Electronic supplementary information

For

Chemical state and isotope ratio analysis of individual uranium particles by a combination of micro-Raman spectroscopy and secondary ion mass spectrometry

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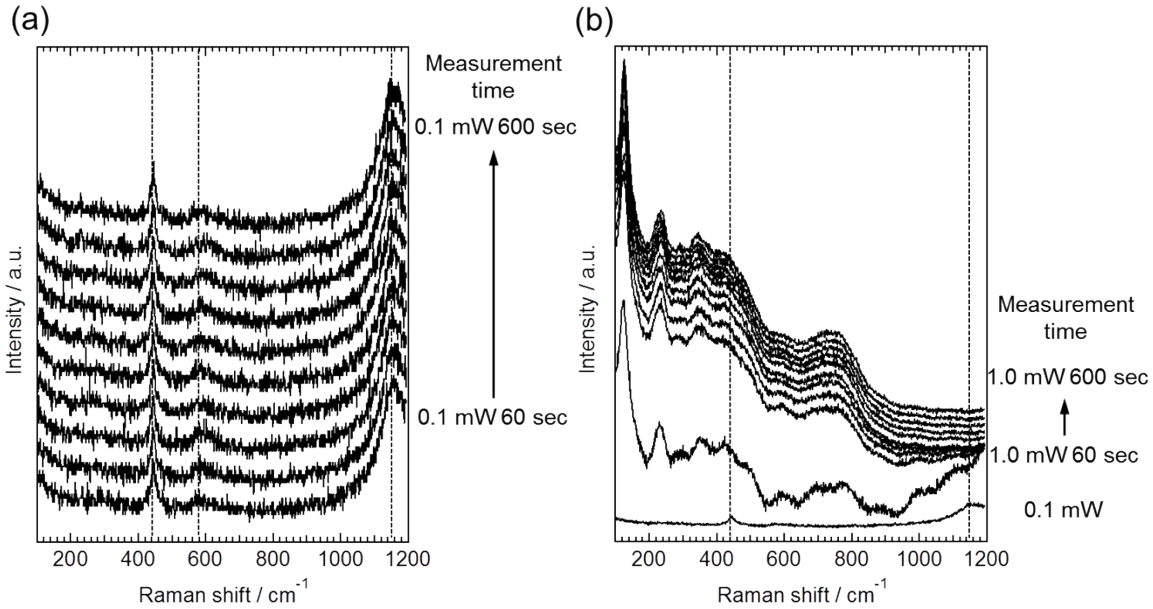

17 Figure S1. Changes in the MRS spectrum of a UO_2 particle with a diameter of 3 μm with
18 measurement time. The laser powers of (a) 0.1 mW and (b) 1.0 mW were used for the
19 measurement.

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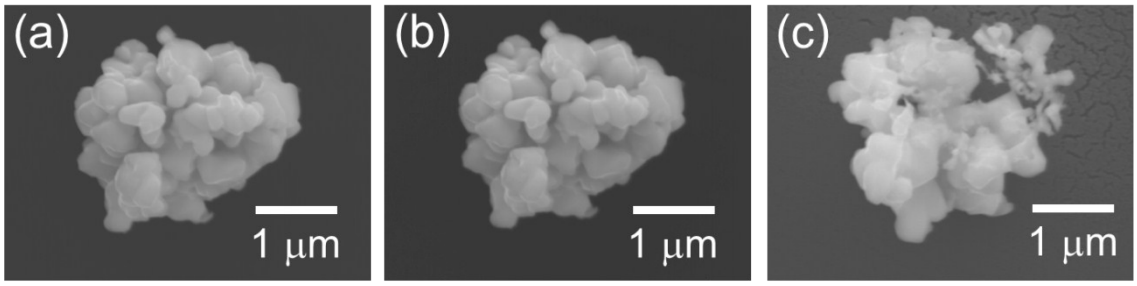

21 Figure S2. The scanning electron images of a uranium particle (UO_2) with a diameter of 3 μm
22 (a) before and after MRS measurements with the laser powers of (b) 0.1 mW and (c) 1.0 mW.

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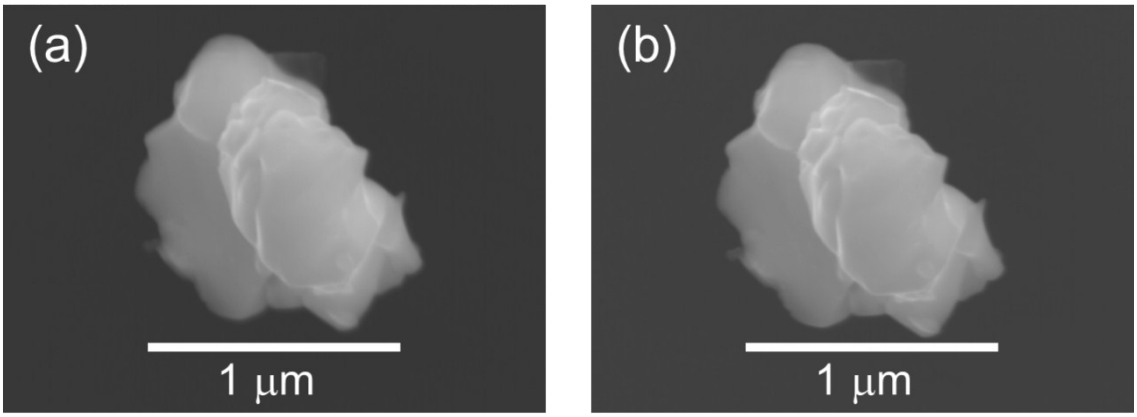

Figure S3. Scanning electron images of a uranium particle (UO_2) with a diameter of 1 mm (a) before and (b) after MRS measurement with a 0.1 mW laser power.

Table. S1 Particle sizes and $^{234}U/^{238}U$ and $^{235}U/^{238}U$ isotope ratios of individual particles in a uranium dioxide sample with natural composition. The errors represent the expanded uncertainties with a coverage factor of $k = 2$.

Particle No.	Size (μm)	$^{234}U/^{238}U$ isotope ratio / $\times 10^{-5}$	$^{235}U/^{238}U$ isotope ratio/ $\times 10^{-3}$
1	5.1	5.1 ± 0.4	7.32 ± 0.07
2	4.1	5.1 ± 0.4	7.23 ± 0.06
3	3.4	5.1 ± 0.2	7.19 ± 0.05
4	1.9	5.1 ± 0.4	7.24 ± 0.07
5	2.2	5.5 ± 0.3	7.21 ± 0.06
6	3.9	4.9 ± 0.4	7.24 ± 0.05
7	3.0	5.4 ± 0.5	7.30 ± 0.07
8	4.2	5.7 ± 0.4	7.24 ± 0.04
9	1.1	5.4 ± 0.5	7.18 ± 0.07
10	4.1	5.2 ± 0.3	7.25 ± 0.07
Average	-	5.2 ± 0.5	7.24 ± 0.09
Literature[*1]	-	5.5 ± 1.0	7.25 ± 0.02

[*1]P. De Bi`evre, P.D.P. Taylor, *Int. J. Mass Spectrom. Ion Processes* **1993**, 123, 149–166.