Proceeding of 3rd International Science Postgraduate Conference 2015(ISPC2015)

NATURAL RADIOACTIVITY FROM NON-NUCLEAR POWER GENERATION INDUSTRIES: REGULATORY CONTROL OF NATURALLY OCCURRING RADIOACTIVE MATERIAL (NORM) FOR ENVIRONMENTAL SUSTAINABILITY.

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Abstract. Coal is a widely used mineral and contains almost all elements which include Naturally Occurring Radioactive Material (NORM) from natural origin such as from Uranium-238 (^{238}U) and Thorium-232 (^{232}Th) Series along with Potassium-40 (^{40}K). It is the most important source of fossil fuel for non-nuclear power generation industries due to itsaccessibility and abundance in nature. The burning of coal generates bottom and fly ash which are released into the atmosphere. This process potentially tends to distribute the natural radionuclides originating from coal and enriched in the environment that could contribute to higher external radiation exposure to the population at large. This study aimed to presents an analysis of radioactivity concentration of feed coal burned and ashes from a typical coal fired power plants (CFPP) which is a non-nuclear power generation in Malaysia. The sample was analyzed for two most important natural radionuclides content namely ^{238}U and ^{232}Th by using Instrumental Neutron Activation Analysis (INAA). An extensive investigation of this nature is warranted for radiation protection towards legislative compliance in ensuring safety of the public and workers and the protection of the environment.

*Keywords.*Natural occurring radioactive material (NORM), radioactivity, coal combustion, coal fired power plant and Instrumental Neutron Activation Analysis (INAA).

1.0 INTRODUCTION

Coal contains almost all elements which include NORM from natural origin. Human activities, such as exploration and processing of these resources and production of consumer items can lead to further enhancement of the natural radioactivity in the products, by-products, residues or wastes, arising from industrial processing [1], and eventually will become Technologically Enhanced or TENORM. Currently, NORM and TENORM have received various attentions, local and international due to its significant amount produced annually.

Coal is widely available source of fossil fuel for non-nuclear power generation industries and it will be the main world's electricity supply for many years ahead. As the availability of natural gas and petroleum decreases, with the subsequent price increases and uncertainty in nuclear power industry has lead coal remained to be relevant [2]. Therefore, for non-nuclear power generation industries sector, CFPP continue to become the most popular choice in power generation industry compared to others. However, combustion of coal in CFPP discharged gaseous, particulates and coal combustion by-products (CCBs) that contain radionuclides released to the atmosphere. In addition, these radionuclides materials are partitioned in the bottom ash (BA), fly ash (FA) and flue gas (FG) when coal is burned. The radionuclides include Uranium decay series such as ^{238}U, Uranium-234 (^{234}U), Thorium-230 (^{230}Th), Radium-226 (^{226}Ra), Radon-222 (^{222}Rn) including daughters Plumbum-210 (^{210}Pb), Polonium-210 (^{210}Po) and Thorium decay series such as ^{232}Th, Radium-228 (^{228}Ra) and Thorium-228 (^{228}Th) [3].The radioactivity level of these radionuclides may enhance by many orders of magnitudes in the ashes compared to raw coal. This happens as fine particles of coal ash with large surface area have a greater tendency to absorb the radionuclides during the combustion process [4-7]. Released of radionuclides from CFPP may cause serious contamination to the atmosphere and the terrestrial environment [7]. This contributes external radiation exposures to the natural environment leading to an increment in the background radiation level. National legislation approach shall consider the impacts of NORM related industries, if any, based on radiological and non-radiological aspects. Thus, it is important to address this issue seriously as to ensure the safety of the public and the protection of the environment for environmental sustainability. This study presents detail radioactivity level by an analysis of activity concentration of feed coal (FC) burned and ashes from a typical CFPP. In addition, the importance of regulatory control of NORM activities for environmental sustainability under normal (not emergency; either accident or incident) operations as discussed.

2.0 EXPERIMENTAL

2.1 The Coal Fired Power Plant

Table 2.1 presents the description of the CFPP in this study, which generated 2x700 Mega Watt (MW) of electricity burning a blended sub-bituminous and bituminous coal. The CFPP burns a total of 2x6,000 Metric Tons (MT) of coal on a daily basis and is equipped separately with dust and gaseous emission control consisting of electrostatic precipitator (EP) and flue gas desulfurization (FGD) unit, respectively.

Table 2.1: Description of the CFPP.

Type of coal	non-local coal, sub-bituminous and bituminous
CFPP capacity (MW)	2 x 700 MW
Total amount of coal burn per day (MT)	2 x 6,000
Number of stack (unit)	1
Stack height (meter, m)	155
Stack diameter (m)	21
Air pollution control (APC) system	EP and FGD

2.2 Sampling and Analysis

A grab sample of FC, BA and FA was taken from the CFPP. The FC, BA and FA were collected at the coal feeder, submerged chain conveyor of the furnace and electrostatic precipitation unit, respectively. Approximately 2.0 kilogram (kg) of material was collected and grinded to fine powder form of 200 micro meters (μm) in size, homogenized and air dried for about 48 hours in an air circulation oven at 110 degree Celsius (^{O}C) in the laboratory and kept in polyethylene (PE) bags. Then, approximately 500 gram (g) of each sample was sealed and kept for a period of thirty days before radioactive counting for

Uranium and Thorium was performed in order to attain the radioactive equilibrium as well as to eradicate ^{222}Rn lost.

The radioactivity content of ^{238}U and ^{232}Th in the sample were measured by instrumental neutron activation analysis or INAA combined with gamma spectrometry system. The Standard Reference Material (SRM) i.e. IAEA 312 for ^{226}Ra, Thorium and Uranium in soil and IAEA 313 for ^{226}Ra, Thorium and Uranium in stream sediment were used as standards in the analysis. Sample was irradiated for 300 minutes at open pool type 1 MW Triga-Mark research reactor of Malaysia Nuclear Agency (Nuclear Malaysia) at a thermal flux of 1.0 x 10^{12} n/cm^2.s and counted sequentially after several days by using a Gamma Spectrometry detection system. The system consist of high performance Germanium (HPGe) detector and Full Width at Half Maximum (FWHM) 1.66 keV for the 1332 kiloelectron volt (keV) photo peak of Cobalt-60 (^{60}Co) and connected to a Canberra n-type multichannel analyzer. All samples were counted twice with peak areas determined by computer code GENIE 2000 software. The ^{238}U specific radioactivity content derived from the weighted mean of the activities Neptunium-239 (^{239}Np) at 228 keV and 278 keV. For ^{232}Th, specific radioactivity content derived from the weighted mean of the activities Protactinium-239 (^{233}Pa) at 312 keV. Systematic errors were taken into account for overall uncertainty calculations. The blank sample was also treated following the same procedures where the final radioactivity content in the sample was determined minus from the blank.

3.0 RESULTS AND DISCUSSIONS

3.1 Activity concentration of natural radionuclides

Table 3.1 presents the mean, standard deviation (sd) as well as the range of activity concentration in natural radionuclides ^{238}U and ^{232}Th in FC, BA and FA which showed that mean of ^{238}U in FC, BA and FA was 9.0 $\pm$ 0, 63.7 $\pm$ 2.5 and 77.7 $\pm$ 3.2 Becquerel per kilogram (Bq/kg), respectively. Meanwhile for ^{238}Th, the mean activity concentration in FC, BA and FA was 10.3 $\pm$ 0.6, 77.7 $\pm$ 2.3 and 77.3 $\pm$ 1.5 Bq/kg, respectively. As expected and clearly indicates that FA has the highest ^{238}U activity concentration compared to the other two samples.

Although, this is not markedly observed for ^{232}Th but the activity concentration level in both ashes were much higher that the FC. Both of the ^{238}U and ^{232}Th activity concentration in FA were enriched by seven orders of magnitude compare to FC. The variability of the activity concentration based to the coefficient of variation for both radionuclides in all samples was less than five percent which shown of their variations were consistent with respect to their means.

Table 3.1: Radioactivity level of natural radionuclides in collected sample.

Sample	**^{238}U (Bq/kg)**		**^{232}Th (Bq/kg)**	
	Mean ± sd	**Range**	**Mean ± sd**	**Range**
FC	9.0 ± 0	9.0-9.0	10.3 ± 0.6	10.0-11.0
BA	63.7 ± 2.5	61.0-66.0	71.7 ± 2.3	69.0-73.0
FA	77.7 ± 3.2	74.0-80.0	77.3 ± 1.5	76.0-79.0

Note: FC=Feed coal; BA= Bottom ash; FA=Fly Ash

Uranium is mainly present in the carbonaceous components of sedimentary rocks and accumulates in coal during coalification [6]. It is mainly present in the organic fraction during the early stages of peat and coal formation, whereas thorium is present in the inorganic phases [4]. Thorium belong to the lithophilic elements of Group I which are associated with aluminosilicate minerals and assumed to have been homogenously incorporated into aluminosilicate-dominated fly ash matrix [5]. Thus, it shows that only slightly or no enrichment on the smaller FA particles. Thorium is a typical lithophile element and has a high affinity for oxygen and so is found mainly in oxygen compounds (oxides, silicates, phosphate and carbonates) [4]. The increase in specific activities of natural radionuclides in combustion products compared to the feed coal depends primarily on the inorganic fraction of the coal [10]. It was found out that coal combustion process eliminates organic components, lead to an enhancement of natural radioactivity level in most of the ashes, are much higher than radioactivity level contain in feed coal. Similarly in this study, the ashes showed higher concentration of thorium compared to uranium.

The combustion of coal in CFPP leads to the enhancement concentration of natural radionuclides from FC to ashes. UNSCEAR [9] reported that radionuclides enrichment from coal to ash is approximately one order of magnitude during the combustion process. Overall in our measurements, the concentrations of radionuclides in FA are much higher than BA and FC. The activity concentration of radionuclides in ash occurs principally as most of the carbonaceous matter in coal oxidizes during combustion and the radionuclides become concentrated in the residue mass [11]. In addition, it shows that at post combustion stage, smaller particle size with high surface area (as in the case of FA) of radionuclides does affect the concentration level of the radionuclides. These results agree with the patterns observed from previous studies [2, 3, 4, 6, 7, 8, and 10]. These variations may occur due to the different compositions and origins of the feed coal and the use of different firing systems, furnace design and furnace temperatures [12]. Thus, these radionuclides showed different physicochemical properties resulting from different kind of behaviour and enrichment at the various stages of the combustion process. The concentration or dispersal of radionuclide, like that of any other chemical element, is controlled by its psysicochemical properties in relation to the ambient conditions [13].

3.2 Legal requirements and regulatory control for natural radioactivity from non-nuclear industries in Malaysia.

Nowadays, NORM residues become a growing issue and led to many discussions among public. Many countries have established certain standard to manage residues containing NORM, but according to International Atomic Energy Agency (IAEA) [13], there has not been any international consensus on the regulations of NORM. Therefore, most countries adopt or implement the available regulatory control based on the international practice.

In Malaysia, activities related to atomic energy are enforced under Atomic Energy Licensing Act 1984 (Act 304) [14]. Malaysia has adopted the international standard set by IAEA; IAEA General Safety Requirements Part No. GSR Part 3 (Interim) 2011 for radiation protection in handling NORM activities. The control limit set, for activity concentration of radionuclides of natural origin shall not exceed 1000Bq/kg for each radionuclide in the Uranium and Thorium decay series [15]. Malaysia has existing interim policy on Radioactive Waste Management that cover activities related to residues or waste generated from various industries. Government of Malaysia had gazette the Atomic Energy Licensing (Radioactive Waste Management) Regulations in 2011. This regulation shall apply to all aspects of radioactive waste and waste management arising from medical, industrial and research applications and any other application which may be specified by the Board (Board has power under Act 304). Radioactive waste means *substance or article that contains or is contaminated with radionuclides at activity concentration or activities greater than Clearance Level and for which no use is foreseen* [16]. For radioactive waste management, the control discharge limit of radioactive waste and it is based on Clearance Level stated in Second Schedule of Atomic Energy Licensing (Radioactive Waste Management) Regulations 2011. The Clearance limit set, whereby the activity concentration of radionuclides of the waste not exceeds 1000Bq/kg for both ^{238}U and ^{232}Th [16]. Clearance Level means *the value established establish by the Board and expressed in terms of activity concentration or total activity, at or below which the source of radiation may be released from the control of the Board as specified in Second Schedule* [16]. The Clearance Level status '*released from the control of the Board'* can only be granting by the regulatory authority once all the requirements meet compliance set by the law.

Table 3.2 shows the legislation limit in Malaysia for natural radioactivity level in NORM and its residues as comparison to the value found in FC, BA and FA obtained from this study which is well below the limits.

Table 3.2: Legislation limit in NORM and its residues in Malaysia.

Sample	**Natural radionuclides**	
	^{238}U (Bq/kg)	**^{232}Th (Bq/kg)**
Limit–NORM& its residues	1000	1000
This study		
FC	9.0	10.3
BA	63.7	71.7
FA	71.7	77.3

It showed that the activity concentration of NORM and its residues from CFPP operation are much lower than the regulatory limit set by the legislation. As reported by UNSCEAR [9], the global mean activity concentration of ^{238}U and ^{232}Th in the fly ashes is 200 and 70 Bq/kg, respectively which is higher than value obtained in this study.

3.3 Issues and challenges arise from NORM use in CFPP.

Few issues recognized here but not limited to, involving coal combustion in CFPP. The release of atmospheric discharge to the environment such as small particulates which escape the air pollution control (APC) devices which contain hazardous and radioactive trace elements. It is one of the major factors affecting human respiratory system. Thus, CFPP operators have to find appropriate APC available technique in order to reduce air pollution especially the greenhouse gases. High demands of energy generated from CFPP result in the continuous growth of high volume solid waste materials and will accumulate if no proper action taken. Solid waste generation need an appropriate waste management and details assessment especially on the waste characteristic. The effect of radiological and non-radiological should be tackled accordingly. In addition, appropriate waste disposal facility will be needed for final disposal, if CCBs are failed to reuse or recycle [2]. Due to this, appropriate stakeholders framework involving the waste polluter and relevant stakeholders consists of the public, local authority, regulatory authority and government shall be established and good communication should imposed accordingly. Polluters-pay-concept is suggested to be applied, where the polluter need to pay for the disposal of their waste. The government should imposed compulsory an appropriate allocation amount of money as the financial security for the waste management as a result of any abundance and cease of operation, decommissioning of facility and others set by the regulatory authority [2]. Secondly, coal combustion potentially enhanced environmental radioactivity level due to CCBs such ashes contain higher radionuclides concentrations. Improve coal combustion technique i.e. integrated gasification combined cycle will increase energy generation, reduce carbon dioxide gas emission and lead to smaller amount of trace elements contains natural radionuclides. Thus, adequate radiation protection for workers and public shall be determined by the management. Workers shall be given appropriate radiation detection equipment (i.e. survey meter) and proper personal protective equipment (PPE) as well as periodic health surveillance should be provided [2]. Continuous assessment on environmental and radiological monitoring study shall be done by all related stakeholders. It is recommended to be done at during the stage of plant siting, pre-commissioning, commissioning and decommissioning. This process will sustain the environment especially in the vicinity of CFPP that involved highly populated area. The biggest challenge to one country to overcome these issues is to provide good legislative framework on radioactive waste management with regards to the use of NORM in non-nuclear industries. Several important factors shall be given detailed consideration such as administrative, technical, social impact, continuous awareness and education to the public and environmental in order to have all-rounded legislative framework. Finally, continuous support from the government in term of financial is likely will increase the effectiveness of regulatory control in CFPP operation in order to ensure the safety and security through-out the CFPP lifespan.

4.0 CONCLUSIONS

The results obtained in this study showed that activity concentration level of ^{238}U and ^{232}Th in ashes were much higher that the feed coal due its combustion in the furnace. In addition, both of the ^{238}U and ^{232}Th activity concentration in fly ash were enriched by seven orders of magnitudes compared to the feed coal. The radioactivity levels of Uranium and Thorium in ashes mostly higher in Thorium compare to Uranium. This study found that the natural radioactivity level obtained from typical CFPP operation is in compliance with the national legislation and international practice. The activity concentration of ^{238}U and ^{232}Th in the fly ashes from this study were much lower value than the global value in which the impact of human exposure to these radionuclides is remote.

ACKNOWLEDGEMENTS

The authors like to express their gratitudes to the Atomic Energy Licensing Board (AELB) of Malaysia for the encouragement and continuous support of the study. This paper is part of the on going study and funded by the Ministry of Science, Technology and Innovation (MOSTI)Malaysia through Science Fund Grant No. LPTA0000005.

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