Uranium Potentiality of Sandstones Collected from North-Eastern Part of Bangladesh

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Abstract: The paper represents an attempt to delineate potential of sandstone-hosted uranium mineralization in Kulaura and Baralekha Upazillas using INNA techniques. This study would be of a great help for interchanging necessary data and ideas for uranium exploration in Bangladesh. As a result of this study, low uranium potentiality of sandstones (average 6×10^{-6} U) has been found in the study area. Uranium is strongly associated with Sc, La, Ce, Sm, Tm and Ta.

Key Words: uranium; thorium; sandstone; potentiality; trace elements

Introduction

Bangladesh Atomic Energy Commission (BAEC) has been entrusted with the responsibility of carrying out the surveys, prospecting and exploration of uranium and thorium bearing atomic minerals (pitchblende, uraninite, thorianite, coffinite, uranophane, autunite, uranothorite, zircon, monazite, xenotime etc.) in the prospective areas throughout the country. Bangladesh is a part of the foreland of the Himalayan Geosyncline and nearly borders the ancient stable platform of Archaean basement of Bihar, the Shillong Massif and the Geanticlinorium of Burma (present day Myanmar) consisting of uranium-bearing granite bodies and metasediments. Most of the geological elements such as the presence of tertiary rock formation, suitable host rock and trap, source area, deep seated faults and lineaments, proximity to the uranium provinces of India, which are considered to be favourable for the accumulation of uranium/thorium bearing atomic minerals occur in the geological set up of Bangladesh(Akon). As Bangladesh is geologically made solely of sedimentary rocks, it is only a possibility to mineralize sedimentary types of uranium deposits under favourable reducing environment, which tends to be deposited as commercial uranium ore. Considering the favourable criteria for uranium formation Bangladesh has been divided into four zones as: (1) Eastern Mobile Belt (EMB), (2) Stable Platform (SP), (3) Dauki Fault Belt (DFB) and (4) Dinajpur Slope (DS) (Majumder et al., 2014). Potential areas in Bangladesh for uranium exploration are shown in the Figure 1.

Factors and processes related to the formation of uranium are sources of uranium (granite, rhyolite, syenite, acidic igneous rocks, older crustal rocks, granitic basement rocks), mobilisation of uranium, transportation of uranium to the site of deposition and deposition of uranium, enrichment and preservation. Inspired by favourable factors of accumulation of uranium in the geological content of Bangladesh, exploration for uranium and thorium bearing atomic minerals was initiated by BAEC in 1977. During the exploration works, geological, geophysical, and radiometric surveys were carried out intermittently in different

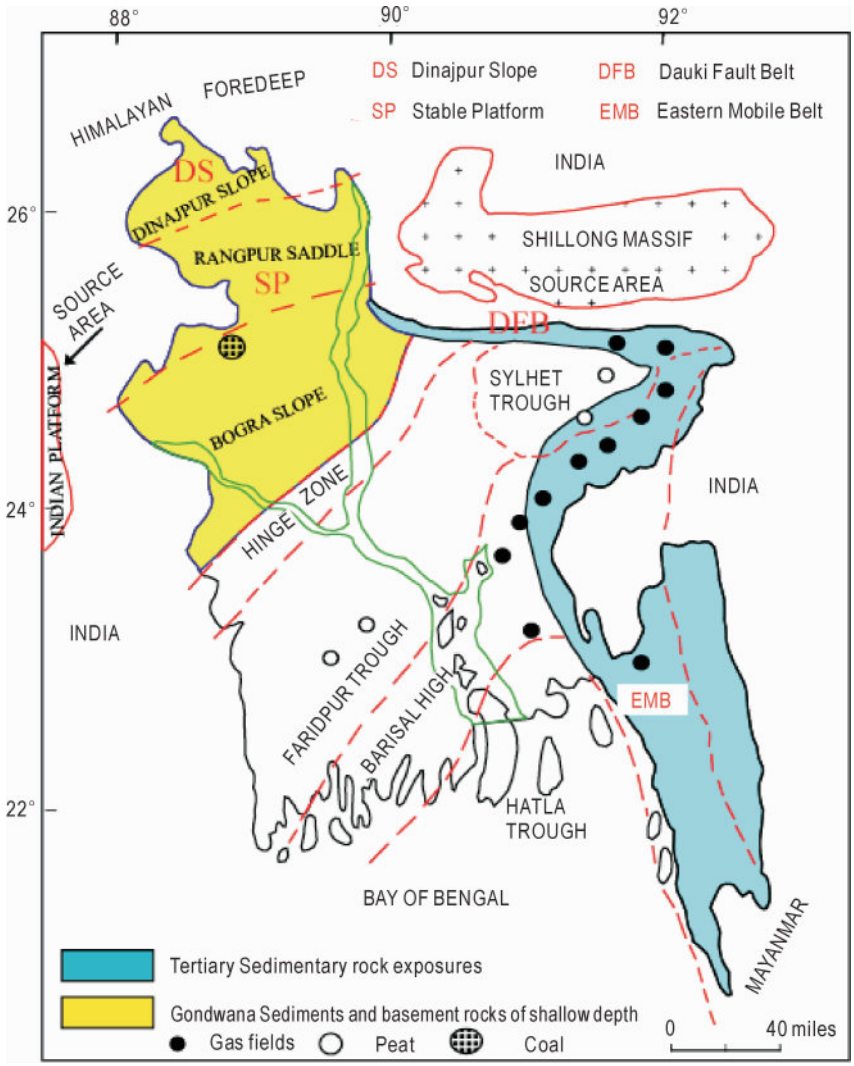

Figure 1 Uranium potential zones of Bangladesh

parts of greater Sylhet district. Laboratory investigations were subsequently carried out. Field methods include radiometric, radon and resistivity survey, portable differential spectrometry, exploratory drilling and geophysical loggings. Laboratory studies include classification of samples according to radio counts; grain size analysis; radioluxography, ore and petrographic microscopy; heavy liquid separation; isodynamic magnetic separation; x-ray fluorescence analysis; mineralogical, geochemical, fluorometric, gamma spectrometric analysis; x-ray diffraction studies and electron probe micro-analysis.

The previous exploration activities carried out so far result in the discovery of a significant number of interesting surface and subsurface radioactive anomalies (3 ~ 60 times to be background) in sandstones of Tipam and Dupitilla Formations of Sylhet and Moulvibazar districts. Anomalies of Harargaj and Lalakhal were quite noteworthy. Radioactive anomalous zone discovered at Fultala of Harargaj Structure, Kulaura, Moulvibazar district was found to bear radioactivity up to 60 times to the background. Mineralogical studies using ore and petrographic microscopy, heavy liquid and isodynamic separator and XRD techniques confirmed the presence of uranium, thorium, rich atomic minerals thorianite and uranothorite in the radioactive sandstone from Fultala area. Main light minerals were quartz and feldspar; the heavy minerals were ilmenite, hematite, zircon, monazite, rutile, garnet and anatase. The presence of sulphide minerals was also indicated. Thorianite and uranothorite contained uranium up to 42% and 15%, respectively. Compositional

mapping of these atomic minerals done by Electron Probe Micro Analyser (EPMA) indicated that they were rich in uranium and thorium. They also contained notable amount of yttrium(Akon).

Chemical analysis of the bulk samples collected from Fultala was carried out using XRF and they were found to contain $(1\ 100 \sim 1\ 400) \times 10^{-6}$ uranium oxide and $(4\ 100 \sim 5\ 000) \times 10^{-6}$ thorium oxide. Anomalous content of uranium in the bulk samples and the presence of uranium rich atomic minerals testified that the processes required for uranium mineralization such as source, mobilization and transportation of uranium to the site of formation and its precipitation as atomic minerals and preservation were accomplished in the sandstone deposits in Bangladesh. The positive findings observed were indicative of the possibility to discover of sandstone type uranium deposits in the Tertiary sedimentary rocks of Bangladesh. Previous investigations showed number of anomalous radioactive sites detected in Harargaj anticline of EMB (Figure 1). These results indicate that uranium bearing solution is still flowing in this zone. So, it can be assumed that the solution has been flowing for very long geologic time and ore might have been formed in and around the sandstones of Harargaj anticline of EMB (NMU/Field (93)/1). Based on the previous survey results, a geological survey was carried out in Kulaura and Baralekha Upazilla of Moulvibazar district in March, 2016. The main objective of the present paper is to find out the potentiality of uranium in sandstones exposed in the study area.

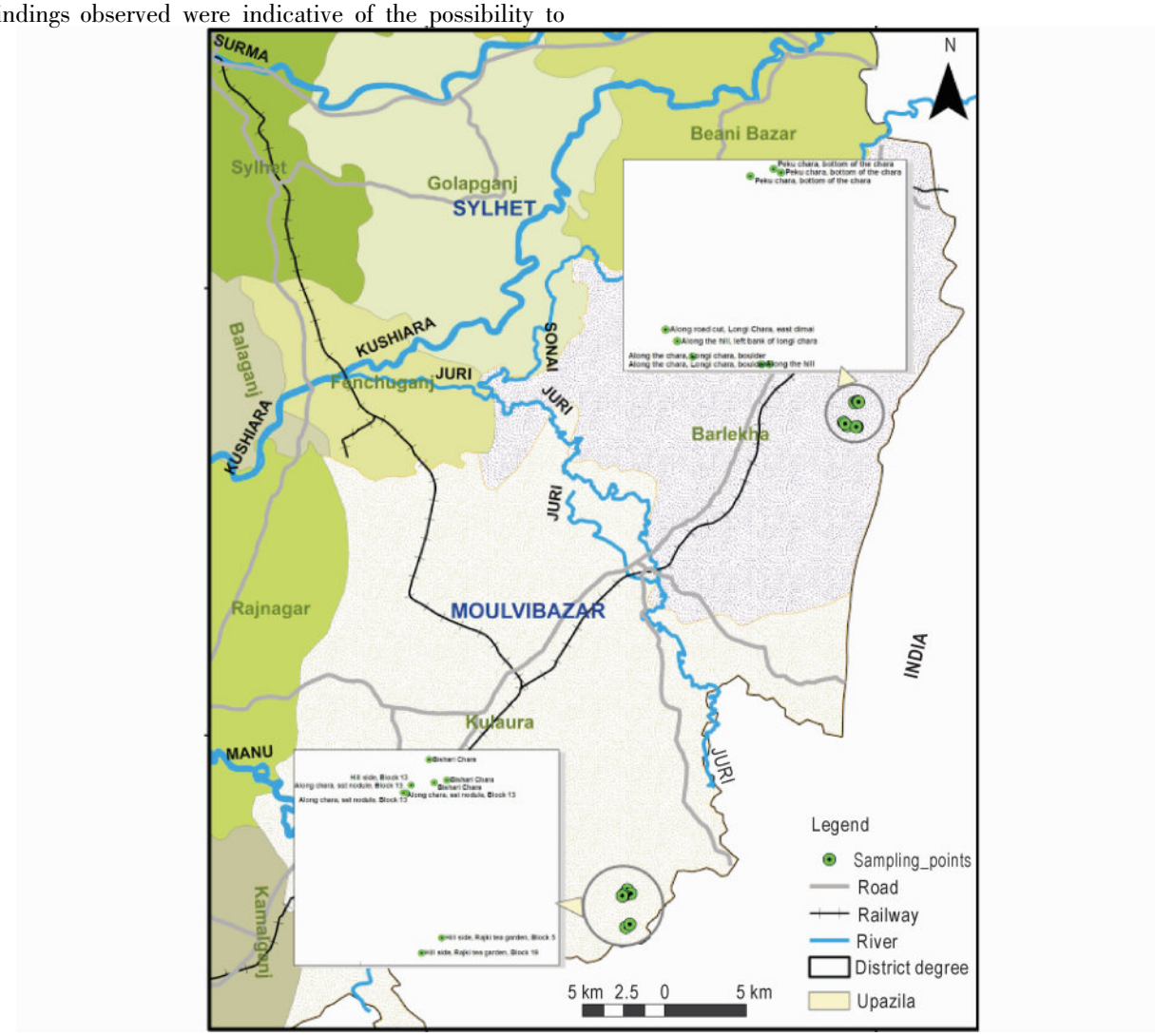

Figure 2 Map of study area with sampling points

1 Location and Genergal Geology

The study area lies (Figure 2) within the western flank of Harargaj anticline in Moulovibazar District. The valley is known as Juri valley. The western flank of Harargaj anticline lies within Bangladesh and the eastern flank in India. The highest peak of the mountain is at an altitude of 375 m. The axial region of Harargaj anticline constitutes Early Miocene Bhuban Formation, the older member of Surma Group. The overlying Boka Bil Formation has developed prominently in the anticline. The Pliocene rock-unit Tipam Sandstone forms the flanks of the anticline. The Harargaj anticline has experienced tremendous tectonic disturbances and thus a great number of faults has developed in different stratigraphic horizons of the structures. The radiometric anomalies occur in pebbles within the Tipam Sandstones.

2 Methods and Techniques

2.1 Sample Collection

A total of 19 stream sediments containing black minerals and sandy/silty pebble samples were collected from Bishari chara (channel) in Kulaura Upazilla and Longi Chara (channel) in Baralekha Upazilla in March, 2016 (Figure 1). The gamma counts of the collected samples ranged from 340 to 917 cps. The geographic coordinates of every sampling point were determined in the field with handheld GPS receiver. The collected samples were kept in polythene bags designated with sampling location, date and sample ID.

2.2 Instrumental Neutron Activation Analysis (INAA)

Neutron activation analysis is an established powerful tool for the determination of trace elements in a variety of matrices (EI-Taher, 2004). The pure instrumental neutron activation analysis (INAA) technique was used in qualitative and quantitative analysis of the rock samples for U, Th, major, trace and rare earth elements analysis (Rahman, 1993). Approximately 100 mg of each dried powder sample was weighted in polyethylene (PE) bag and heat sealed. These PE bags were then enfolded by a second layer of PE bag and heat sealed. For relative standardization approach, reference material (RM): IAEA-Soil-7 and standard reference material NIST-1633b (Coal Fly Ash) were used in this study. Each of the standards was prepared as the same way as those of samples. Samples and standards were placed in a vial for irradiation. IAEA-Soil-7 was used as the standard while NIST-1633b ($n=4$) was used as the control sample.

Two irradiation schemes were performed using pneumatic transfer (rabbit) system at the 3 MW TRIGA Mark-II research reactor of Bangladesh Atomic Energy Commission, Savar: (Ⅰ) short irradiation was performed separately for each sample with the thermal neutron flux of 5.28×10^{12} n · cm^{-2} · sec^{-1} for 60 seconds at 250 kW and (Ⅱ) long irradiation was performed simultaneously with all the samples and standards with the thermal neutron flux of 5.06×10^{13} n · cm^{-2} · sec^{-1} for 7 minutes at 2.4 MW. To determine the neutron flux gradient within the sample stack, three IRMM-530RA Al-0.1% Au (0.1 mm foil) monitor foils were also irradiated by placing them at the bottom, middle and top of the sample stack for the long irradiation scheme. After irradiation second layer of PE bag was replaced by a new one and gamma-ray counting was performed with a high purity germanium (HPGe) detector (CANBERRA, 25% relative efficiency, 1.8 keV resolution at 1 332.5 keV of ^{60}Co) coupled with a digital gamma spectrometer (ORTEC, DSPEC JrTM).

For short irradiation, first counting was performed for 300 seconds after a decay time of about 300 seconds and second counting for 600 seconds after a decay time of 2 to 3 hours. For long irradiated samples, first counting was performed for 40 minutes after a decay time of 2 days while the second counting was performed for 2 hours after a decay time of 7 ~ 10 days. Aluminum, V, Ca, Ti, Mn, U, Dy and Na were determined from the short irradiation scheme whereas Na, K, Sc, Cr, Fe, La, Ce, Nd, Sm, Eu, Tb, Dy, Tm, Yb, Lu, Hf, Ta, Th and U were determined from the long irradiation scheme.

Elemental concentrations obtained from independent replicate measurements of NIST-1633b ($n = 4$) were consistent with the certified value (or the literature data) within the limit of analytical uncertainties (counting statistics, 1σ) and the relative standard deviations (RSDs) were within 10%, except those for Ca, Nd, Tm and Lu (within 12% to 31%).

3 Results

The elements concentrations were quantitatively determined by NAA and the concentrations of major, trace and rare earth elements of collected samples are presented in Table 1. The INAA method has been used to determine the elemental concentration of uranium via ^{238}U and thorium via ^{232}Th in the collected samples. The average U and Th concentration in the collected samples were about 6×10^{-6} (0.000 6%) and 47.1×10^{-6} (0.005%) respectively. The average concentration of Na, Al, K, Ca, Ti and Fe in collected samples were 0.567%, 6.71%, 2.08%, 0.69%, 0.53% and 8.12% respectively. Besides, the average concentrations of Sc, V, Cr, Mn, La, Ce, Nd, Sm, Eu, Tb, Dy, Tm, Yb, Lu, Hf and Ta were 14.7×10^{-6}, 106×10^{-6}, 96×10^{-6}, $3\,918 \times 10^{-6}$, 60.5×10^{-6}, 172×10^{-6}, 85×10^{-6}, 8.23×10^{-6}, 1.57×10^{-6}, 1.19×10^{-6}, 11.91×10^{-6}, 2.92×10^{-6}, 4.93×10^{-6}, 0.505×10^{-6}, 9.9×10^{-6} and 1.77×10^{-6} respectively.

4 Discussion

The data presented in this work show insignificant concentrations of U and Th in the collected samples. Correlation analysis plays a significant role in explaining U and Th associations with other elements in the study area. The results show that the contents of K, Ca, Sc, Ti, V, Cr, Mn, La, Ce, Nd, Sm, Eu, Tb, Dy, Tm, Yb, Lu, Hf and Ta are strongly correlated with U and Th (Table 2). U and Th are negatively correlated with Na, moderately correlated with Al and insignificantly correlated with Fe. But the highest correlation coefficients are found for Sc, La, Ce, Sm, Tm and Ta ($r^2 > 0.90$). Therefore, these significant relationships among the measured variables indicate their common sources and also their moving together, especially from U-Th bearing rocks around the study area. It can be considered that transportation of U and Th-bearing minerals has been taken place in the study area and has been absorbed by pebbly sandstones. Under some circumstances, it is also possible for uranium to be moved over considerable distances by water.

5 Conclusions

Neutron activation analysis is an established powerful tool for the determination of trace elements in a variety of matrices. The results obtained for U concentration by INAA have been found insignificant which are not reasonable for the potentiality of sandstones in the study area. But the evidence of absorbed uranium in pebbly sandstones in the study area has increased the importance for carrying out extensive exploration activities for uranium in and around the study area.

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Table 1 U, Th, major, trace and rare earth element concentrations of analysed samples

Sample ID	Na /%	Al /%	K /%	Ca /%	Sc /10^{-6}	Ti /%	V /10^{-6}	Cr /10^{-6}	Mn /10^{-6}	Fe /%	La /10^{-6}	Ce /10^{-6}	Nd /10^{-6}	Sm /10^{-6}	Eu /10^{-6}	Tb /10^{-6}	Dy /10^{-6}	Tm /10^{-6}	Yb /10^{-6}	Lu /10^{-6}	Hf /10^{-6}	Ta /10^{-6}	Th /10^{-6}	U /10^{-6}
SS-1	1.319	7.38	2.13	1.05	13.8	0.351	104	91	6 185	4.16	52.7	119	61	9.90	1.44	1.16	8.66	2.91	4.40	0.497	12.4		22.8	5.7
SS-2	0.873	6.57	2.07	0.42	10.9	0.365	70	58	546	5.97	41.5	100	46	7.19	1.29	0.93	4.73		3.21	0.367	7.3	0.96	25.0	5.6
SS-3	0.917	7.64	2.37	0.49	12.1	0.453	69	61	6 147	5.12	52.7	122	62	8.38	1.67	0.82	7.16	1.83	3.31	0.427	6.7	1.31	28.1	6.2
SS-4	0.276	4.95	1.41	0.95	15.2	0.301	101	71	2 782	20.30	28.4	73	45	5.89	1.22	0.68	6.67	3.75	4.08	0.475	5.0	0.72	13.3	4.4
SS-5	0.605	11.91	1.10	3.30	40.1	2.572	476	307	6 191	11.23	398.6	895	417	47.38	3.81	4.58	67.19	6.60	15.94	1.832	21.2	4.58	227.1	16.7
SS-6	0.618	5.95	1.75	0.97	15.6	0.432	100	85	4 386	12.61	45.6	117	55	9.35	1.69	1.11	7.60	2.82	5.45	0.475	6.7	0.72	18.5	5.6
SS-7	0.080	7.46	4.64	1.28	57.4	1.355	275	269	9 921	17.80	339.1	966	359	44.66	3.76	3.86	33.01	7.62	18.90	1.558	26.8	12.52	363.9	29.0
SS-8	0.303	7.02	1.99	0.16	11.8	0.342	82	83	5 166	19.42	20.2	57	18	3.69	0.86	0.51		2.39	2.08	0.188	5.3	0.85	12.5	3.5
SS-9	0.237	7.95	2.02	0.22	12.1	0.347	83	95	1 304	16.77	27.8	58	42	5.20	1.06	0.60	4.48	2.92	3.11	0.343	7.4	0.98	20.6	3.5
SS-10	0.775	6.77	2.22	0.12	11.1	0.492	81	81	5 086	3.37	38.5	96	44	6.23	0.91	0.74		2.75	3.22	0.335	9.4	1.07	18.6	5.8
SS-11	1.339	5.56	1.97	0.61	8.5	0.251	52	55	4 553	2.81	36.4	82	52	6.32	1.20	0.74	6.05	1.75	3.19	0.356	8.9	0.94	16.8	4.9
SS-12	0.240	4.82	1.99	0.26	6.0	0.221	71	65	745	13.83	2.2	30	18	0.11	0.58	0.33	2.16	1.72	1.65	0.199	4.4	0.53	11.4	1.8
SS-13	0.596	5.87	2.10	0.43	9.8	0.354	59	68	3 340	3.26	0.5	86	51	0.26	1.33	0.71	4.74	2.03	3.19	0.299	9.5	0.99	15.2	2.9
SS-14	0.214	6.46	2.02	0.33	11.1	0.502	63	92	2 774	3.04	1.6	100	50	0.42	0.93	0.71	5.89	2.59	3.88	0.425	17.1	1.20	27.6	3.8
SS-15	0.393	6.30	2.09	0.16	9.5	0.319	66	62	3 821	2.95	1.8	66	40	0.24	1.08	0.48	7.51	1.84	2.42	0.262	6.5	0.83	13.7	2.3
SS-16	0.579	5.29	1.77	0.82	8.2	0.346	54	75	4 640	2.58	55.0	99	67	0.36	1.69	1.02	8.81	2.38	3.71	0.415	12.8	1.18	19.7	2.6
SS-17	0.545	6.49	2.16	0.45	10.5	0.384	74	80	477	3.31	0.9	77	44	0.29	1.26	0.77	6.76	1.85	3.07	0.332	7.4	0.95	14.4	3.0
SS-18	0.597	6.59	2.08	0.54	10.4	0.387	77	81	5 626	3.52	0.7	91	110	0.27	3.30	2.30	14.12	2.81	7.18	0.626	8.8	1.03	15.7	3.8
SS-19	0.272	6.39	1.58	0.47	5.2	0.205	60	38	744	2.17	4.6	36	30	0.20	0.76	0.51	6.86	1.97	1.63	0.175	4.2	0.45	10.2	2.2
Avg. Conc.	0.567	6.71	2.08	0.69	14.7	0.53	106	96	3918	8.12	60.5	172	85	8.23	1.57	1.19	11.91	2.92	4.93	0.505	9.9	1.77	47.1	6.0

Table 2 Correlation coefficients between U, Th, major, trace and rare earth elements concentrations in samples

	Na	Al	K	Ca	Sc	Ti	V	Cr	Mn	Fe	La	Ce	Nd	Sm	Eu	Tb	Dy	Tm	Yb	Lu	Hf	Ta	Th	U
Na	1.000																							
Al	0.057	1.000																						
K	−0.178	−0.035	1.000																					
Ca	0.087	0.712	−0.141	1.000																				
Sc	−0.236	0.576	0.579	0.680	1.000																			
Ti	−0.100	0.842	0.080	0.896	0.807	1.000																		
V	−0.124	0.816	0.089	0.916	0.837	0.983	1.000																	
Cr	−0.214	0.750	0.331	0.820	0.932	0.946	0.955	1.000																
Mn	0.172	0.377	0.521	0.431	0.675	0.490	0.488	0.591	1.000															
Fe	−0.505	0.119	0.163	0.200	0.448	0.230	0.344	0.376	0.144	1.000														
La	−0.093	0.733	0.321	0.853	0.928	0.939	0.951	0.966	0.616	0.334	1.000													
Ce	−0.176	0.675	0.451	0.784	0.968	0.907	0.916	0.971	0.638	0.319	0.981	1.000												
Nd	−0.133	0.727	0.331	0.846	0.920	0.942	0.944	0.970	0.621	0.252	0.973	0.981	1.000											
Sm	−0.062	0.727	0.371	0.823	0.948	0.910	0.935	0.954	0.634	0.386	0.987	0.974	0.953	1.000										
Eu	−0.026	0.591	0.345	0.731	0.796	0.768	0.765	0.809	0.686	0.147	0.802	0.830	0.891	0.786	1.000									
Tb	−0.072	0.712	0.301	0.839	0.877	0.906	0.909	0.933	0.643	0.212	0.925	0.935	0.977	0.908	0.952	1.000								
Dy	−0.084	0.835	0.046	0.933	0.785	0.985	0.979	0.928	0.542	0.252	0.929	0.894	0.951	0.893	0.820	0.933	1.000							
Tm	−0.265	0.602	0.427	0.745	0.965	0.834	0.874	0.942	0.634	0.484	0.926	0.944	0.924	0.936	0.802	0.898	0.827	1.000						
Yb	−0.171	0.613	0.478	0.763	0.963	0.856	0.871	0.946	0.687	0.321	0.935	0.971	0.970	0.936	0.913	0.965	0.862	0.957	1.000					
Lu	−0.099	0.722	0.302	0.872	0.921	0.938	0.945	0.968	0.631	0.284	0.966	0.968	0.990	0.955	0.898	0.978	0.946	0.938	0.973	1.000				
Hf	−0.101	0.529	0.512	0.622	0.841	0.750	0.713	0.852	0.653	0.071	0.816	0.864	0.844	0.794	0.726	0.811	0.720	0.824	0.865	0.845	1.000			
Ta	−0.294	0.395	0.783	0.477	0.946	0.632	0.656	0.815	0.695	0.349	0.819	0.895	0.821	0.836	0.718	0.771	0.611	0.873	0.893	0.800	0.843	1.000		
Th	−0.256	0.556	0.614	0.649	0.981	0.801	0.819	0.922	0.641	0.361	0.929	0.976	0.928	0.933	0.783	0.874	0.782	0.935	0.954	0.908	0.863	0.968	1.000	
U	−0.152	0.524	0.666	0.609	0.981	0.748	0.771	0.880	0.698	0.369	0.904	0.949	0.889	0.931	0.772	0.846	0.726	0.927	0.941	0.883	0.838	0.969	0.981	1.000