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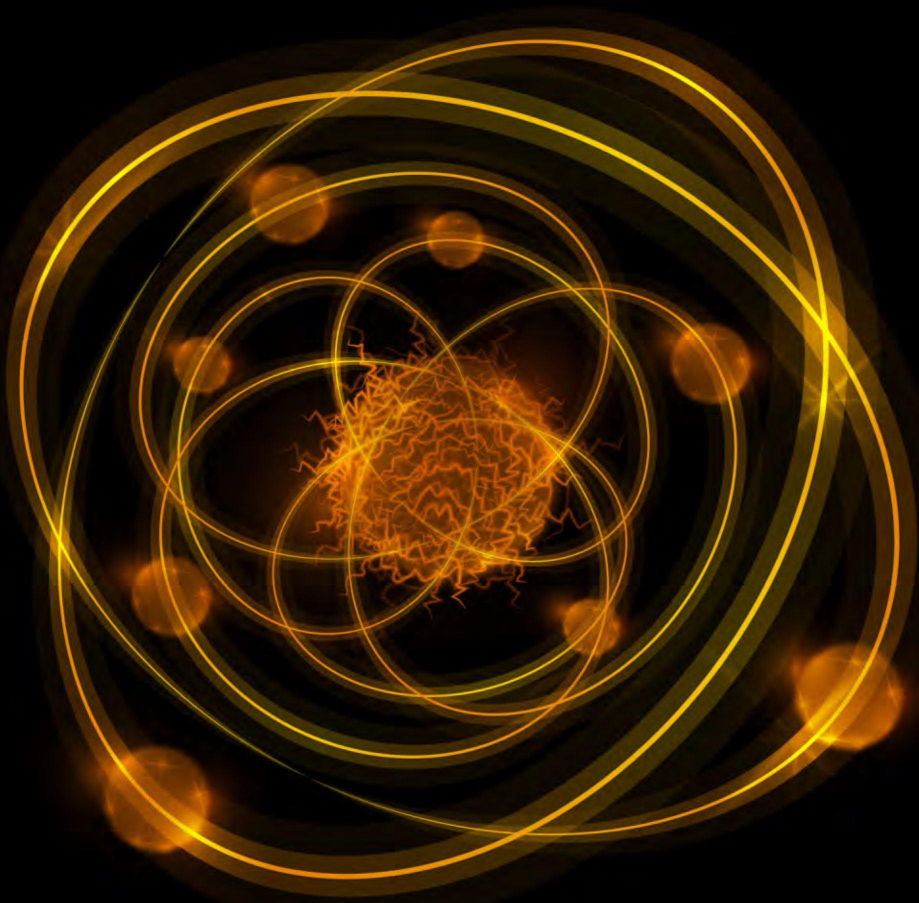

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Establishing a Relationship between Coal Quality and the Enrichment of Radionuclides in Coal Combustion Residues

Uwais Al Qarni Ahmed[1,2*], **Adriaan Joubert**[1]

[1]National Nuclear Regulator, Eco-Park Estate, Centurion, South Africa
[2]CIMERA, Department of Geology, University of Johannesburg, Kingsway Campus, Auckland Park, Johannesburg, South Africa
Email: *uwais78692@yahoo.com

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Abstract

Coal-fired power plants (CFPP) provide approximately 40% of the world's energy demand. Naturally occurring radioactive materials (NORM) contained in coal become enriched in coal combustion residues as a result of the elimination of carbon during combustion. The fly ash and bottom ash produced from CFPP may be significant sources of exposure to naturally occurring radionuclides for the population near the combustion plant or ash dumps. Despite this fact, very few studies have actually addressed the relationship of the NORM enrichment factors and the quality of coal used. This paper aims to relate the quality of coal to the enrichment factors for the radionuclides of interest (K^{40}, Ra^{226}, Th^{232} and Po^{210}) in coal combustion residues from three South African CFPP. The data from other CFPP was also taken into account to establish this correlation. The feedstock coal used in these CFPP is typically low quality, with ash content in the range of 25 - 45 wt%. The radionuclides investigated were determined by gamma spectrometry with the exception of Po^{210}, which was determined by alpha spectrometry. The enrichment factors for the radionuclides of K^{40}, Ra^{226}, Th^{232} and Po^{210} in the fly ash and bottom ash (except Po^{210}) was found to be directly proportional to the quality of coal. That is when the ash percentage increased (coal quality decreased) the enrichment factor decreased. The Po^{210} radionuclide in the bottom ash had an enrichment factor less than one. The relationship between coal quality and enrichment factors for the radionuclides of K^{40}, Ra^{226}, Th^{232} and Po^{210} in both the fly ash and bottom ash (except Po^{210} in the bottom ash) was demonstrated by the following mathematical equation:

$\text{Enrichment Factor} = \dfrac{1}{\text{Ash}(\%)\,\text{in feed coal}}$. This equation may be used as a good indication in obtaining an estimate in determining the enrichment of the mentioned radionuclides in coal combustion products such as fly ash and bottom ash.

Keywords

Radionuclides, Coal, Enrichment Factor, Coal Quality

1. Introduction

Coal-fired power plants (CFPP) provide approximately 40% of the world's energy demand [1]. In developing countries, such as South Africa, coal has a significant role in power generation and contributes to approximately 77% of the country's energy [2]. During coal combustion, the elements in minerals and organic fractions of coal are liberated and distributed into combustion products *i.e.* fly ash, bottom ash and flue gas [3]. The increase in thermal generation capacity and subsequently a deterioration of the quality of coal used has resulted in increased generation of fly ash and bottom ash of varying properties. Naturally occurring radioactive materials (NORM) are among the inorganic constituents that are present in coal. These radioactive materials are enriched in coal combustion products such as fly ash and bottom ash following the combustion of coal [4]. The installation of ash collectors like cyclones, electrostatic separators and bag filters significantly reduces the emission of radionuclides to the atmosphere [5]. In contrast, the treatment and disposal of power plant ash continues to be problematic, particularly in South Africa where the coal has an inherent high ash content (low quality), typically between 25% - 50%, and thus a significant amount of ash is generated.

The interest in measuring NORM concentrations in coal and resulting combustion residues (such as fly ash and bottom ash) is due to the awareness of health hazards and environmental pollution [6]. The fly ash and bottom ash produced from CFPP are significant sources of exposure for the population near the plant to naturally occurring radionuclides [7] [8]. The naturally occurring radionuclides, particularly K^{40}, Ra^{226}, Th^{232} and Po^{210} released by these plants pose a potential health hazard [9]. Due to its short half-life and the highly energetic cell-disrupting alpha particles (5.3 MeV) emitted during its decay process, Po^{210} is considered a major health hazard [10]. Once Po^{210} from the air is assimilated into flora and fauna, it may be bio-concentrated in the food chain and, thus, poses further threat to human health [11].

The concentration of most radioactive elements in solid combustion wastes, such as fly ash and bottom ash, will be multiple times higher than the concentration in the original coal [12]. The enrichment factors of certain radionuclides can sometimes be a few folds or even several magnitudes, relative to the feed coals [13]. The number of natural radionuclides discharged into the atmosphere via the ash produced from a CFPP depends on the ash content of coal, the temperature of combustion, partitioning between fly ash and bottom ash, and the efficiency of control devices [5] [14].

Several studies wherein coal enrichment factors with respect to certain radionuclides in coal and coal combustion products were conducted by other authors [4] [5] [10] [15]. However, none of these studies actually relates the quality of coal to the enrichment of radionuclides (enrichment factors) in the fly ash and bottom ash. This attribute of coal quality is one that every CFPP is aware of in terms of the coal being burnt. Therefore, knowing the relationship between coal quality and enrichment factors may enable us to determine the concentration of radionuclides in the combustion products prior to the combustion of coal. This may prevent scenarios such as those in China whereby it was found that some coal ash was too radioactive for reuse in building materials [16]. For this very reason, radioactivity in coal and coal combustion residues' uses are being limited [17].

It is hypothesized that the relationship between the concentration of radionuclides in coal and the combustion products *i.e.* fly ash and bottom ash, is closely related to the quality of coal. Hence, the objectives of this study are to evaluate the ash percentage content (hence quality) of 3 different coals fed to 3 different CFPP and evaluate the enrichment factors in the fly ash and bottom ash (in comparison to other studies as well). As a result, thereof, a mathematical correlation between coal quality and the enrichment factor of radionuclides in coal (in terms of an equation) is to be established, since it does not exist in present-day literature.

2. Materials and Methods

2.1. Sample Collection

The feed coal to 3 different CFPP from 3 different coalfields in South Africa was used in this study. The coal mines (**Figure 1**) are in close proximity to the CFPP. Monthly composite samples were sampled on alternate days over a period of 3 months. The three-month period of sampling assured that the samples were representative of the feed coal to each CFPP.

The proximate and ultimate analysis for the feed coal provided for these samples by the power supplier indicated that, over the three months, the feed to the plant was consistent. On the same days as the feed coal was sampled, the fly ash (from the hoppers) and bottom ash (from the boiler) from the 3 CFPP were also sampled. This ensured that the fly ash and bottom ash were indeed products of the sampled feed coal. The gross samples were then air-dried, milled (coal and bottom ash) and carefully split in accordance with ISO recommendations in order to obtain a representative sample of particle size < 250 um prior to chemical analyses.

2.2. Sample Processing and Analyses

The feed coal samples were supplied together with the proximate data by the power utility and were analysed in accordance with ISO 18283:2006 [19] and

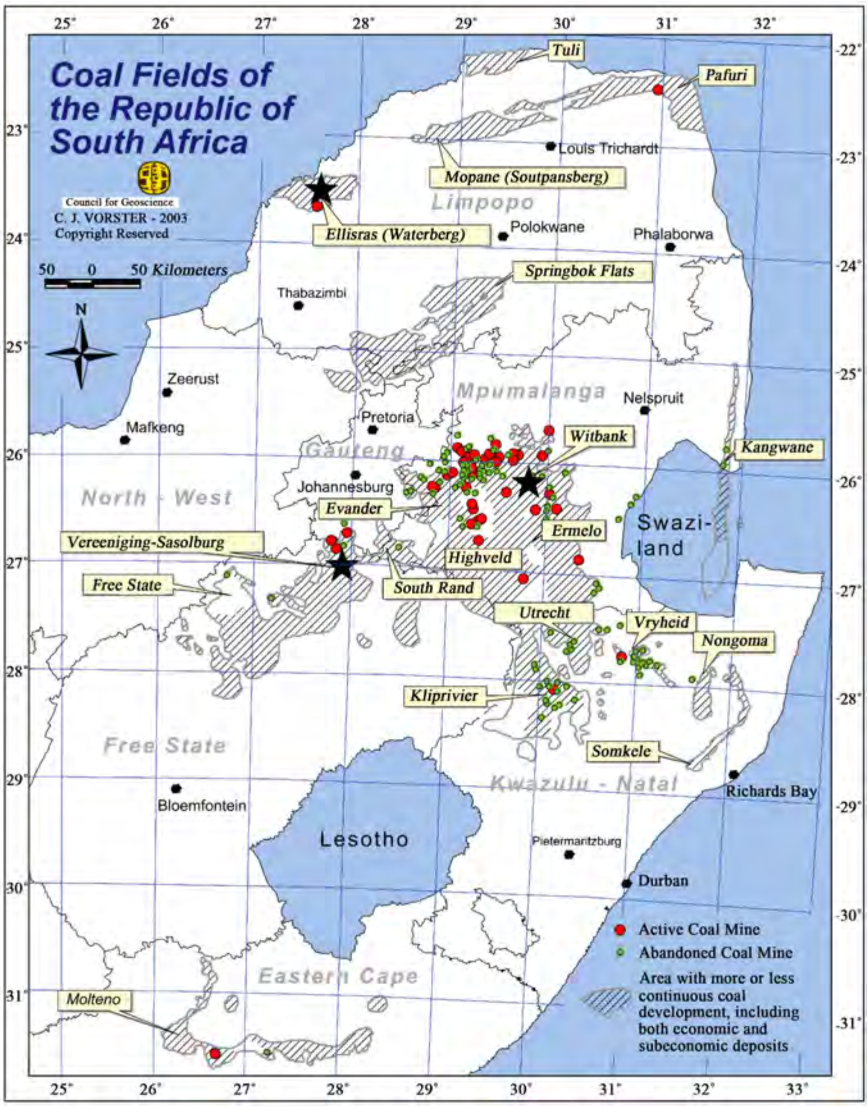

Figure 1. Coalfields of the Republic of South Africa [18]. The location of the CFPP are indicated.

ISO 13909:2001 [20]. Therefore, the quality of coal was concluded based on the ash yield and classified accordingly as indicated in **Table 1**.

In order to determine the radionuclides of interest (K^{40}, Ra^{226}, Th^{232} and Po^{210}) in the samples, two routes were followed. For K^{40}, Ra^{226} and Th^{232}, gamma spectrometry was conducted; for Po^{210} alpha spectrometry was conducted.

For gamma spectrometry, the coal and ash samples were dried for 24 hours in an air-circulation oven at 110˚C. Samples were further pulverized to obtain a fine powder and sieved for homogeneity. Thereafter, 100 g of each sample was placed in plastic containers of 6.5 cm diameter × 7.5 cm height, and sealed to make them airtight. The samples were left for a period of 1 month in a designated laboratory cupboard to ascertain the establishment of secular equilibrium between Ra^{226} and Th^{228} with their progeny and to prevent Rn loss. The specific radionuclides of the samples—*i.e.* K^{40}, Ra^{226} and Th^{232}—were determined using a

Table 1. Ash categorisation of the feed coals.

CFPP location	Ash Yield	Ash Class Category (According to ISO 11760:2005) [21]
Witbank	25.27% (≥20 and <30)	Moderately high ash
Waterberg	32.84% (≥30 and <50)	High ash
Sasolburg	40.85% (≥30 and <50)	High ash

high-resolution, p-type coaxial HPGe γ-ray spectrometer shielded by cylindrical lead. The detector relative efficiency was 28.2% and energy resolution of 1.67 keV-FWHM at the 1.33 MeV peak of Co^{60}. A cylindrical multi-nuclide source was used for detector energy calibration and efficiency determination [22]. The measured detection efficiencies were fitted by using a polynomial fitting function, as described by Khandaker *et al.* [23], and the fitted efficiencies were used in activity determination of the samples. The minimum detectable activity (MDA) of the γ-ray measurement system at 95% confidence level was calculated according to the procedure by Khandaker *et al.* [23]. Each sample was counted for 86,400 s, and similarly for background counts, in order to obtain the net activity.

The same method used by Sahu *et al.* [10] was used to determine the Po^{210} using alpha spectrometry. Samples (5 g of each) were first digested with HNO_3 and 4 N HCl sequentially. The acids were evaporated to near dryness and made up to 80 mL by adding 1 N HCl with ascorbic acid to reduce interfering Fe(III). Tracer activity 3.0 Bq/ml including Po^{209} was added to the aliquots. Then silver planchets were submerged into the solutions and were kept at a temperature of about 85˚C for 7 h with continuous stirring. The planchets thereafter were dried under infra-red lamp and alpha activities were determined in the alpha spectrometer. Samples were counted in an eight-chamber integrated alpha spectrometry system equipped with ion-implanted Si-charged-particle detectors, with an active detector-surface area of 450 mm^2, and a source-to-detector distance of approximately 10 mm. Samples and blanks were counted for nominally 250,000 s. Background measurements were made immediately prior to the measurements.

3. Results and Discussion

Table 2 presents the quality of coal used in the three CFPP and their coal and resultant coal combustion residues' radionuclide concentration in comparison with other studies [24] [25].

3.1. Radionuclide Concentrations in Coal, Fly Ash and Bottom Ash

The quality of coal used in South Africa varies between the CFPP [26], and is generally of low grade. The values obtained here are comparable to those reported by [27] for coals typically supplied to these three CFPP's. The quality of coal used can be classified as moderately high ash to high ash coal according to

Table 2. Ash categorisation of the feed coals and radionuclide concentrations in coal, fly ash and bottom ash for the three CFPP in comparison with other studies [24] [25].

Sample		Ash	N	Activity of radionuclide (Bq/kg)			
				K^{40}	Ra^{226}	Th^{232}	Po^{210}
Witbank (Present Study)	Fuel (Coal)	25%	9	88 ± 6	21 ± 1	22 ± 4	46 ± 1
	Fly ash		9	348 ± 8	80± 3	88 ± 6	193± 8
	Bottom ash		9	334 ± 8	63± 3	84 ± 6	41 ± 7
Waterberg (Present Study)	Fuel (Coal)	33%	9	91 ± 3	52 ± 5	21 ± 1	70 ± 5
	Fly ash		9	319 ± 4	166 ± 8	63 ± 6	280 ± 8
	Bottom ash		9	302 ± 5	150 ± 13	82 ± 8	3 ± 1
Sasolburg (Present Study)	Fuel (Coal)	41%	9	110 ± 4	24 ± 5	19 ± 3	83 ± 3
	Fly ash		9	286 ± 4	65± 6	45 ± 6	265 ± 13
	Bottom ash		9	242 ± 13	51± 6	44 ± 2	2 ± 1
Spain: Teruel UPT [25]	Fuel (Coal)	22%	2	66 ± 15	54 ± 3	21 ± 3	65 ± 11
	Fly ash		2	306 ± 13	191 ± 9	74 ± 3	257 ± 30
	Bottom ash		2	235 ± 11	149 ± 6	66 ± 3	57 ± 7
Spain: Litoral UPT [25]	Fuel (Coal)	16%	2	70 ± 16	15 ± 3	13 ± 3	33 ± 4
	Fly ash		2	338 ± 16	107± 4	87 ± 4	300 ± 40
	Bottom ash		2	278 ± 16	81 ± 4	64 ± 4	7 ± 1
Spain: Compostilla II [24]	Fuel (Coal)	37%	2	334 ± 13	34 ± 3	33 ± 3	118 ± 40
	Fly ash		2	1109 ± 49	94 ± 4	93 ± 4	416 ± 57
	Bottom ash		2	1077 ± 48	86 ± 2	89 ± 3	24 ± 12
Spain: Teruel [24]	Fuel (Coal)	28%	2	77 ± 32	56 ± 5	21 ± 2	65 ± 11
	Fly ash		2	310 ± 14	190 ± 2	74 ± 2	257 ±30
	Bottom ash		2	238 ± 13	149 ± 5	66 ± 3	57 ± 7
Spain: Litoral [24]	Fuel (Coal)	15%	4	62 ± 30	18 ± 4	20 ± 6	47 ± 20
	Fly ash		4	250 ± 144	158 ± 48	154 ± 53	505 ± 321
	Bottom ash		4	224 ± 105	144 ± 55	138 ± 61	9 ± 4

ISO [21]. For all of the CFPP samples, the radionuclide concentrations in the coals are in close proximity with the world averages (K^{40}, 4 - 785 Bq/kg; Ra^{226}, 1 - 206 Bq/kg ; Th^{232}, 1 - 170 Bq/kg; and Po^{210}, 3 - 52 Bq/kg) as indicated by the IAEA [28].

Although the radionuclide concentration in coal may be in line with the world averages, the concentrations of these are expected to be multiple times higher in the coal combustion products (fly ash and bottom ash) than the concentration in the original coal [12] and are commonly termed as the enrichment factor or enrichment ratio. The enrichment ratio is the concentration of the nuclide in ash to its concentration in the feed coal [10]. The results of radionuclides in the fly ash and bottom ash from **Table 2** certainly indicate the enrichment of radionuclides from the coal to the fly ash and bottom ash. The activity levels of K^{40}, Ra^{226}, and Th^{232} are very similar to each other in both the fly ash and bottom ash samples for all three plants in the current study. These correlate with the results from the two other studies tabulated. However, there are significant differences between the Po^{210} concentrations in the fly ash and bottom ash. This is because most of the Po^{210} is vaporized during combustion in the boiler [25]. This phenomenon will be expanded on in the next section. The K^{40}, Ra^{226}, Th^{232} and Po^{210}activity concentration levels for the fly ash and bottom ash for this study are in close proximity to the values reported by Mora *et al.* [25] and Baeza *et al.* [24], which are presented in **Table 2**. The average activity levels given in the UNSCEAR report [29] for fly ash are 265 Bq/kg for K^{40} , 240 Bq/kg for Ra^{226} , 70 Bq/kg for Th^{232} and 1700 Bq/kg for Po^{210}.

3.2. Enrichment Factors and Coal Quality

The enrichment factor for each radionuclide for the fly ash and bottom ash for each of the CFPP is reported in **Table 3** together with the results from other studies [24] [25].

Table 3. Coal quality and their respective enrichment factors for the three CFPP in comparison with other studies [24] [25].

Sample		Ash	Enrichment Factor			
			K^{40}	Ra^{226}	Th^{232}	Po^{210}
Witbank	Fly ash	25%	3.9 ± 0.8	3.8 ± 0.3	4 ± 0.7	4.1 ± 0.1
	Bottom ash		3.8 ± 0.8	3 ± 0.3	3.8 ± 0.7	0.9 ± 0.1
Waterberg	Fly ash	33%	3.5 ± 0.8	3.1 ± 0.6	3 ± 0.2	4 ± 0.6
	Bottom ash		3.3 ± 0.6	2.9 ± 0.4	3.9 ± 0.1	0
Sasolburg	Fly ash	41%	2.6 ± 1	2.7 ± 0.8	2.4 ± 0.5	3.2 ± 0.2
	Bottom ash		2.2 ± 0.3	2.1 ± 0.8	2.3 ± 0.7	0.0
Spain: Teruel UPT [25]	Fly ash	22%	4.6 ± 0.9	3.5 ± 0.3	3.5 ± 0.2	4 ± 0.3
	Bottom ash		3.6 ± 0.7	2.8 ± 0.7	3.1 ± 0.1	0.9 ± 0.6
Spain: Litoral UPT[25]	Fly ash	16%	4.8 ± 0.1	7.1 ± 0.8	6.7 ± 0.1	9 ± 0.1
	Bottom ash		4 ± 0.1	5.4 ± 0.8	4.9 ± 0.1	0.2 ± 0.3
Spain: Compostilla II [24]	Fly ash	37%	3.3 ± 0.2	2.8 ± 0.3	2.8 ± 0.3	4 ± 1
	Bottom ash		3.2 ± 0.2	2.5 ± 0.2	2.7 ± 0.3	0.2 ± 0.1
Spain: Teruel [24]	Fly ash	28%	4 ± 2	3.4 ± 0.3	3.5 ± 0.3	4 ± 1
	Bottom ash		3 ± 1	2.7 ± 0.3	3.1 ± 0.3	1 ± 0.2
Spain: Litoral [24]	Fly ash	15%	4 ± 3	9 ± 3	8 ± 3	11 ± 8
	Bottom ash		4 ± 2	8 ± 3	7 ± 4	0.2 ± 0.1

3.2.1. Fly Ash and Coal Quality

In most cases, the enrichment factors in this study were approximately 2 - 4 times higher in the ashes than in the feed coal. This is commonly observed in other studies [10] [30]. The enrichment factor for all the samples were higher for the fly ash compared to the bottom ash. This is because the radionuclides concentrate on the smaller fly ash particles that have a larger surface area-to-volume ratio and the hot flu gases cool down on their way to stack [31]. An exception to the enrichment of 2 - 4 times is that of Po^{210} in the bottom ash.

For the present study, the highest enrichment factor was for Po^{210} in the Witbank fly ash. The enrichment of Po^{210} in the fly ash is 2 - 4 times higher. The lowest enrichment factor in the fly ash was Th^{232} that was for the Sasolburg coal. In bottom ash the highest enrichment factor of 3.90 was observed for Th^{232} in the Waterberg coal and the lowest enrichment factor was 2.13 for Ra^{226} in the Sasolburg coal. In all cases, the fly ash had higher enrichment factors for all the radionuclides when compared to bottom ash, which is consistent with previous studies conducted [10] [32] [33].

The enrichment factor for each radionuclide determined in the fly ash (**Table 3**) was plotted against the ash content of the coal in order to establish a correlation (**Figures 2-6**).

It is apparent (from **Figures 2-5**) that the enrichment factor for K^{40}, Ra^{226}, Th^{232} and Po^{210} in the fly ash samples decreased as the %ash increased. This indicates that the enrichment factors for these specific radionuclides in these coals are directly proportional to coal quality as expressed by ash. A strong correlation (indicated by the R^2 values which are approximately > 0.7 in **Figures 2-5**) was obtained for the concentration the K^{40}, Ra^{226} and Th^{232} radionuclides in relation to the quality of coal, whilst the correlation for Po^{210} was moderately strong *i.e.* $R^2 = 0.62634$.

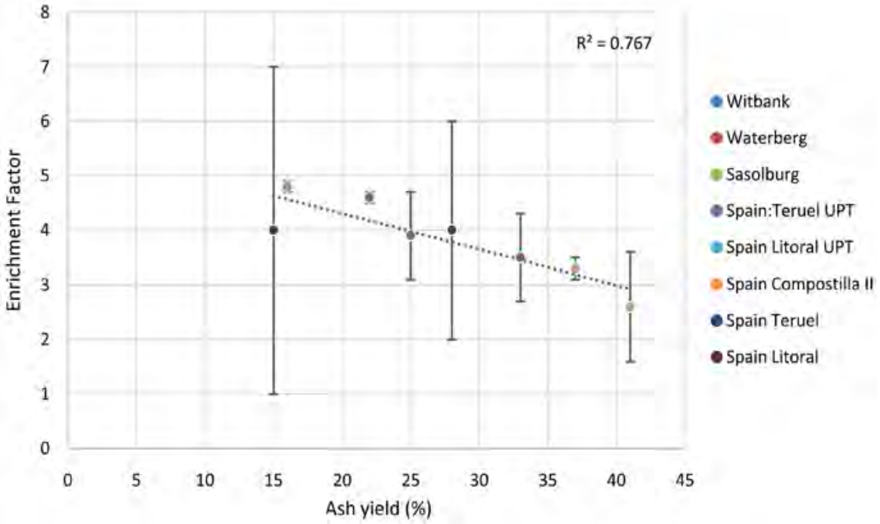

Figure 2. Correlation for K^{40} in the fly ash for the enrichment factor and the ash content in various coal samples.

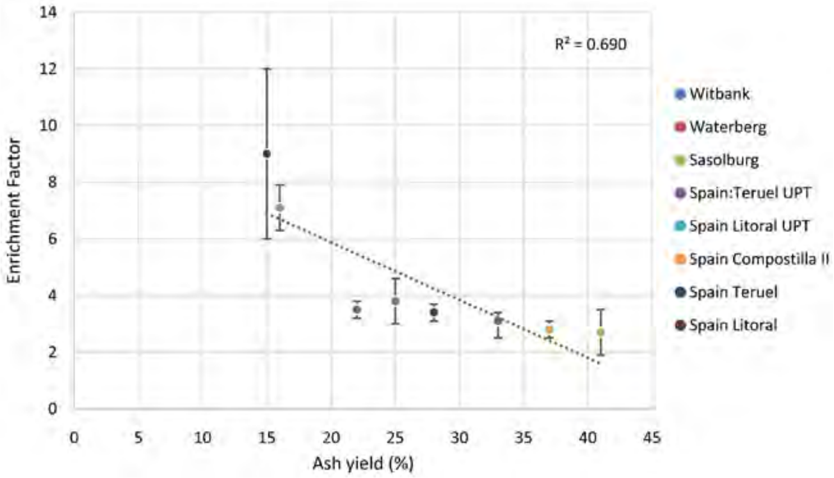

Figure 3. Correlation for Ra^{226} in the fly ash for the enrichment factor and the ash content in various coal samples.

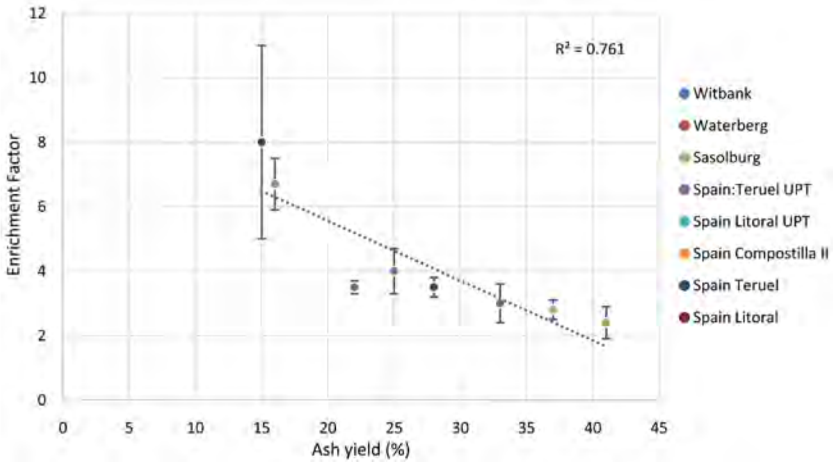

Figure 4. Correlation for Th^{232} in the fly ash for the enrichment factor and the ash content in various coal samples.

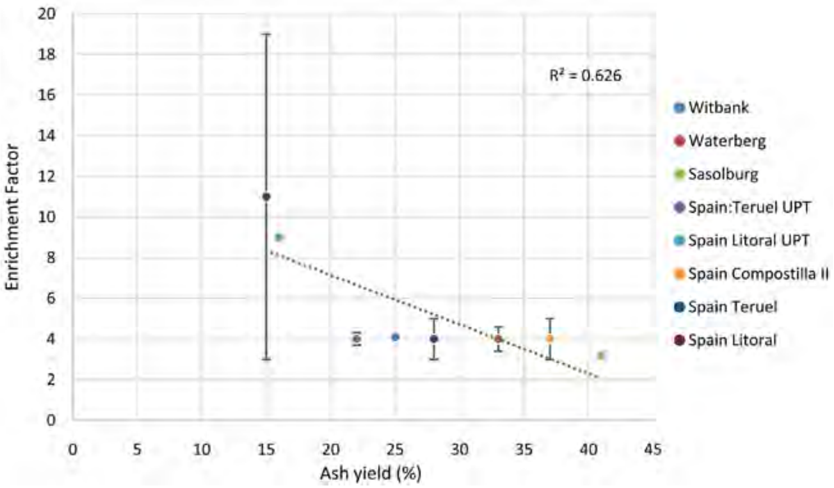

Figure 5. Correlation for Po^{210} in the fly ash for the enrichment factor and the ash content in various coal samples.

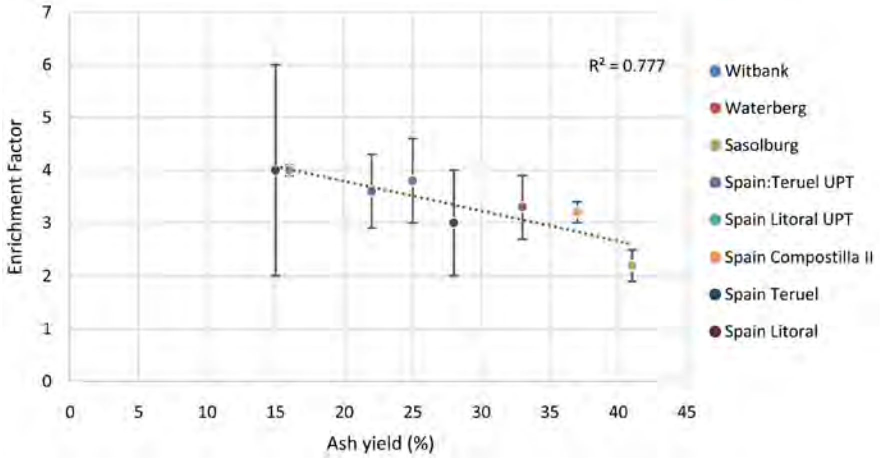

Figure 6. Correlation for K^{40} in the bottom ash for the enrichment factor and the ash content in various coal samples.

3.2.2. Bottom Ash and Coal Quality

The enrichment factors for each radionuclide in the bottom ash (**Table 3**) was correlated with the ash content of coal as shown in **Figures 6-9**.

In terms of the bottom ash, the enrichment factors also seems to be directly proportional to the ash content. However, a strong correlation is only present in K^{40} ($R^2 = 0.7777$) whereas the Ra^{226} and Th^{232} present only a moderately strong correlation (as opposed to strong correlations in the fly ash). It is interesting to note that this relationship is applicable to all the radionuclides (K^{40}, Ra^{226} and Th^{232}) except Po^{210}. In fact, Po^{210} exhibits no correlation ($R^2 = 0.0854$) and the enrichment factors for Po^{210} in the bottom ash for all the CFPP is less than unity, and therefore indicates that Po^{210} is depleted from the feed coal to bottom ash. This is because, as mentioned, the Po compounds are associated with sulphide minerals and are mostly volatilized during the combustion process; they later condense onto smaller fly ash particles which have larger specific areas and thus their levels get elevated in the fine fly ash fraction [10].

The results indicate that the enrichment factors for the radionuclides K^{40}, Ra^{226}, Th^{232} and Po^{210} in the fly ash and bottom ash are in close proximity with each other (except Po^{210} in the bottom ash). The results indicate that the enrichment factor is approximately 4, 3 and 2.4 when the ash content is 25%, 33% and 41% respectively. Mora *et al.* [25] observed that the enrichment factors in fly ash in relation to coal were 6.1 and 4.6 when the ash content in coals was 16% and 22%respectively. Lauer *et al.* [4] conclude that a 7 - 10 fold enrichment is expected from the elimination of carbon during combustion from coals containing 10% - 15% ash content, which is typical for low ash U.S coals. This leads to the following equation to determine the enrichment factor for radionuclides in coal prior to the combustion of the coal by simply considering the % ash content:

$$\text{Enrichment Factor} = \frac{1}{\text{Ash}(\%)\,\text{in feed coal}} \tag{1}$$

Although not precise, the equation may be used as an excellent estimate (or

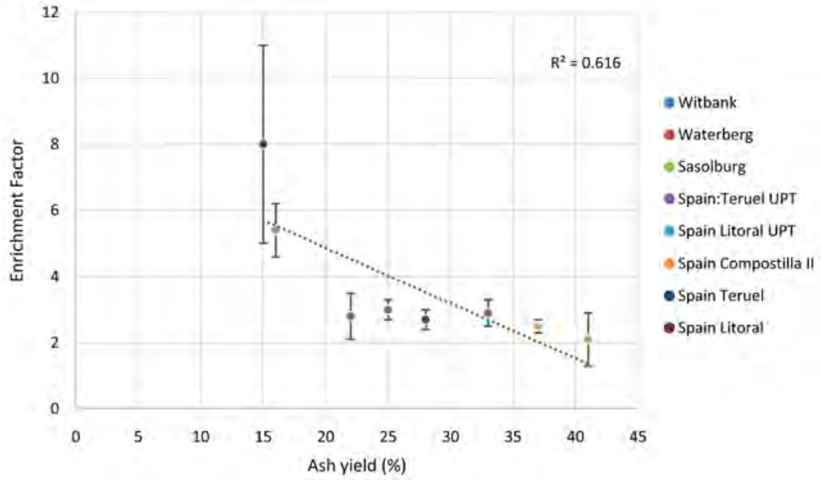

Figure 7. Correlation for Ra^{226} in the bottom ash for the enrichment factor and the ash content in various coal samples.

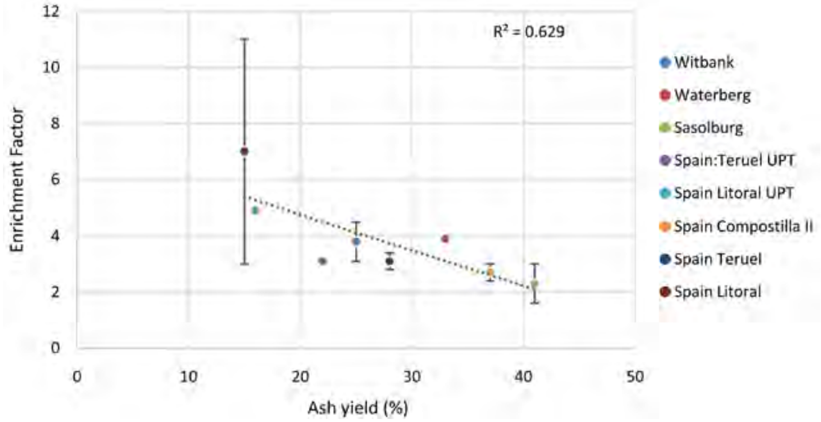

Figure 8. Correlation for Th^{232} in the bottom ash for the enrichment factor and the ash content in various coal samples.

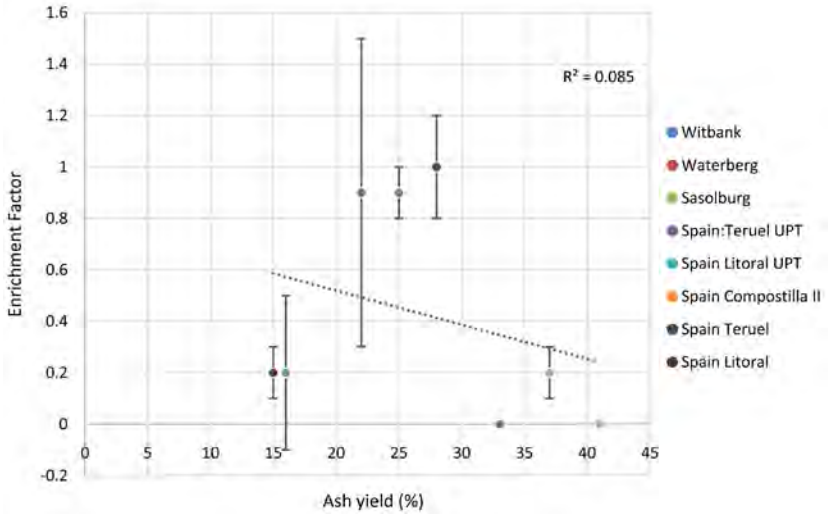

Figure 9. Correlation for Po^{210} in the bottom ash for the enrichment factor and the ash content in various coal samples.

rule of thumb) in determining the enrichment of radionuclides such as K^{40}, Ra^{226} and Th^{232} in coal combustion products.

4. Conclusions

When coal is burnt, the radionuclide concentration increases multiple times from the coal to the fly ash and bottom ash. The relationship between this enrichment of radionuclides (in the fly ash and bottom ash) and the quality of coal was investigated using samples from 3 different CFPP in South Africa in comparison with other studies.

It was found that the coals used in these CFPP were typically low quality coal, ranging between 25% - 45% ash content. The enrichment factor for the radionuclides K^{40}, Ra^{226}, Th^{232} and Po^{210} were 2 - 4 times higher in the ashes than in the feed coal (with the exception of Po^{210} in the bottom ash). The enrichment factor for the radionuclides of K^{40}, Ra^{226}, Th^{232} and Po^{210} and in the fly ash was found to be directly proportional to the ash content; *i.e.* when the ash % increased (coal quality decreases) the enrichment factor decreased. The relationship for the enrichment factor for the radionuclides K^{40}, Ra^{226} and Th^{232} in the bottom ash also showed the same directly proportional relationship to coal quality *i.e.* as the coal quality decreased the enrichment factor also decreased. However, this was not obeyed for the Po^{210} radionuclide, which had an enrichment factor less than unity in the bottom ash samples.

The relationship between coal quality and enrichment factors for the radionuclides of K^{40}, Ra^{226}, Th^{232}, and Po^{210} in both the fly ash and bottom ash (except Po^{210} in the bottom ash) was demonstrated by using the following mathematical equation:

$$\text{Enrichment Factor} = \frac{1}{\text{Ash}(\%)\text{in feed coal}} \tag{1}$$

This equation may be used a good indication in obtaining a ball park figure in determining the enrichment of radionuclides such as K^{40}, Ra^{226} and Th^{232} in coal combustion products such as fly ash and bottom ash.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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Safe Controlled Storage of SVBR-100 Spent Nuclear Fuel in the Extended-Range Future

Georgy Toshinsky[1,2*], Sergey Grigoriev[2], Alexander Dedul[2], Oleg Komlev[2], Ivan Tormyshev[1]

[1]JSC "SSC RF-IPPE", Obninsk, Russia
[2]JSC "AKME-Engineering", Moscow, Russia
Email: *toshinsky@ippe.ru

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Abstract

Experience of operating reactor facilities (RF) with lead-bismuth coolant (LBC) has revealed that it is possible to perform safe refueling in short terms if the whole core is replaced and a kit of the special refueling equipment is used. However, comparing with RFs of nuclear submarines (NS), in which at the moment of performance of refueling the residual heat release is small, at RF SVBR-100 in a month after the reactor has been shut down, at the moment of performance of refueling the residual heat release is about 500 kW. Therefore, it is required to place the spent removable unit (SRU) with spent fuel subassemblies (SFSA) into the temporal storage tank (TST) filled with liquid LBC, in which the conditions for coolant natural circulation (NC) and heat removal via the tank vessel to the water cooling system are provided. After the residual heat release has been lowered to the level allowing transportation of the TST with SRU in the transporting-package container (TPC), it is proposed to consider a variant of TPCs transportation to the special site. On that site after the SRU has been reloaded into the long storage tank (LST) filled with quickly solidifying liquid lead, the TPCs can be stored during the necessary period. Thus, the controlled storage of LSTs is realized during several decades untill the time when SNF reprocessing and NFC closing are becoming economically expedient. On that storage, the four safety barriers are formed on the way of the release of radioactive products into the environment, namely: fuel matrix, fuel element cladding, solid lead and steel casing of the LST.

Keywords

Spent Nuclear Fuel, Controlled Storage, Lead-Bismuth Coolant, Safety Barriers, Radioactive Waste

1. Introduction

The design of reactor SVBR-100 allows its operation in such a way that without changes in design that reactor can operate using different types of fuel both in an open nuclear fuel cycle (NFC) with postponed reprocessing of spent nuclear fuel (SNF) and in the closed NFC [1]. The option for the fuel and NFC type is determined by economical expedience and technological mastering of the fuel and fuel cycle.

It can be expected that at the nearest first stage of implementation of fast reactors (FR) in the nuclear power (NP) in conditions of low costs of natural uranium and services on its enrichment, the operation of reactor SVBR-100 with use of oxide uranium fuel in an open NFC with postponed reprocessing of SNF will be more economically expedient than that in the closed NFC despite the fact of significantly higher (approximately by a factor of 2 - 2.5 as compared with that of VVER-1000 reactors) specific (per 1 kWh) consumption of natural uranium. As an example, it was demonstrated by the results of conceptual project of the modular NPP with reactors SVBR-75/100 cooled by heavy liquid-metal coolant (HLMC) lead-bismuth alloy (2002) [2]. Use of that fuel enriched in less than 20% with postponed reprocessing of SNF is also the most expedient for the reasons of nonproliferation that is important when those reactors are exported to developing countries.

Of course, when the costs of natural uranium and the cost of SNF storage increase much while the SNF reprocessing and its cost are decreasing, the changeover to the closed NFC will become economically expedient. At that point, reactors SVBR-100 will operate in a mode of fuel self-providing with a breeding ratio (BR) slightly exceeding one. Launching of FRs using uranium fuel with further changeover to the closed NFC (as it is shown in [3]) is more economically expedient than launching of FRs using mixed uranium-plutonium fuel with the use of plutonium extracted upon reprocessing of thermal reactors (TR) SNF.

At that point, there is no necessity to construct the expensive factories on reprocessing of TR SNF and manufacturing of uranium-plutonium fuel prior to launching of FRs[1].

However, operation of FRs with postponed reprocessing of SNF will require to find the solution to the issue of safe controlled and economic storage of the SNF with retention of the opportunity to use it upon NFC closing.

2. SNF Management Problems

Up to nowadays, the problem of NPP SNF management has not been solved in full in any country. That is a challenge to the current NP and is a cause of concern for a certain part of the population and national governments because of high long-lived radioactivity accumulated in the SNF. That fact and some other reasons, namely: safety, economics, nonproliferation, are hampering NP devel-

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[1] In Japan the cost of Rokkasho Reprocessing Plant (RRP) with an annual capacity to reprocess 800 tonnes of SNF is $25 billion. This is equivalent to the capacity to reprocess spent nuclear fuel from 40 PWR type reactors, the capacity of each reactor is about 1 GWe.

opment. To hasten the process of finding the solution to that postponed problem, the following ethical principle has been put forward: "*Radioactive waste shall be managed in such a way that will not impose undue burdens on future generations*" [4].

Though the principal scientific ways for finding the solution to that problem are clear in general terms, the practical realization of the highlighted problem is requiring performance of the corresponding R&D, demonstration of the entire cycle of SNF management and harmonization of national strategy and policy in the area of nuclear fuel cycle in the world.

That is conditioned by the fact that now the nuclear community does not possess the necessary knowledge, experience and opportunities for finding the safe and economically efficient solution to that complex problem. In the process of development of SNF management methods, it should be accounted that at the existing technologies of SNF reprocessing, two first defense barriers are damaged, namely: fuel matrix and fuel element cladding. And the fuel, in which the huge amount of radioactivity has been accumulated, is converted from a solid state into liquid one (or gas phase required for gas-fluoride technology) when radioactivity release into the environment is much possible.

When the most developed water-chemical reprocessing of SNF is used, arising of the accidents caused by the emergence of over-critical configuration or concentration of fissile nuclei cannot be excluded as water is the best moderator of neutrons. The Tokaimura Criticality Event occurred on September 30, 1999 in Japan was among those accidents. In 1957 at "Mayak" (a plutonium production site for nuclear weapons and nuclear fuel reprocessing plant, Chelyabinsk Region, Russia) the catastrophic exhaust of radioactivity occurred in the repository for liquid radioactive wastes received from reprocessing of SNF of weapon uranium-graphite reactors. Moreover, in the process of SNF reprocessing, it is more difficult to perform the account and control of nuclear fissile materials (NFM) that is heightening the risk of their unauthorized proliferation.

At present, there are three approaches used to find the solution to SNF management.

2.1. The SNF Is the NP Radioactive Waste

The SNF is considered as the NP radioactive waste that must be finally buried in deep geological formations, where SNF radioactive substances can be securely isolated for hundreds of thousands of years.

However, in that case, the following conflict appears, namely: a desire to bury the SNF as deep as possible for its maximal safe isolation from the biosphere and opportunity to remove the residual heat. Heat removal via thick rock layers by thermal conductivity will take a very long time before the residual heat is reduced to the permitted level eliminating the necessity for temperature monitoring and organized heat removal. Otherwise, it will be necessary to provide heat removal into the atmosphere by organizing a multiple branch system of air-supply and exhaust ducts. For a long time, it will be necessary to provide temperature monitoring of storage packages and air flow rate in the air ducts, via which the radionuclides can be released in the biosphere in an event of degradation of protection barriers. It will be also required to organize reliable accounting, control and physical protection of SNF packages with NFMs.

During the last 30 - 40 years that approach has been taken up in the USA. The Yucca Mountain Nuclear Waste Repository was built on the site located in the mountains in the State of Nevada. That geological storage facility was provided for final burial of packages with spent fuel subassemblies (SFSA) after they had been cooled for many years in the plant spent nuclear fuel storages (SNFS) purposed to reduce radioactivity and corresponding heat release. The SFSAs should be located in special cans equipped by multi-barrier shielding.

The cost of that repository equipped with ventilation channels, and which required 40 miles of tunnels to store 70,000 metric tons of SNF was estimated as $13.5 billion [5]. So, a special fund was established to build the Yucca Mountain project [6]. All nuclear power plants had to pay a tax of 0.1 cent on each kilowatt-hour of produced electricity. By estimations, the cost for 100 years of operation will reach $90 billion. The repository square is 2×2 km^2.

Currently, it is considered that the highlighted approach does not comply with requirements of large-scale NP development and implementation of the Yucca Mountain repository is held up.

The main reasons for that are presumed to be as follows:

First, as a matter of fact, the NP radioactive waste (RAW) are fission products, which mass content in the SNF is as follows: about 5% for thermal reactors (TR), about 10% for fast reactors (FR). Others are as follows: uranium-238 (about 90%), which is a nuclear raw material for producing of plutonium-239 for the future NP based on FRs operating in the closed NFC and eliminating the problem of NP fuel providing for centuries (within the reasonable scales); built up plutonium-239 along with its higher isotopes, and unburned out uranium-235 that is a nuclear fuel, which will be used upon NFC closing; small amount of transuranium elements (so-called minor actinides: isotopes of neptunium, americium, curium), which are generated while capturing of neutrons without fission by uranium-235 and plutonium-239 and their daughter nuclei.

Thus, general application of that approach can doom the NP to use as fuel only the rare isotope of uranium-235 (that is about 0.7% in natural uranium), which economically available resources can be expired in the nearest 50 - 100 years upon the high paces of development of the NP based on existing TRs operating in the open NFC. NFC closing for TRs, in which due to degradation of nuclear properties of plutonium a single recycle of plutonium is possible, is reducing the specific consumption of natural uranium by a little without finding the solution to the other problems of SNF management. Such fuel cycle is realized, e.g. in France. However, at this point, the cost of mixed oxide uranium-plutonium fuel (MOX fuel) is by a factor of 4.5 times higher than the cost of uranium fuel [7]. Thus, the cost of produced electricity is heightened approximately by 20% [7].

In the USA one of major reasons for final burial of the SNF was fear of unauthorized proliferation of plutonium in an event of its releasing from SNF in the process of reprocessing in the closed NFC.

Second, though the content of minor actinides (MA) in the SNF is low, it is MA but not fission products, which are determining the long-lived radioactivity of SNF, as their half-life is hundreds of thousands of years (against dozens of years for the most of fission products). For such long storage period, the geologists cannot assure the absence of tectonic shifts in the earth's crust, which can cause destruction of protection barriers in the SNF packages and release of radioactive products into the ecosystem.

2.2. Controlled Storage of SNF

At present, long (dozens of years) controlled storage of SNF in plants or central SNFSs is realized almost in all countries.

Currently that method of SNF management is the cheapest and meets the requirements of plutonium nonproliferation because in the SNFS the last is under protection of strong gamma-irradiation of fission products that facilitates account and control of the NFMs and leaves open the possibility to use SNF in the closed NFC when the conditions in economics become ready for that.

Along with that, the following limitations are peculiar to the existing technologies:

- Long storage of SNF in the cooling water pools are resulting in the development of corrosion processes in fuel elements claddings and opportunity of radioactive contamination of water.
- The necessity for uninterruptible electricity supply for the purpose to provide operation of cooling systems. In compliance with that requirement is resulting in water boil-off, heating of fuel, steam-zirconium reaction with generation of hydrogen in large amounts, explosions and radioactivity exhausts (Fukushima 1). For that reason, for a long time, there is in practice the process of changeover to "dry" cask storage with cooling realized by air natural circulation after the SNF has been cooled in the water pool for three years or more for the purpose to reduce the residual heat. Such newly built central "dry" cask storage repository is implemented on the site of Mining and Chemical Combine (Zheleznogorsk, Krasnoyarsk Krai, Russia).
- The cost of SNF storage is increasing while the number of NPPs and their operation time are growing much. Though the "dry" SNFSs with air natural cooling does not require large operating expenses, the capital costs needed for their construction are large due to the necessity to protect them against external impacts. These costs will be decreasing with increasing storage density measured in metric tons of heavy metal per one square meter of the repository.

Along with that when the issues of storage of power reactors SNF were considered at the IAEA Scientific Forum held within the frameworks of the 47th IAEA General Conference (Vienna, 2003), the certain countries expressed a desire to extend the SNF storage time up to 100 years and over. They explained that by the factor of delays in implementation of the programs on SNF burial in geological repositories. In addition, significant expanding of the time of SNF storage in "dry" repositories makes a possible saving of financial resources for the construction of geological repositories. Absence of social agreement on the issue concerning how to consider the spent fuel, namely: as a waste or fuel for the future NP, lack of political will in activities on options for the sites for geological repositories and their construction are also the motivation in the certain countries to extend the period of "dry" storage of the SNF.

2.3. NFC Closing

Organizing of the closed NFC with the implementation of FRs and large-scale reprocessing of TR and FR SNF in a unified NFC. Thus, the fission products are separated for their further immobilization and final isolation (really, radioactive wastes). The remaining uranium and built plutonium are used for manufacturing of fresh fuel. At the same time, the task of noticeable reduction of amounts of stored SNF and unloading of repositories is solved.

To solve the problem of MA incineration, the different methods of nuclear transmutation of MA in fast critical or subcritical (accelerator driven) reactors are studied. In those reactors, the long-lived MA are fissioned by fast neutrons and transmuted into relatively short-lived fission products. After required cooling in the controlled repository those fission products can be vitrified similar to fission products of uranium and plutonium and then safely buried in geological repositories.

Realization of the highlighted approach needs implementation of the high number of FRs in the NP structure, which allow increasing approximately by a factor of hundred the efficiency of use of energy potential of natural uranium as compared with that of TRs. In many countries the FRs have been highly developed since the second half of the last century due to the existence of two challenges, namely: 1) predictable high paces of NP development, 2) insufficiency in explored economically available resources of natural uranium.

Sodium cooled fast reactors with a breeding ratio exceeding one (BR > 1) could provide short plutonium doubling time and upon operating in the closed NFC made possible elimination of limitations on the paces of implementation of new nuclear power plants (NPPs)due to insufficiency in economically available resources of natural uranium. However, they were more expensive as compared with TRs operating in the open NFC, and were not economically demanded in conditions of low costs of natural uranium and services on its enrichment.

3. Possible Duration of Temporal SNF Storage Prior to Reprocessing

At present it is not easy to determine the time when reprocessing of the SNF with recycling of plutonium and MA, separation of fission products and their final isolation will become economically expedient. That time will depend on specific consumption of natural uranium by existing TRs and its contribution into a fuel component of the electricity cost, forecast for resources of natural uranium and their dependence on the cost of natural uranium, escalation of natural uranium costs (now they are not increasing), prospect for the paces of NP development in the current century, the cost of SNF storage and reprocessing, the cost of manufacturing of refabricated fuel, economic characteristics of fast reactors, the cost of final isolation of long-lived RAW.

For the purpose to make a preliminary decision for NFC variants for future large-scale NP, the comparative economic analysis of the open NFC with postponed SNF reprocessing and closed NFC for the USA conditions was performed in the Massachusetts Institute of Technology (MIT Reports) [7] and earlier [8]. In those Reports, it was revealed that up to the end of the current century the thermal reactors operating in the open NFC would not lose their competitiveness because of rising in the cost of natural uranium.

However, as the commercial FRs operating in the closed NFC have not been implemented yet, there are many uncertainties in estimations of their economic characteristics. That point is concerning both economic characteristics of FRs and economic parameters of the closed NFC. All highlighted points will effect on forecasting determination of the time for economically expedient NFC closing. The same can be said about the assessment of economically available resources of natural uranium and paces of NP development.

According to the common geological regularity, on average the mineral resources are growing in proportion to the squared cost that the consumer is ready to pay. Of course, that refers to uranium as well. For instance, according to the data presented by UICA (Uranium Information Centre Australia) [9], doubling of the cost of uranium ($30/kg) that existed in 2000 could presumably result in 10 times more growth of natural uranium resources, which cost is $80/kg or less, i.e. the resources of natural uranium will increase from 3 to 30 million metric tons.

Along with that, even considerable increase in the cost of uranium extracted from depleted deposits can only slightly raise the cost of electricity due to the fact that the cost contribution of natural uranium into the cost of produced electric power is low. For example, according to the data given in [8], in case the cost of uranium is increased by a factor of twice, i.e. from 30 to $60/kg, the electricity cost of the existing NPPs is growing by 0.11˚ cent per kilowatt hour or by 2.2%.

When uranium is extracted together with other minerals, the uranium resources are growing appreciably along with depletion of uranium content in the extracted ore. The example is phosphate deposits where uranium content is low, namely from 10 to 300 ppm. However, it is forecasted in [10] that in the uranium-ore deposits the mentioned content of uranium is contained in approximately 22 million metric tons of uranium.

According to the data presented in [7], electricity produced by light water reactors (LWR) will be competitive until the uranium cost reaches the values in the range of $300 - $400/kg. The highlighted cost range is covering the range of predicted costs of uranium extracted from sea water. However, even provided that forecast is true, it is difficult to agree that the SNF is a radioactive waste that must be buried once and for all.

Among the world countries, which possess the uranium resources, Russia, where the uranium cost is less than $130/kg thus providing competitiveness of the NP based on TRs, is on the fourth place, right after Australia, Kazakhstan and Canada [10]. These resources (both determined and forecasted ones) are as much as 1.3 million metric tons. However, because of the low market costs of natural uranium, at present the certain deposits are unprofitable. Today the repeated use of waste uranium that has been piled in great amounts at the uranium enrichment plants is more profitable. The additional resource is the use of regenerated uranium obtained on the extraction of weapon plutonium at the Mayak plant for its after-enrichment to the content of ^{235}U in natural uranium with the application of the most effective gas-centrifugal technology for separation of isotopes.

On the assumption that till the end of the century the average value of overall installed power capacities of NPPs with VVER in Russia is about 70 GW (it corresponds to the average pace of annual implementing in operation of 1.5 GW or more with account of decommissioning of power-units) and 115 GW is a value of overall installed power capacities of NPPs in 2100, it will be required about 1.2 million metric tons of natural uranium upon reactors operation in the open NFC with postponed reprocessing of SNF.

Bearing in mind it is unlikely that by the end of the century the total power capacities of SVBR-100 reactors will exceed 10 GW while their serial implementation is beginning from 2030, their operation in the open NFC with postponed reprocessing, provided it is profitable, will require 280 thousands of tons of natural uranium.

With due account of the highlighted above, it can be expected that prior to the SNF has been involved in the closed NFC, duration of the period of storage of SVBR-100 SNF can take dozens of years. For that reason, the development of the concept of sufficiently long controlled storage of SNF from SVBR-100 reactors is expedient.

4. The Concept of Safe Controlled Storage of SNF from SVBR-100 Reactors

For the design of experimental-industrial power unit (EIPU) with reactor facility (RF) SVBR-100 the variant of cassette-by-cassette extraction of the SFSA with their further placing in capsules filled with lead was adopted. Then those capsules had to be stored for a long time in the in-plant "honeycomb" type storage facility with natural air cooling that provided removal of residual heat from the

SFSA. The storage duration can be hundred of years or more because on the way of radioactivity release into the environment there are four safety barriers, which are not subject to damaging factors during storage.

Along with that, the experience of operation of RF with lead-bismuth coolant (LBC) has revealed that it is possible to perform safe refueling in short terms provided the whole core is replaced with the use of the special refueling equipment set [11]. At that point, cooling and long storing of SNF extracted from the reactor were realized in long storage tanks (LST) filled with liquid LBC, which was solidifying after that. Removal of residual heat was performed via the LST casing by natural circulation (NC) of atmospheric air. By present, the obtained storage time without any signs of radioactivity release is about fifty years. Such technology simplifies the technological process of refueling, shortens its duration, and makes possible diminishing of dimensions of the main building and reducing of its cost.

However, in contrast to the nuclear submarines (NS) RFs, in which the residual heat was low when unloading was performed, for RF SVBR-100 in a month from the moment of reactor shutdown the residual heat upon unloading is about 500 kW. For that reason, it is necessary to place the spent removable unit (SRU) with SFSA into the temporal storage tank (TST) filled with liquid LBC, in which there are conditions for NC of LBC and removal of heat via the tank casing to the water cooling system.

In a year of cooling the heat, decay is reduced to 170 kW. That makes it possible to extract the TST together with SRU out of the shaft and install it in the in-plant transport-package container (ITPC), where further cooling of the SRU is realized till complete solidification of LBC in the TST. And that will take ten years. TST cooling in the ITPC is realized by natural circulation of atmospheric air. For the purpose to reduce the cost of LBC in the TST, the LBC can be manufactured from lead and bismuth of cheap technical grades instead of those of reactor fineness.

After the residual heat has been decreased to the level that makes possible transportation of the TST with SRU in the transport-package container (TPC) without taking the special measures on organizing of heat removal and meeting of the railway requirements and normative documentation requirements on transportation of packages with nuclear fissile materials, the following two variants of SNF management are possible:

1) To transport the TPCs to the Mayak plant, where unloading of SRUs from the TST is performed and then dismantled for further reprocessing of the SNF. The technology for performance of works on the unloading of the SRU and description of the equipment is presented in paper [11]. That variant of SNF reprocessing can be realized at the Mayak plant on the expiration of 10 years of SNF storage in the ITPCs on the NPP site provided SNF reprocessing has been organized by that time.

2) To transport the TPCs to the special site. At that site after the SRUs have

been reloaded into the LSTs filled with quickly solidifying liquid lead (on ten years of cooling the residual heat is about 25 kW and can be easily removed by atmospheric air NC under the temperature of the LST wall being not more than 200˚C), the LST are stored during the required time. Thus, the controlled storage of LSTs is realized for several decades up to the time when SNF reprocessing and NFC closing are becoming economically expedient. Upon that storage, the four safety barriers are formed on the way of releasing of radioactive products into the environment, namely: fuel matrix, fuel element cladding, hard lead and steel casing of the LST. In addition to the reduction of the cost, replacement of LBC by lead is providing more reliable protection of SRUs against extremal external effects due to the higher temperature of lead solidifying (327˚C). The additional shielding is a reinforced concrete hood (RCH), which walls are of a required thickness, that covers each LST. By performed assessments, the RCH thickness is within the range of (50 - 100) cm.

It is expected that separation of functions of SRU transportation in the expensive TPCs, whose number is not large, and long storage of the LSTs protected against external effects by comparatively cheap RCHs can be more economically expedient than use of a large number of dual-purposed containers, the cost of each is about $2 million [5].

That practice of temporal controlled storage of thermal reactors SNF in the reinforced concrete containers on the open site is realized in the USA [7] and in Germany where cast-iron dual-purposed containers are used for SNF storage and transportation [5]. "Dry" storage facility of SNF in the USA is shown in **Figure 1**. CASTOR V (Interim storage facility Surry) is shown in **Figure 2**. With reference to the data presented in [7], the cost of such storage heightens the cost of electricity only by 1% - 2% upon the density of SNF storage being 0.5 tons of heavy metal per square meter.

Upon that density of storage on the site, which square is 200 × 200 m^2, for safe controlled storage it is possible to place 13,000 tons of the SNF from reactors SVBR-100 (about 1500 LSTs with spent removable units). That corresponds to

Figure 1. "Dry" storage facility of SNF in the USA.

Figure 2. CASTOR V (Interim storage facility Surry).

approximately 10 GWeof total capacity of 100 year operating NPPs with reactors SVBR-100.

5. Conclusion

- SNF management is an NP postponed problem that has not been solved in full in any country in the world. It is possible to find the most complete solution to that problem when the closing of the NFC is realized and a large number of FRs are implemented in the NP structure.
- At present, it is difficult to determine the real time when the implementation of FRs operating in the closed NFC is becoming economically expedient. That period is determined by economically available resources of natural uranium and economic characteristics of FRs and closed NFC. For that reason, it is a universal practice of SNF storing in cooling water pools and then in "dry" storage facilities on the NPP sites or centralized repositories.
- For operated LBC cooled reactors, which operated at NSs, the practice of long storage of unloaded cores in long storage tanks filled with solidified LBC was successfully realized. Under such storage, the four defense-in-depth barriers are formed on the way of radioactivity release into the environment, namely: fuel matrix, fuel element cladding, solid LBC and tank casing. And that is assuring the high level of safety.
- It is expedient to consider the similar solution for reactors SVBR-100, in which LBC is replaced by lead providing the higher level of effective protection due to its higher melting temperature. The duration of such storage can reach several decades when the realization of the closed NFC is becoming economically efficient.

- On the site, which square is 200 × 200 m^2, it is possible to place 13000 tons of the SNF that corresponds to approximately 10 GWe of total power capacity of 100-year operating NPPs with reactors SVBR-100.

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Conflicts of Interest

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Abbreviations and Acronyms

BN: Sodium Fast Reactor
BR: Breeding Ratio
EIPU: Experimental-Industrial Power Unit
FR: Fast Reactor
HLMC: Heavy Liquid-Metal Coolant
IAEA: International Atomic Energy Agency
ITPC: In-Plant Transport-Package Container
LBC: Lead-Bismuth Coolant
LST: Long Storage Tank
LWR: Light Water Reactor
MA: Minor Actinides
MIT: Massachusetts Institute of Technology
MOX fuel: Mixed Oxide Uranium-Plutonium fuel
NC: Natural Circulation
NFC: Nuclear Fuel Cycle
NFM: Nuclear Fissile Materials
NP: Nuclear Power
NPP: Nuclear Power Plant
NS: Nuclear Submarine
PA: Production Association
RAW: Radioactive Waste
RCH: Reinforced Concrete Hood
RF: Reactor Facility
SFSA: Spent Fuel Sub-Assembly
SNF: Spent Nuclear Fuel
SNFS: Spent Nuclear Fuel Storages
SRU: Spent Removable Unit
SVBR: Lead-Bismuth Fast Reactor
TPC: Transport-Package Container
TR: Thermal Reactor
TST: Temporal Storage Tank
VVER (PWR): Pressurized Water Reactor

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Atom's Nuclear Structure and the Periodic Table of the Chemical Elements

Faustino Menegus

V. Europa 15, Bussero, Italy
Email: menegus.faustino@gmail.com

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Abstract

A new method for the identification of the chemical Elements isotopes takes advantage of the isotope Neutron Excess (NE) number. The representation of the natural isotopes in the Z-NE plane reveals a surprising correspondence between atom's nuclear and electronic structures. Nuclear directs the atom electronic structure in spite of the alternative set of numbers ruling the two main atom's compartments. These compartments appear better integrated than actually considered. The Mendeleev periodic table is rooted in the atom's nuclear structure. Two recent studies arrive to identical conclusions.

Keywords

Periodic Table, Atom's Structure, Nuclear Structure, Electronic Structure

1. Introduction

Atomic nuclei birth takes place in the star interior at temperatures and pressures allowing their nucleosynthesis. Following migration at the star periphery, the reduced temperature and pressure allow nuclei to acquire the necessary electrons to become the stabilized atoms constituting the chemical Elements of our world [1]. The heavy central nuclear, $r = 10^{-15}$ m and close to 100% of the atomic mass, and the external electronic cortex, $r = 10^{-10}$ m and close to 100% of the atomic volume, appear as very distinct and somehow independent atom's compartments. Two distinct branches of physics, nuclear and atomic, describe the above compartments notably ruled by two different sets of numbers: 2, 8, 20, 28, 50, 82, 126 the proton-neutron nuclear magic numbers and 2, 10, 18, 36, 54, 86 the noble gases electron numbers, respectively. While the noble gases numbers satisfactory describe the chemical Elements electronic structure and properties, this is not the case for the magic numbers. The nuclear structure is still an open field [1]. So different compartments constitute, nevertheless, stable and defined objects: the atoms of the chemical Elements. We suggest here that subtle influences exist between the nuclear and the electronic structure, the latter, likely dictated by the nuclear one. Besides the strong electrostatic interaction between the nucleus and the electrons, a cross-talk of electromagnetic nature takes place between the atom's main compartments: the analysis of atom's electronic spectra allows the retrieval of important nuclear information [2]. Mendeleev maintained that, in addition to the periodic properties, each element has its own personality [3]. Properties like the diagonal behavior, Li and Be have similarities with Mg and Al respectively, or the knight's move, whereby Zn resembles Sn, may be of nuclear origin. To provide the ground for the discussion of the above ideas, it was decided to employ the representation of the chemical Elements isotopes by the Neutron Excess number [4] [5]. Such a representation contains the basic elements of both the nuclear and electronic atom's compartments.

Recently Mao J., guided by the most abundant isotope of each Element, proposed the atom's nuclear structure as the root of the periodic table [6]. Identical conclusions where reached, Bonchev D. [7], using the tools of the Information Theory [8]. An idea of the present work is contained in a poster presentation at the 15th International Conference on Condensed Matter Nuclear Science: Rome 2009 [9].

2. Methods

The Excess Neutron (EN) number of the isotope ${}_{Z}X^{A}$ of the element X was calculated by the equation: $EN = A - 2Z$. The mass numbers A were from *K.* Lodders [10]. The results constitute a new method for the representation of the chemical Element's isotopes in the Z-NE plane.

3. Results

The representation of the chemical Elements isotopes by the Neutron Excess number

In **Figure 1**, the electronic structure of the Elements is represented by the principal quantum number, $n = 1, 2, 3\ldots.$, and by the orbital angular momentum quantum number, the latter, as usual, by the letters s, p, d, f. The natural isotopes of the chemical Elements are represented by the atomic Z number and by their Neutron Excess number (NE). With $Z = P =$ proton number, $N =$ neutron number, $A = P + N =$ mass number, $NE = N - P =$ Neutron Excess number, the relationship $A = NE + 2Z$ holds for each isotope represented in **Figure 1**.

Highlighted in **Figure 1** are: the period numbers of the Mendeleev table, the noble gases location and the proton-neutron couples of the nuclear magic numbers. The maximum and minimum NE level of each Element define the borders of the β-stability valley. The figure inset shows same basic correlations between the isotopes of the chemical Elements.

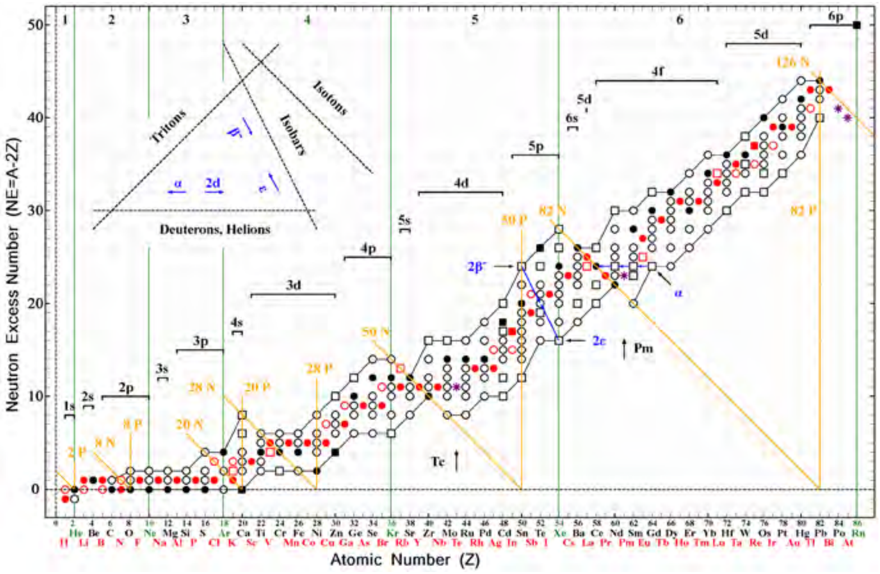

Figure 1. Representation of the Elements isotopes by the Neutron Excess number, $NE = A - 2Z$, in the Z-NE plane and the respective electronic structure. The noble gases limiting the periods of the periodic table, indicated at the top of the figure, are green; s, p, d, f, indicate the electron's angular momentum quantum numbers. Highlighted are the proton-neutron couples of the nuclear magic numbers. Even and odd Element's symbols are black and red respectively; (○) (□) represent stable and radioactive isotopes respectively, with the full symbol indicating the most abundant isotope of each Element; (✳) represents the most stable isotope of the artificial Elements. The inset shows basic relationships between the isotopes along with radioactive decay paths.

What matters to be noted here is that, with the progress of the *Z* number, the nuclear *EN* addition takes place with pauses, upsurges and reductions that parallel the periodicity of the chemical Elements properties, the latter marked by the noble gas location. In fact following He, Ne, Ar, Kr and Xe there is first a pause, period 2, 3 and 4 or a reduction, period 5 and 6 of the maxima of the NE level addition to the Elements nuclei; see discussion for the anomalous situation at Ca. Most important is the net *NE* upsurge, coincident with the electron filling of the p orbitals, reaching completeness at the noble gases. The electron filling of the 3 d orbitals takes place with a clear *EN* addition pause. That pause, still present in the 4 d and in the subsequent f and d orbitals, progressively vanish because of a robust *NE* addition. The *NE* addition is required to counteract the nuclear proton-proton repulsion caused by the *Z* progress [4]. Closure of the Table periods, with maxima of the nuclear *NE* levels, is a strong indication of a nuclear periodicity, repeating that of the electronic compartments.

The drop of the *NE* level behind Kr and Xe appears directly related to the 50 and 82 isotons. The lack of natural isotopes for Tc and Pm, exactly seven places beyond the noble gases, suggests a gearing of the electronic with the nuclear structure. That these structures are at the least partially geared, is indicated by the identical four proton gap existing between the 50 P and 82 P magic numbers of Sn and Pb and the noble gases Xe and Rn. The most abundant isotope of each element convey the same information contained in the upper border of the β-stability valley, see also [6].

Further support to the idea that the electronic may reflect atom's nuclear structure comes from **Figure 2**, representing the weighted means of the Elements *NE* content. Periods 4, 5 and 6 show again a clear correspondence of the electronic with the nuclear structure: a sharp nuclear *NE* increase accompanies the electron filling of the p orbitals, reaching maxima at the noble gases. The *NE* pause after Ar and its reduction after Kr and Xe mark the electron filling of the d and f orbitals. The message of **Figure 2** reinforces that of **Figure 1** in spite of the notable discrepancy for the light Elements up to Ti. The evolvement of the Elements molar atomic volume [11], with the progress of *Z*, is anything but a monotonous-one, like that of the *NE* addition to nuclei. The sharp volume increase of the alkaline and alkaline earth metals, coinciding with the s orbitals filling next to the noble gases *NE* maxima, stresses a nuclear-electronic structure correspondence.

It is well known from inorganic chemistry that in addition to the periodical similarities of Zn and Cd with Cd and Hg respectively, Zn and Cd show in addition similarities with Sn and Pb respectively. This last property is known as the Knight's move relationship [12]: one step down and two steps right in the periodic table, as in the game of chess [3]. **Figure 2** shows that relationship from the nuclear point of view.

4. Discussion

In **Figure 1** the ^{46}Ca and the ^{48}Ca isotopes, at variance with the situation in Mg,

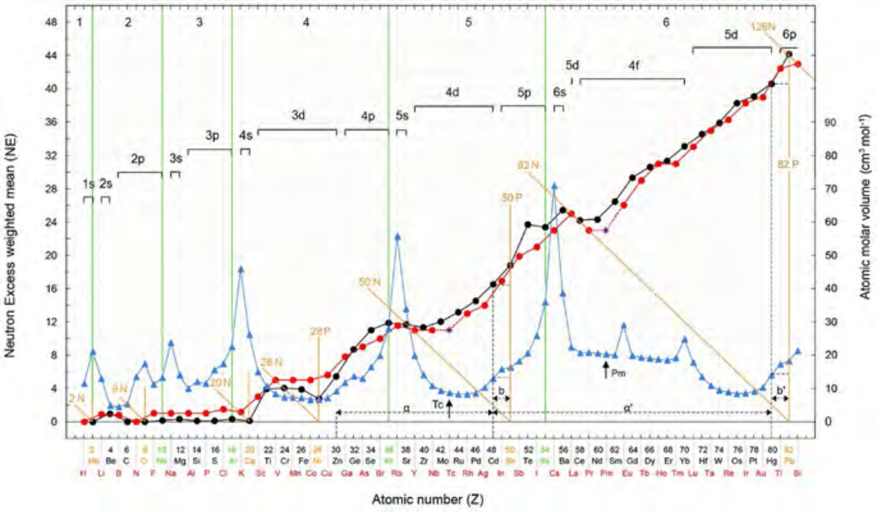

Figure 2. The representation element's *NE* weighted means and of the Elements Atomic Molar Volumes. The *NE* means are black or red for even or odd Elements respectively. (▲) indicate the Atomic molar Volumes. Other symbols as in **Figure 1**. Both properties show clear periodicity matching that of the Element's electronic structure for periods 4, 5 and 6. a, a' and b, b' indicate one step down and two steps right respectively in the periodic table as in the chess Knight's move.

Sr and Ba, show an abnormal *NE* increase corresponding to the electron filling of the s orbitals. This exception may be the result of the paramount influence, on the nuclear structure, of the combined effects of the 20 P and the 28 N magic numbers. The *NE* drop behind Kr and Xe, followed by its resumption with the progress of *Z*, is the result of the *NE* reduction connected to the 50 N and the 82 N respectively.

The lack of *NE* maxima at Ne and Ar in **Figure 2** is the result of the nucleosynthesis mechanisms: nuclear fusion, fading at Fe, for the light Elements, then replaced by the neutron capture processes [1]. The *NE* drop, connected with the 50 N and the 82 N isotons, shows up again with the 126 N isoton, accompanied by dramatic consequences: beyond Bi atom's nuclei become unstable. The *NE* drop is evidenced by the reduced *NE* content of the most long-lived isotopes of Po and At. A *NE* recovery takes place with the most long-lived isotope of Rn, confirming *NE* maxima for the noble gases. Notably the 126 N isoton does not relate to a noble gas. It relates however with the complete filling of the 4f and 5d orbitals.

At variance with the upper, the lower border of the β-stability valley doesn't reflect the Element's electronic structure, because it is differently affected by the 50 and 82 neutron magic numbers (**Figure 1**). As proposed by J. Mao [6], the most abundant isotope of the Elements reflects the atom electronic structure: a suggestion confirmed by the full symbol isotopes in **Figure 1**. The weighted means of the Elements *NE* content (**Figure 2**) confirm again the nuclear-electronic structure correlation.

In **Figure 2**, the nuclear interpretation of the knight's move shows that moving two steps at the right of Cd and Hg, Sn and Pb are reached respectively. In both cases the identical nuclear circumstance is found: the shift zeroes the influence of the 50 and 82 neutron magic numbers [4] [5], with the consequent increase of the Elements molar atomic volume [11] and as well as that of the *NE* level. Changes that may offer a key for the interpretation of the knight's move, presently still debated [13] and, at the same time, may constitute a validation test for the suggestions of the present work.

5. Conclusion

The representation of the natural isotopes in the Z-NE plane, together with the basic elements of their nuclear and electronic structures, convey the idea that nuclear directs the atom's electronic structure. In particular, the Element's electronic structure is reflected in the upper border of the β-stability valley, and in the evolution of the *NE* number of the most abundant isotope of each Element (**Figure 1**). This last observation agrees with the findings of J. Mao [6]. **Figure 2** beautifully confirms the above conclusions. The Neutron Excess addition mode to nuclei, a determinant of the nuclear stability of the natural isotopes, acts also as a determinant of the atom's nuclear structure. The periodic table of the chemical Elements is rooted in the atom's nuclear structure.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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