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Comparison Analysis of Heavy Metal Detection in Soil Samples; TANDEM Accelerator and VDG Accelerator

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Abstract: Proton Induced X-Ray Emission (PIXE) technique, using accelerator facility is a well known for detection heavy metal in different samples upto micro level range. In this study PIXE technique is used for both TANDEM accelerator and Van de Graaff (VDG) accelerator to detect heavy metal in some soil samples of two Aila affected upozilas of two districts. A total 22 common samples are irradiated by both the systems and their data was compared and analyzed. The obtained data was nearly same and comparable for both the systems except very few. Obtained data from both the facilities provided us to the consistence of work of relevant issues.

Keywords: PIXE, TANDEM, VDG, Comparison, Heavy Metal.

1 Introduction

Ion Beam Analysis (IBA) technique is one of the prominent ways of determining many elements simultaneously on a typical sample; since it is multi elemental, sensitive and virtually nondestructive [1]**.** PIXE method is suitable for the analysis of heavy elements of high Z region [2-8]. PIXE is a powerful yet non-destructive elemental analysis technique now used routinely by geologists, archaeologists, art conservators and others to help answer questions of provenance, dating and authenticity. The fundamental interest of the PIXE method is to determine the concentrations (relative or absolute) of elemental constituents in the sample under investigation.

Some of the experiments in this research work have been performed using the 3 MeV VDG accelerator of the Atomic Energy Center, Dhaka (AECD), supplied by High Voltage Engineering Corporation, USA and a modified form of VDG known as TANDEM accelerator, Institution of Nuclear Science and Technology (INST), Atomic Energy Research Establishment (AERE), Savar, Dhaka, manufactured by the High Voltage Engineering Corporation Europa BV (HVEE), the Netherland.

VDG accelerator at AECD established in 1964 which is using for IBA techniques for nuclear analysis while TANDEM accelerator started its function in 2011. A new state-of-the-art 3 MV TANDEM Accelerator facility has been installed at the AERE, Ganakbari, Savar under Bangladesh Atomic Energy Commission (BAEC), in order to continue the research and contribution to the society using accelerator related IBA techniques such as PIXE. A total 22 soil samples are irradiated in both the facilities using PIXE method. All the samples are prepared for irradiate at Accelerator Facility Division (AFD), Atomic Energy Center, Dhaka (AECD). The obtained results has taken into consideration and analyzed for the comparison of performance of both the facilities as well as the concentration of heavy metal into the samples.

2 Experimental Procedure

2.1 Sample Collection and Preparation:

To determine the material concentration in the present study of two selected Aila affected Upazila, in two District, a total 22 soil samples were collected. Soil samples were collected from 5 to 20 cm depth to avoid the very recent upper surface soil and dust deposition and to ensure the cultivated soil for agricultural purpose. For ensuring the

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homogeneity of the soil a considerable amount of soil was collected randomly within space limit of 4 sq.ft (2ft x 2ft) from each location. All the soil samples were dried at 80^{O}C into a temperature controlled electric oven for a week (168 hours). Then the dried samples were mashed by using an agate motor and hand grandeur. To determine the elemental concentration, powdered samples of the soil were pressed at a pressure of 250 kg per cm^2 using a hydraulic press to prepare pellets (~ 0.2 gm) of 7 mm diameter and about 1 mm thickness. Several pellets of same size and shapes also were prepared adopting the same criteria on the standard reference soil provided by International Atomic Energy Agency (IAEA) and National Institute of Standard and Technology (NIST). The preparation work was carried out at AFD, AECD.

2.2 Experimental Methodology

After preparing the sample pelletsat a time 13 soil samples, a quartz and 2 standard samples (Total 16 slides of samples) were set with the wheeler which was then set in the proton beam scattering chamber. Then the chamber was made vacuum by using vacuum pump up to the level of $\sim 10^{-5}$ Pascal. Achieving the required vacuum, the samples were irradiated by the proton beam of energy 2.2 – 2.5 MeV with a beam current of 10 to 15 nA. The obtained data from PIXE technique in 3MV Van de Graaff accelerator at Atomic Energy Center, Dhaka (AECD) were analyzed by MAESTRO-32 PIXE program and the Guelph GUPIX PIXE program with the interface of DAN32. DAN32 is a program designed to run under Windows. The program is designed to facilitate the analysis of PIXE and RBS spectra and to implement the "Q-factor" normalization technique [9]. The X-ray spectrum were analyzed with the help of well established PIXE technique and by the use of the Si(Li) detector.

3 Result and Discussion

The elemental concentration values of the soil samples measured using the Tandem Facilty (PIXE method) for the two different upazilas (Moralgonj Upazila, Bagerhat and Koira Upazila, Khulna) are presented in Table 1 and Table 2 respectively. Elemental concentrations also measured in the soil samples of those two different Upazilas (Moralgonj Upazila, Bagerhat and Koira Upazila, Khulna) using the VGD Facility (PIXE method) are presented in Table 3 and Table 4 respectively.

From the obtained results presented in Tables 1 and 2 for Tandem accelerator and Tables 3 and 4 for the VDG accelerator, one may find that the results from measurements made using the Tandem accelerator and VDG are very close. So both the results for elemental concentration are reliable and thus acceptable.

The main goal of this present study was to detect the presence of heavy metals like Ni, Cu, Zn, Hg and Pb and hence to determine the concentration of it in the samples of some selected areas affected by the natural disaster Aila. This is being discussed now.

Ni concentration as observed in the work fall within the range 0 to 3725 µg/g. Only one sample (G3_10475.gpx) collected from Moralgonj Upazila, Bagerhat and irradiated by the Tandem accelerator facility shows a very high Ni concentration. Whereas the Ni concentration at Moralgonj soil samples irradiated by the VDG ranges from 0 to 502 µg/g, which is comparable with the results of other upazilas. Concentration for the element at Koira Upazila, Khulna ranges from 0 to 411 µg/g as observed through irradiation using the Tandem accelerator facility. VDG irradiated soils had the concentration within 0 to 458 µg/g. In total 6 samples (G3_10472.gpx, G3_10450.gpx, G3_10478.gpx, G3_10445.gpx, G3_10479.gpx, G3_10452.gpx of Table 2) out of 9 and 5 samples (PX-014, PX-059, PX-060, PX-061, PX-092 of Table 4) from 10 have shown the presence of Ni after being irradiated by Tandem and VDG facilities respectively.

Thirteen soil samples of Moralgonj Upazila irradiated by the Tandem facility and by the VDG facility show the Cu concentration ranging within 0 to 2582 µg/g and 0 to 2954 µg/g respectively. Out of 13 soil samples 8 show the presence of Cu (Sample ID: G3_10473.gpx, G3_10474.gpx, G3_10475.gpx, G3_10470.gpx, G3_10476.gpx, G3_10471.gpx, G3_10477.gpx, G3_10453.gpx, Table 1) in the Tandem irradiated samples and 9 show the presence of the element in the VDG irradiated samples (Sample ID: PX-002, PX-003, PX-004, PX-005, PX-007, PX-008, PX-009, PX-012, PX-065 in Table 3). The Zn concentration ranging from 0 to 4775 µg/g and 0 to 3481 µg/g shown by Tandem and VDG irradiated sample respectively. Out of 13 soil samples 8 from each samples show the Zn concentration. Only 2 samples from 13 show the presence of Hg - concentration of which falls within 13 and 21 µg/g. This has been observed amongst the VDG irradiated samples, where as the Tandem facility could not identify them. The concentration of Pb ranges from 0 to 427 µg/g as observed by the Tandem facility and 0 to 435 µg/g by the VDG. Three (Sample ID: G3_10473.gpx, G3_10470.gpx, G3_10476.gpx Table 1) from 13 samples and 4 (Sample ID: PX-002, PX-005, PX-012, PX-065 in Table 3) from 13 samples has shown the presence of Pb as observed by the Tandem and the VDG facilities respectively.

The samples in Koira Upazila, irradiated by the Tandem accelerator show concentration level of Cu (observed in 2 out of 9 samples, Sample ID: G3_10472.gpx, G3_10478.gpx in Table 2) ranging from 298 to 1313 µg/g; that of Zn (observed in 5 out of 9 samples, Sample ID: G3_10472.gpx, G3_10449.gpx, G3_10478.gpx, G3_10445.gpx, G3_10454.gpx, in Table 2) ranging from 94 to 6174 µg/g, and Pb (observed in 3 out of 9 samples, Sample ID: G3_10478.gpx, G3_10445.gpx, G3_10479.gpx in Table 2) concentration ranges from 108 to 164 µg/g. Presence of Hg could not be considered for detection at Tandem facility. While the samples irradiated by the VDG

Table 1: Elemental concentrations in soil samples of Moralgonj Upazila measured using the Tandem facility.

Sample ID	Material Concentration in µg/g										
	K	Ca	Ti	Cr	Mn	Fe	Ni	Cu	Zn	Hg	Pb
G3_10446.gpx	23826	10494	7223	283	250	51730	-	0	-	Not done	0
G3_10473.gpx	12589	17999	4472	252	144	35321	303	2462	510	Not done	427
G3_10474.gpx	15553	21932	7972	183	686	48881	240	1290	637	Not done	0
G3_10475.gpx	16254	11567	5134	1704	1724	82041	3725	234	-	Not done	0
G3_10470.gpx	14695	19347	5778	205	297	41502	324	2180	847	Not done	313
G3_10447.gpx	23171	28118	8388	270	660	53511	0	0	-	Not done	0
G3_10476.gpx	20814	18842	7008	355	396	53571	458	2582	1229	Not done	294
G3_10471.gpx	16290	13561	6190	287	91	46879	252	1305	4745	Not done	0
G3_10477.gpx	14906	23053	7195	85	787	49000	0	400	1634	Not done	0
G3_10457.gpx	12163	14112	8006	251	623	51738	-	-	104	Not done	0
G3_10448.gpx	23652	42964	7376	220	437	53117	208	0	0	Not done	0
G3_10456.gpx	11423	22777	8313	316	600	60439	201	-	-	Not done	0
G3_10453.gpx	24179	30718	8026	101	155	48859	0	81	76	Not done	0

Table 2: Elemental concentrations in soil samples of Koira Upazila measured using the Tandem facility.

Sample ID	Material Concentration in µg/g										
	K	Ca	Ti	Cr	Mn	Fe	Ni	Cu	Zn	Hg	Pb
G3_10472.gpx	14097	10288	5665	112	214	41357	171	298	1161	Not done	0
G3_10449.gpx	25210	7424	7654	0	197	47568	0	-	94	Not done	-
G3_10451.gpx	16774	30886	7877	300	0	45983	-	0	-	Not done	0
G3_10450.gpx	18313	25748	7557	196	505	47533	168	0	0	Not done	0
G3_10478.gpx	13541	44224	4735	-	376	33932	199	1313	6174	Not done	164
G3_10445.gpx	25376	5689	7559	149	0	46913	177	0	94	Not done	118
G3_10479.gpx	16087	58651	5410	1847	428	45584	411	-	-	Not done	108
G3_10454.gpx	20268	23459	8524	0	504	46191	-	0	95	Not done	0
G3_10452.gpx	25075	8789	6670	205	-	48075	181	-	0	Not done	-

Table 3: Elemental concentration in soil samples of Moralgonj Upazila, Bagerhat measured using VDG Facility, AECD

Sample ID	Material Concentration in µg/g										
	K	Ca	Ti	Cr	Mn	Fe	Ni	Cu	Zn	Hg	Pb
PX-001	28750	14182	6995	186	231	49368	0	-	117	0	0
PX-002	10819	19153	8603	160	254	37320	346	2954	3481	0	352
PX-003	18322	12857	5334	-	448	54286	-	152	486	13	0
PX-004	14476	31232	6619	1537	1670	87134	314	283	0	0	0
PX-005	12615	21322	5123	198	302	46343	264	1354	584	21	435
PX-006	18171	27124	7206	179	731	58217	0	-	-	0	0
PX-007	20242	22155	5678	402	367	58524	502	2711	918	0	0
PX-008	14096	15263	6564	321	-	41835	316	905	2646	0	0
PX-009	13944	24321	4289	-	573	48073	-	451	1139	0	0
PX-010	12745	17314	6280	289	801	53812	183	-	206	0	0
PX-011	19634	36783	9321	198	532	59423	251	-	0	0	0
PX-012	13454	26362	7312	405	554	54434	226	87	-	0	348
PX-065	21173	28712	6270	-	-	51296	-	96	-	0	256

Table 4: Elemental concentrations in soil samples of Koira Upazila, Khulna measured using the VDG Facility, AECD

Sample ID	Material Concentration in µg/g										
	K	Ca	Ti	Cr	Mn	Fe	Ni	Cu	Zn	Hg	Pb
PX-014	14097	10288	5665	112	214	41357	171	538	1161	0	321
PX-015	25210	7424	7654	0	197	47568	0	-	94	0	0
PX-047	16774	30886	7877	300	0	45983	0	0	43	0	0
PX-048	18313	25748	7557	196	505	47533	-	0	61	25	0
PX-059	10257	34297	4036	-	310	25044	407	433	2312	0	156
PX-060	18308	11715	7910	524	654	46491	458	0	288	14	0
PX-061	20682	57041	6033	1323	501	53589	360	601	290	20	289
PX-062	21247	51542	6507	-	456	52188	0	0	-	0	0
PX-091	26322	11746	8216	-	0	54752	-	-	-	0	0
PX-092	23241	29154	6824	352	-	39458	235	381	-	0	0

Table 5: Reference value and obtained value of SRM 2586 soil and sample of Soil-7.

Standard Soil Samples	Elemental concentration in µg/g										
	K	Ca	Ti	V	Cr	Mn	Fe	Ni	Cu	Zn	Pb
Reference value (SRM 2586)	9760	22100	6050	160	301	1000	51610	75	81	352	432
Obtained value (SRM 2586)	9160	24922	5281	179	289	885	46140	89	93	302	350
Reference value (Soil-7)	12100	163000	3000	66	60	631	25700	26	11	104	60
Obtained value (Soil-7)	13068	135296	3392		74	588	31531			125	

facility, the concentration level of Cu has been observed in 4 (Sample ID: PX-014, PX-059, PX-061 and PX-092 in Table 4) out of 10 samples whose range falls within 381 to 601 µg/g, Zn concentration falls within (observed in 7 out of 10 samples, Sample ID: PX-014, PX-015, PX-047, PX-048, PX-059, PX-060 and PX-061 in Table 4) 43 to 2312 µg/g, Hg (observed in 3 from 10 samples, Sample ID: PX-048, PX-060 and PX-061 in Table 4) within 14 to 25 µg/g and Pb (observed in 3 from 10 samples, Sample ID: PX-014, PX-059 and PX-061 in Table 4) concentration falls within 156 to 321 µg/g.

The obtained results of measurements of elemental concentration were compared with the IAEA reference values and NIST reference values. Table 5 shows the results for the reference values and obtained values of SRM 2586 soil and Soil-7 reference soil. Ratios between the reference values and obtained values for the selective materials are shown in Figs. 1 and 2 respectively.

For most of the elements the ratios are very close to one. Thus, the good consistency between the measured and NIST values confirms the reliability of our measurements of soil samples using the Tandem accelerator. Similarly the ratios of the reference and measured values of the Soil-7 samples using the 3MV VDG accelerator assure us of the system to be reliable for the study.

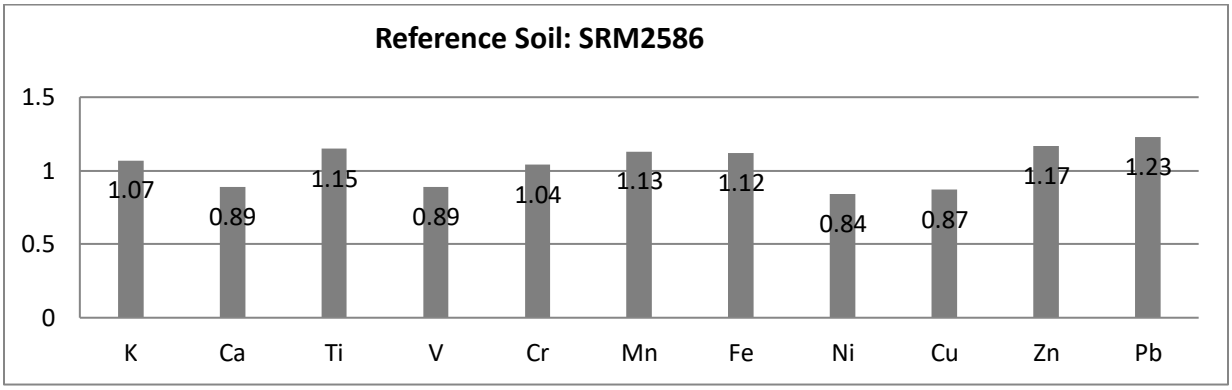

Fig.1: Ratio between the reference values and obtained values of the selective material of reference soil: SRM 2586.

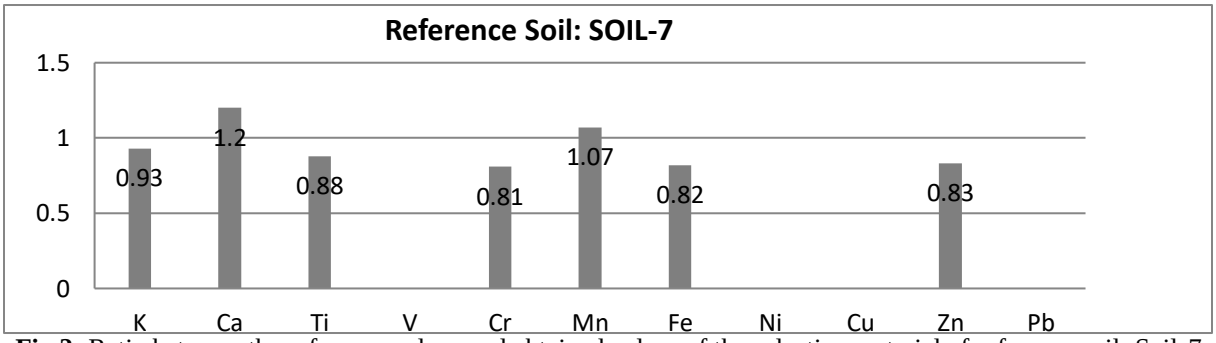

Fig.2: Ratio between the reference values and obtained values of the selective material of reference soil: Soil-7

4 Conclusions

Elemental analysis of the collected samples was carried out by using PIXE technique at 3 MV Van-de-Graaff Accelerator, AFD, AECD and 3 MV Tandem Accelerator, INST, AERE, Savar, Dhaka, Bangladesh. The obtained x-ray spectra were analyzed using the GUPIXWIN software for Tandem irradiated samples and Maestro-32 with GUPIX/DAN 32 for Van-de Graaff Accelerator samples. Quality control (QC) tests of the measurements were also done for both the accelerators. In this work the elemental concentration values of samples of two upazilas (Moralgonj Upazila, Bagerhat and Koira Upazila, Khulna) were found nearly same when measured using the 2 different facilities, e.g., Tandem facility and VDG facility. The heavy metals present in a particular sample of those locations could be detected using both the facilities; the results also become nearly same. So, we may conclude that, for heavy metal detection and analyze both facilities (Tandem facility and VDG facility) are equally applicable.

It also observed that in some of the samples the elemental concentration of Ni, Cu, Zn, Hg and Pb in the investigated soil samples is slightly higher than the values obtained from reference soil samples provided from IAEA and NIST. However, Prompt investigation is necessary to check the soil status after any natural devastation like cyclone of the locality.

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