Electronic Supplementary Material (ESI) for Journal of Analytical Atomic Spectrometry.

ESI, Table 1. Amount of heavy element (U) and burn-up monitor (Nd) taken for the preparation target pellet (Set-I). The source of the starting materials are nuclear grade natural UO_2 and Nd_2O_3 (99.9% pure) obtained from M/s Indian Rare Earths, Mumbai, India.

S. No.	**Sample ID**	**Calculated burn-up (at.%)**	**UO_2 (g)**	**Nd_2O_3 (g)**	**Atom ratio of $^{238}U/(^{145}Nd+^{146}Nd)$**
1.	S-I-1	0.79	3.5311	0.0038	2.3E+03
2.	S-I-2	3.58	3.5337	0.0178	4.8E+02
3.	S-I-3	6.83	3.5386	0.0352	2.4E+02
4.	S-I-4	9.80	3.5338	0.0521	1.6E+02
5.	S-I-5	12.54	3.5377	0.0688	1.2E+02
6.	S-I-6	15.44	3.5318	0.0875	9.8E+01

ESI, Table 2. Amount of heavy element (U), burn-up monitor (Nd), and other major fission products (Cs, Zr and Sr) taken for the preparation target pellet (Set-II). The source of the starting materials are nuclear grade natural UO_2; Nd_2O_3 (99.9% pure) obtained from M/s Indian Rare Earths, Mumbai, India; CsCl (99.9% pure) and $ZrOCl_2.8H_2O$ (98% pure) procured from M/s. Sigma Aldrich Chemicals Pvt. Ltd., Bengaluru, India and $SrCO_3$ (99.9% pure) obtained from M/s. Otto Chemie Pvt. Ltd., Mumbai, India.

S. No.	**Sample ID**	**Calculated burn-up (at.%)**	**UO_2 (g)**	**Nd_2O_3 (g)**	**$ZrOCl_2$, $8H_2O$ (g)**	**CsCl (g)**	**$SrCO_3$ (g)**	**Atom ratio of $^{238}U/(^{145}Nd + ^{146}Nd)$**
1.	S-II-1	0.73	5.8817	0.0059	0.0145	0.0104	0.0027	2.4E+03
2.	S-II-2	3.44	5.8801	0.0284	0.0695	0.032	0.0128	5.0E+02
3.	S-II-3	6.73	5.9195	0.0579	0.1399	0.0613	0.0258	2.5E+02
4.	S-II-4	9.91	5.8959	0.088	0.2100	0.1008	0.0384	1.6E+02
5.	S-II-5	12.63	5.9050	0.1158	0.2793	0.1236	0.0516	1.2E+02
6.	S-II-6	15.44	5.8884	0.1458	0.3489	0.1548	0.0642	9.8E+01

ESI, Table 3. Details and optimized operating parameters for ICPMS (M/s. SPECTRO MS, Germany. Model: MSS001, 2015).

S. No.	ICPMS instrument parameters	Description
1.	Mass analyzer	Double focusing sector field in Mattauch-Herzog geometry
2.	Mass resolution	500-800 at ^{107}Ag and ^{208}Pb
3.	Sample introduction system	Laser ablation (LA)
4.	Length between LA cell & ICP	~ 1 m
5.	Plasma torch	Fixed quartz with platinum shield
6.	Cooling gas flow rate	12 L/min
7.	Auxiliary gas flow rate	2.3 L/min
8.	Carrier gas flow rate	1.7 L/min
9.	RF-power	1460 W
10.	Detector	Complementary metal oxide semiconductor active pixel direct charge detector array (CMOS-AP-DCD)
11.	Detector mass range	6 to 238 amu (simultaneous measurement)
12.	Detector dynamic Range	> 8 order of magnitude
13.	BaO^+/Ba^+ ratio	≤ 0.3 %
14.	Mass calibration	20 ppb multi-element standard MERCK- VI solution
15.	No. of measurements	21 for each sample
16.	Total integration time	20 s
17.	Base interval	10 ms
18.	Integration Mode	Threshold
19.	Measured isotopes	Relatively high abundance and non-interfering isotopes

ESI, Table 4. Details of the ArF (193 nm) excimer laser used.

S. No.	Laser system	Description
1.	Manufacturer and model	M/s. ATL Lasertechnik GmbH, Germany; ATLEX SP300i.
2.	Pulse width	4-6 ns
3.	Repetition rate	10 Hz
4.	Pulse energy used	2 mJ
5.	Laser spot size	150 μm x 150 μm
6.	Energy fluence at focused spot	1.3 J/cm^2
7.	Target	Sintered pellet (as in Table 1 and 2)

ESI, Table 5. Details of the laser beam homogenizer used.

S. No.	LBH system	Description
1.	Manufacturer and model	M/s. ATL Lasertechnik GmbH, Germany; Light shot micro-machining system
2.	Beam homogenization	Fourier lens in conjunction with an engineered diffuser on motorized rotating mount
3.	Homogenization	Both spatial and temporal
4.	Mask	Stainless-steel sheet cut with 32 different shapes with motorized positioning, providing spot size 2 x 2 (micron) to 150 x 150 (micron) on sample
5.	Energy Control	Motorized beam attenuator: 20-85%
6.	No of focal lens elements	6
7.	Lens materials	UV grade fused silica and CaF_2
8.	Lens clear aperture	ø 22 mm
9.	Useful aperture	ø 20 mm
10.	Effective focal length	50-150 mm
11.	Demagnification	16/10 X
12.	UV optical resolution	<1 µm
13.	UV Field	upto 1 x 1 mm
14.	Focus	0-50 mm
15.	Sample stage X, Y travel	100 x 100 mm
16.	Positioning resolution	1 µm
17.	Positioning repeatability	<3 µm
18.	Positioning speed	<25 mm/s

ESI, Table 6. Atom ratio (AR) obtained from the weight composition [ESI Table 1-2] and measured intensity ratio (IR) of $^{143}Nd/^{238}U$, $(^{145}Nd + ^{146}Nd)/^{238}U$, $^{143}Nd/^{146}Nd$ by LA-ICPMS, the corresponding RSD and error for $^{143}Nd/^{146}Nd$ implies the deviation from natural abundance ratio.

S. No.	$^{143}Nd/^{238}U$			$(^{145}Nd + ^{146}Nd)/^{238}U$			$^{143}Nd/^{146}Nd$		
	AR	IR	RSD (%)	AR	IR	RSD (%)	IR	RSD (%)	Error (%)
	Set-I pellets								
1	2.12E-04	0.27	0.7	4.43E-04	0.58	0.9	0.707	0.5	0.3
2	9.92E-04	1.75	0.3	2.08E-03	3.62	0.4	0.710	0.5	0.2
3	1.96E-03	3.55	0.8	4.10E-03	7.55	0.8	0.709	0.3	0.1
4	2.90E-03	5.32	0.2	6.08E-03	11.33	0.2	0.711	0.3	0.4
5	3.83E-03	6.99	0.2	8.01E-03	14.97	0.1	0.707	0.4	0.3
6	4.88E-03	8.99	0.4	1.02E-02	19.15	0.4	0.709	0.3	0.01
	Linear fit								
	$y = (1832 \pm 9) * x$			$y = (1866 \pm 11) * x$					
	$R^2 = 0.99997$			$R^2 = 0.99998$					
	Set-II pellets								
7	1.98E-04	0.27	0.9	4.13E-04	0.59	0.7	0.707	0.4	0.2
8	9.51E-04	1.72	0.5	1.99E-03	3.67	0.9	0.709	0.4	0.1
9	1.93E-03	3.54	0.6	4.03E-03	7.52	0.6	0.711	0.5	0.3
10	2.94E-03	5.35	0.6	6.15E-03	11.47	0.3	0.709	0.2	0.1
11	3.86E-03	7.11	0.4	8.08E-03	15.12	0.2	0.706	0.3	0.4
12	4.88E-03	9.01	0.3	1.02E-02	19.20	0.5	0.708	0.2	0.1
	Linear fit								
	$y = (1839 \pm 7) * x$			$y = (1874 \pm 7) * x$					
	$R^2 = 0.99993$			$R^2 = 0.99996$					

ESI, Table 7. Atom ratio (AR) obtained from the weight composition [ESI Table 2] and measured intensity ratio (IR) of $^{133}Cs/^{238}U$, $^{90}Zr/^{238}U$ and $^{88}Sr/^{238}U$ by LA-ICPMS for Set-II pellets and the corresponding RSD value.

S. No.	$^{133}Cs/^{238}U$			$^{90}Zr/^{238}U$			$^{88}Sr/^{238}U$		
	AR	**IR**	**RSD (%)**	**AR**	**IR**	**RSD (%)**	**AR**	**IR**	**RSD (%)**
	Set-II pellets								
1	2.86E-03	0.08	0.9	1.07E-03	0.30	0.8	6.98E-04	0.22	0.7
2	8.79E-03	0.25	0.8	5.13E-03	1.60	0.5	3.31E-03	1.22	0.4
3	1.67E-02	0.51	0.6	1.03E-02	3.19	0.3	6.63E-03	2.49	0.3
4	2.76E-02	0.84	0.6	1.55E-02	4.83	0.6	9.91E-03	3.71	0.2
5	3.38E-02	1.04	0.4	2.05E-02	6.41	0.3	1.33E-02	5.05	0.2
6	4.25E-02	1.31	0.3	2.57E-02	8.12	0.2	1.66E-02	6.26	0.6
	Linear fit								
	$y = (31 \pm 0.1) * x$			$y = (314 \pm 1) * x$			$y = (377 \pm 1) * x$		
	$R^2 = 0.99992$			$R^2 = 0.99994$			$R^2 = 0.99993$		

ESI, Table 8. Measured intensity ratio (IR) of $^{143}Nd/^{238}U$, $(^{145}Nd + ^{146}Nd)/^{238}U$, $^{143}Nd/^{146}Nd$ by LA-ICPMS for S-I-3 pellet [ESI Table 1], the corresponding RSD and error (for $^{143}Nd/^{238}U$ and $(^{145}Nd + ^{146}Nd)/^{238}U$ error implies the deviation from the mean value; for $^{143}Nd/^{146}Nd$ error implies the deviation from natural abundance ratio).

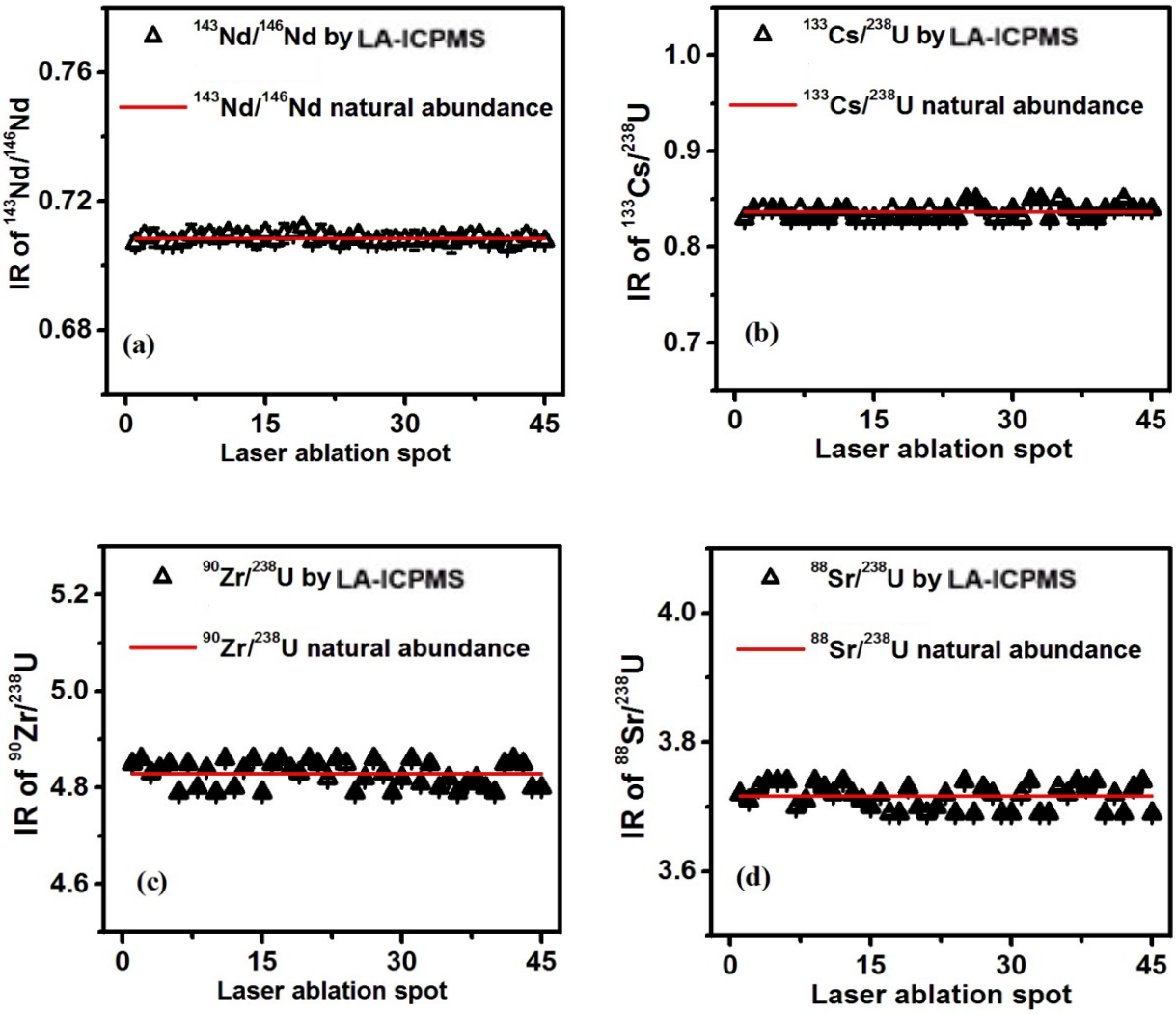
S. No.	No. of laser spots	$^{143}Nd/^{238}U$			$(^{145}Nd + ^{146}Nd)/^{238}U$			$^{143}Nd/^{146}Nd$		
		IR	RSD (%)	Error (%)	IR	RSD	Error (%)	IR	RSD (%)	Error (%)
1	1	3.5	0.5	0.6	7.5	0.4	0.5	0.710	0.3	0.1
2	2	3.5	0.3	0.3	7.5	0.5	0.4	0.709	0.3	0.1
3	3	3.5	0.7	0.1	7.5	0.4	0.2	0.709	0.4	0.03
4	4	3.6	0.4	0.4	7.6	0.7	0.7	0.706	0.2	0.3
5	5	3.5	0.5	0.8	7.5	0.3	0.5	0.708	0.2	0.1
6	6	3.5	0.4	0.7	7.5	0.7	0.2	0.706	0.1	0.4
7	7	3.5	0.7	0.1	7.6	0.2	0.4	0.710	0.4	0.2
8	8	3.5	0.5	0.1	7.5	0.4	0.4	0.708	0.1	0.1
9	9	3.5	0.5	0.5	7.5	0.5	0.5	0.709	0.3	0.03
10	10	3.6	0.7	0.8	7.6	0.4	0.8	0.706	0.2	0.4
11	11	3.6	0.5	0.9	7.6	0.3	0.7	0.710	0.2	0.3
12	12	3.6	0.5	0.5	7.6	0.4	0.2	0.708	0.1	0.02
13	13	3.5	0.6	0.6	7.5	0.5	0.6	0.709	0.2	0.1
14	14	3.6	0.7	0.5	7.6	0.5	0.6	0.712	0.4	0.4
15	**<x>**	3.5			7.5			0.709		
16	**RSD**	0.6			0.5			0.24		

ESI, Table 9. Atom ratio (AR) obtained from the weight composition [ESI Table 1] and measured intensity ratio (IR) of $^{143}Nd/^{238}U$, $(^{145}Nd + ^{146}Nd)/^{238}U$, $^{143}Nd/^{146}Nd$ by liquid sampling ICPMS, the corresponding RSD and error for $^{143}Nd/^{146}Nd$ implies the deviation from natural abundance ratio.

S. No.	$^{143}Nd/^{238}U$			$(^{145}Nd + ^{146}Nd)/^{238}U$			$^{143}Nd/^{146}Nd$		
	AR	**IR**	**RSD (%)**	**AR**	**IR**	**RSD (%)**	**IR**	**RSD (%)**	**Error (%)**
	Set-I pellets, Liquid sampling ICPMS								
1	2.12E-04	0.08	0.8	4.43E-04	0.18	0.6	0.708	0.5	0.1
2	9.92E-04	0.40	0.9	2.08E-03	0.82	0.9	0.709	0.2	0.1
3	1.96E-03	0.80	0.4	4.10E-03	1.72	0.3	0.706	0.4	0.3
4	2.90E-03	1.21	0.5	6.08E-03	2.52	0.7	0.710	0.3	0.2
5	3.83E-03	1.59	0.3	8.01E-03	3.35	0.2	0.707	0.2	0.2
6	4.88E-03	2.03	0.2	1.02E-02	4.31	0.3	0.711	0.3	0.3
	Linear fit								
	$y = (415 \pm 1) * x$			$y = (420 \pm 1) * x$					
	$R^2 = 0.99993$			$R^2 = 0.99994$					

ESI, Table 10. AR determined from calibration plot of liquid ICPMS and measured IR by LA-ICPMS for $^{143}Nd/^{238}U$ and $(^{145}Nd + ^{146}Nd)/^{238}U$.

S. No.	$^{143}Nd/^{238}U$			$(^{145}Nd + ^{146}Nd)/^{238}U$		
	AR	**IR**	**RSD (%)**	**AR**	**IR**	**RSD (%)**
	Set-I pellets					
1	(1.99 ± 0.02)*E-04	0.27	0.7	(4.25 ± 0.03)*E-04	0.58	0.9
2	(9.67 ± 0.09)*E-04	1.75	0.3	(1.95 ± 0.17)*E-03	3.62	0.4
3	(1.92 ± 0.08)*E-03	3.55	0.8	(4.10 ± 0.13)*E-03	7.55	0.8
4	(2.91 ± 0.16)*E-03	5.32	0.2	(6.01 ± 0.44)*E-03	11.33	0.2
5	(3.83 ± 0.12)*E-03	6.99	0.2	(7.97 ± 0.16)*E-03	14.97	0.1
6	(4.89 ± 0.10)*E-03	8.99	0.4	(1.03 ± 0.34)*E-02	19.15	0.4
	Linear fit					
	y = (1833 ± 7) * x			y = (1870 ± 8) * x		
	R^2 = 0.9999			R^2 = 0.9998		



ESI, Figure 1. Plot of intensity ratios (IR) obtained for Set-II-4 pellet [ESI Table 2] using LA-ICPMS - (a) $^{143}Nd/^{146}Nd$; (b) $^{133}Cs/^{238}U$; (c) $^{90}Zr/^{238}U$ and (d) $^{88}Sr/^{238}U$, against different laser ablation spots on the target pellet; the corresponding natural abundance ratio is indicated as a line.