HCl. Other possible contaminants (e.g., Cu and Cr) were eluted from the resin using 30 ml of 4 M HCl. Iron was eluted using 9 ml of 0.4 M HCl. The samples not previously processed at the Tokyo Institute of Technology were purified using the same procedure, but the elution was repeated using new resin. The overall Fe yield is >99%, and the procedural blank is negligible (~70 ng) compared to the amount of Fe purified for each sample (0.5 to 1 mg Fe). Interfering elements Cr (^{54}Cr on ^{54}Fe) and Ni (^{58}Ni on ^{58}Fe) were present at low enough levels (Cr/Fe $\leq 9 \times 10^{-6}$ and Ni/Fe $\leq 2 \times 10^{-5}$) to not affect the accuracy of the analyses (*24*).

Iron isotopic measurements

High-precision Fe isotopic compositions were measured following the protocol used for analysis of Fe isotopic anomalies in iron meteorites (*24*). Measurements were conducted with a Thermo Fisher Scientific Neptune multicollector inductively coupled plasma mass spectrometer (MC-ICP-MS) at the Origins Laboratory of the University of Chicago. Measurements were made on the flat-topped peak shoulder in medium-resolution mode (*63*). Ion beams of $^{54}Fe^+$, $^{56}Fe^+$, $^{57}Fe^+$, and $^{58}Fe^+$ were analyzed statically on Faraday collectors. All isotopes were measured using 10^{11}-ohm amplifiers, except for high-abundance $^{56}Fe^+$, which was measured using a 10^{10}-ohm amplifier. Isobaric interferences from $^{54}Cr^+$ and $^{58}Ni^+$ were determined simultaneously by monitoring $^{53}Cr^+$ and $^{60}Ni^+$ using 10^{12}-ohm amplifiers. The purified Fe solutions (10 μg/g in 0.3 M HNO_3) were introduced into the MC-ICP-MS using an Elemental Scientific Inc. (ESI) PFA nebulizer with an uptake rate of ~100 μl/min combined with a cyclonic glass spray chamber. Iron isotopic composition was measured at a typical $^{56}Fe^+$ ion signal intensity of 1.3 nA. Each measurement consisted of 50 cycles of 8.369 s each. Sample analyses were bracketed by measurements of IRMM-524a in a standard-sample-standard scheme. On peak zero, intensities from a blank solution measured at the start of each sequence were subtracted from the measurements. A washout time of 210 s was used between each analysis. The Fe concentrations of the samples and standards were matched to within ≤2%, which is required for accurate and precise Fe isotopic analysis (*24*).

SUPPLEMENTARY MATERIALS

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