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Araştırma Makalesi / Research Article

Uranium Potentiality of Coal Occurences in Dinar (Afyonkarahisar, western Turkey) region: Geologic Factors Controlling the Accumulation of the Uranium

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Abstract

Random distribution of energy resources in the world, rapidly increasing energy prices, environmental problems such as global warming and climate change, the depletion of fossil fuels in the near future and their negative impact on the environment and human health, increasing dependence on foreign energy, limited energy resources of big consumer countries and being dependent on a small number of specific countries lead states to seek various sources of energy. The uranium fuel-based nuclear energy generation is an alternative energy source for Turkey. Proven uranium reserves in Turkey are 32.4 kt and exploration of sedimentary basins for uranium mineralization still continues. In this context, Dinar (Afyonkarahisar, western Turkey) coal occurences can attract attention for economic possibilities of uranium related with Plio-Miocene aged extensional tectonic regime. A total of 17 coal samples were collected for analyses from five boreholes at various depths. The uranium contents of studied coals are up to 1065 µg/g. The origin of the uranium in this basin are considered as a stratal epigenetic origin, which the uranium was introduced in the coal after coalification and consolidation of the enclosing sediments by ground water deriving uranium from hydrothermal sources or from unconformably overlying volcanic rocks. In addition, it can be highlighted that the higher contents of uranium occur in the upper parts of the stratigraphically highest lignite beds, which is compatible with other epigenetic lignite deposits.

Keywords
Uranium; Lignite; Afyonkarahisar/Dinar; Western Turkey

Dinar (Afyonkarahisar, Batı Türkiye) Bölgesindeki Kömür Oluşumlarının Uranyum Potansiyeli: Uranyum Birikimini Kontrol Eden Jeolojik Faktörler

Öz

Dünyada enerji kaynaklarının rastgele dağılmış olması, yükselen enerji fiyatları, küresel ısınma ve iklim değişikliği gibi çevresel sorunlar, yakın gelecekte fosil yakıtların tükenecek olması, bu enerji kaynaklarının çevre ve insan sağlığı üzerindeki etkileri, yabancı kaynaklara ve az sayıda enerji zengini ülkeye bağımlılık sebebiyle ülkeler farklı enerji kaynaklarına yönelmiştir. Uranyumu yakıt olarak kullanan nükleer enerji üretimi Türkiye için alternatif bir kaynak olarak ele alınmaktadır. Türkiye'nin kanıtlanmış uranyum rezervi 32.4 kiloton olup, halen uranyum içerebilecek havzaların araştırılması devam etmektedir. Bu kapsamda, Pliyo-Miyosen genişlemeli rejim sonucu gelişen Dinar (Afyonkarahisar, Batı Anadolu) kömür oluşumları ekonomik açıdan önem arz edebilecek uranyum içerikleri bakımından ilgi çekebilmektedir. Çeşitli derinliklerden ve 5 sondaj kuyusundan toplam 17 adet kömür numunesi alınmıştır. Bu numunelerin uranyum içeriklerinin 1065 µg/g mertebelerine kadar çıktığı görülmüştür. Havzada mevcut olan uranyumun, kömürleşme süreci ve kömür tabakalarının üzerinin volkanoklastik sedimanlar tarafından örtülmesinden sonra, hidrotermal akışkanlar ile karışan

Anahtar kelimeler
Uranyum; Linyit; Afyonkarahisar/Dinar; Batı Türkiye

yeraltı sularının etkisiyle katmanlı epijenetik köken modeline uygun şekilde oluştuğu düşünülmektedir. Ayrıca, yüksek uranyum içeriğinin, linyit damarlarının üst katmanlarında yoğunlaşması sebebiyle havza diğer epijenetik linyit oluşumları ile benzerlik göstermektedir.

1. Introduction

Random distribution of energy resources in the world, rapidly increasing energy prices, environmental problems such as global warming and climate change, the depletion of fossil fuels in the near future and their negative impact on the environment and human health, increasing dependence on foreign energy, limited energy resources of big consumer countries and being dependent on a small number of specific countries lead states to seek various sources of energy. The uranium fuel-based nuclear energy generation is an alternative energy source for Turkey. Proven uranium reserves in Turkey are 32.4 kt and exploration of sedimentary basins for uranium mineralization still continues. In this context, Dinar (Afyonkarahisar, western Turkey) coal occurences can attract attention for economic possibilities of uranium related with Plio-Miocene aged extensional tectonic regime.

Radioactive elements' existence in lignites and other hydorcarbon basins are reported at multiple locations (Gentry et al. 1976, Zelinski and Meier 1988, Dill 1987, Douglas et al. 2011). In earth crust, concentration of uranium is around 1-3 ppm (Hazen et al. 2009, Fayek et al. 2011).

The solubility of the solutions with low temperature increases significantly with the presence of strong anions such as F^-, Cl^-, $(SO_4)^{2-}$, $(CO_3)^{2-}$, $(PO_4)^{2-}$ (Langmuir 1978). It increases the solubility and the mobility of uranium in groundwater (Cuney and Kyser 2008). The solubility of the uranium minerals in aqueous media and the behavior of the uranyl complexes are given in Figure 1.

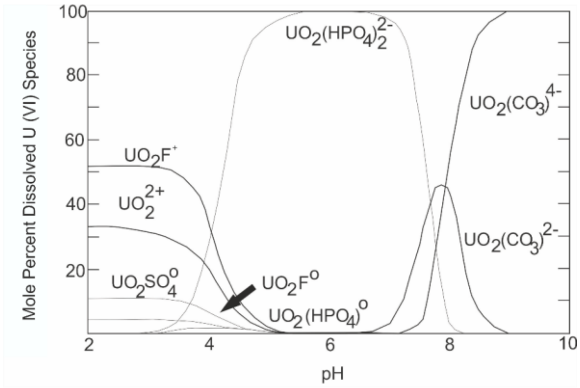

Figure 1. Distribution of uranyl complexes versus pH for some typical ligand concentrations in ground waters of the Wind River Formation at 25°C. PCO_2=10-2.5 atm, ∑F= 0,3 ppm, ∑Cl=10 ppm, $∑SO_4$=100 ppm, $∑PO_4$=0.01 ppm, $∑SiO_2$=30 ppm (Langmuir 1978).

The highest concentrations of uranium in ground water are found in areas of known deposits (about 70-300 parts per billion) and in ground water from silicic tuffs and tuffaceous rocks of Oligocene and Miocene age (about 20-45 parts per billion). The most highly mineralized water from tuffaceous terranes has an alkaline pH range of 7.5-9.5 (Denson 1959). In general, the greater the alkalinity of the waters from these rocks, the more uranium the waters contain. A large part of the uranium deposits (sedimentary deposits) could be dissolved from a prominent uranium source (metamorphic massives, peralkalin or peralumin) and deposited in a suitable environment (Eh-pH conditions, solution rock reactions, permeability of rocks). The genetic factors for uranium enrichment in coals are shown in Figure 2.

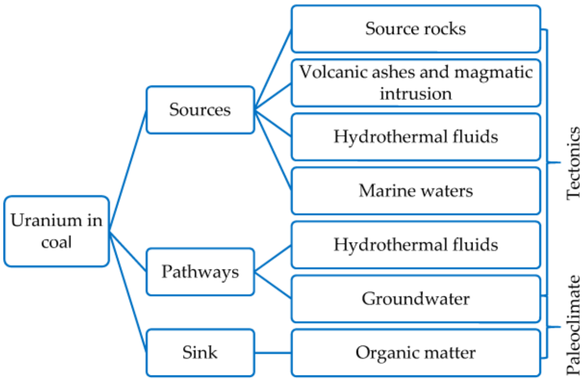

Figure 2. Genetic factors for uranium enrichment in coals. The schematic diagram is reorganized from Chen et al. (2017).

The uranium concentrations of the coal deposits ranging from 0.005 to 0.02 % generally are not uniform. Within the low-grade uranium-bearing coal deposits, smaller irregular masses of coal containing 0.1 percent or more uranium have been found in some deposits around the globe. The large quantities of uranium can be expected in the low-rank coal of Plio-Miocene aged lacustrine sedimentary basins in western Anatolia.

The aim of this study is to evaluate the economic possibilities where uranium was found in sufficient amount in coal and to determine the geologic factors controlling the accumulation of the uranium in Dinar (Afyonkarahisar, western Anatolia).

2. Material and Methods

A total of 17 coal samples were collected for analyses from five boreholes at various depths. In order to obtain the original basis moisture, ash, volatile matter and fixed carbon core samples are prepared (ASTM International 2013). Firstly, they are fed into a crusher, then, pulverised with a lab scale ring mill. Then, proximate analysis is carried out with determining moisture content (ASTM International 2017), ash content (ASTM International 2012) and volatile matter (ASTM International 2017) by utilizing a Binder brand stove and a Carbolite brand oven. Also, some other part of pulverized samples are put in the oven at 750 °C for at least 6 hours until all organic matter is removed.

Elemental analyses were conducted on powders grounded using an agate mortar muller milling device for Elan DRC-e Perkin Elmer Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) in Geochemistry Research Laboratories of Istanbul Technical University (ITU/JAL) to determine the U, Th and REE element contents. A two-step digestion process used approximately 50 mg of powdered samples: (1) 6 ml of 37% HCl, 2 ml of 65% HNO_3 and 1 ml of 38–40% HF acid mixer put in a pressure- and temperature-controlled Teflon beaker using a Berghoff Microwave at 135 °C; (2) 6 ml of 5% boric acid solution was added to the step one mixer for ICP-MS analyses.

3. Geological Setting

The Dinar (Afyonkarahisar) lignite deposit is located approximately 120 km^2 in the western Turkey. The Dinar sedimentary basin is bordered between the Menderes massif in north and the Lycian nappes in east-southeast direction. The autochthon Pan-African basement overlain by the Menderes massif Lower Devonian-Eocene schist and marbles and Mesozoic-Early Cenozoic platform carbonates. The Menderes massif, described as Pan-African basement, is overlained by Lower Devonian-Eocene schist and marbles and Mesozoic-Lower Cenozoic platform carbonates. The carbonates consist mainly of marble, phyllite, schist, gneiss, dolomite and limestone. Lycian nappes contain fragments of the metamorphic rocks of Taurides overlained by the Mesozoic platform carbonates, and fragmented ophiolite and volcano-sedimentary units at the top of the nappes (Yılmaz et al. 2000).

Pre-Eocene carbonate rocks and Eocene Sarıdere formation (mainly claystone and sandstone) can be considered as the basement of the basin. Oligocene marine conglomerates which are named as Akcakoy formation deposited on these units. Lacustrine sediments and fluvial sediments composed of Upper Miocene-Pliocene sandstone, siltstone, claystone, marl, limestones, lignite with volcanic intercalations (Kumalar formation) were deposited uncomformably. Along the northern border of the graben, Pliocene limestones and carbonate cemented conglomerates (Karatas formation) unconformably overlie the basement rocks. Towards the basin, stream-lacustrine units contain conglomerate claystone, marl and siltstone. Plio-Quaternary units (Kepeztepe formation), which are stored under the control of active tectonism, comprise alluvially-phase and river-lake sediments. In the eastern part of the study area Miocene-Pliocene volcanic sequences with trachyandesites, phonolites and pyroclastics are observed (Figure 3).

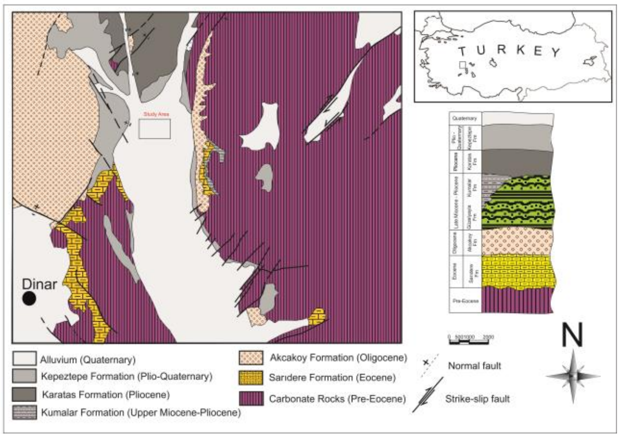

Figure 3. Geological map of the study area (modified from Bechtel et al. 2016)

4. Results and Discussion

4.1 Geochemical and Petrographical Properties of Studied Coals

A previous petrographic observation reveals that the Afyonkarahisar/Dinar uranium-bearing coals are rich in huminite, in which textinite and ulminite (telohuminite subgroup), attrinite, densinite (detrohuminite subgroup), and corpohuminite (gelohuminite subgroup) are more abundant than other macerals (Bechtel et al. 2016).

In addition, geochemical composition of selected major oxides and trace elements are listed in Tables 1 and 2. The geochemical studies focus on the original and dry-ash-free moisture, ash, volatile matter and fixed carbon values of the core samples as well as the U-Th-REE element contents. The dry-ash-free volatile contents of all samples from different boreholes are greater than 50%, indicating that they are in the lignite class.

The uranium contents in ash of studied lignite samples from five boreholes at various depths are up to 1065 µg/g. The ash content of the lignite ranges from 10 percent or less to about 20 percent, which indicates that after ignition of the lignite the uranium content of the ash is at least 5 times and generally 7-10 times the uranium content of the lignite. On the other hand, it can be concluded that uranium contents of studied samples are up to 106.5 µg/g in lignite. Furthermore, there is no evident dependency between uranium content and depth. The uranium behaves extreme mobile due to the Eh-pH conditions of basin. The Eh conditions of coals are sufficient for uranium enrichments regardless of depth.

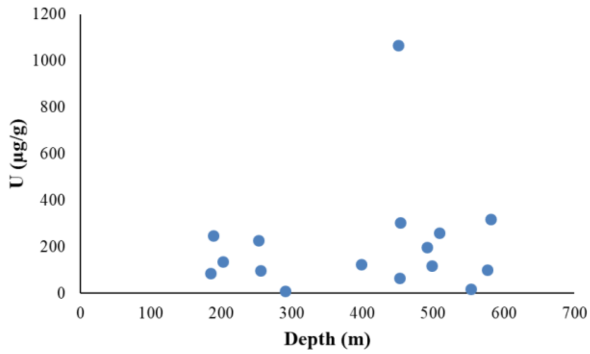

Figure 4. Uranium concentration vs. depth.

The total rare earth elements abundances (ΣREE) are variable for studied samples, from 27.3 to 250.6 µg/g (average of 140.9 µg/g) (Table 1). The ratio of total light REE/ heavy REE (ΣLREE/ΣHREE) of the coals range from 8.00 to 16.7 (average of 12.4). These enrichment of LREEs relative to HREEs values of coals show a consistency with most distinctive features of the REE patterns of the coals (Yan et al. 2009). The LREEs have a similar behaviour with element Th that there is high positive correlation between them (Figure 4). In addition, it can be said that high LREE contents were observed at between 300 and 400 m average coal depth (Figure 5).

Ash content of the samples fluctuates in low to medium interval. Thus, the seams have different burial conditions in terms of mineral matter considering multiple seams and depth range. However, uranium and other elements consistently available in all samples in some degree because deposition of uranium extends over time.

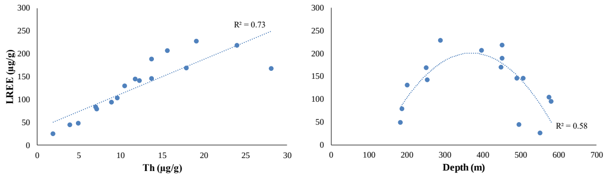

Figure 5. The correlations of LREE contents vs. Th and depth

High correlation values between TiO_2 vs. Th and ΣREE strongly suggest that the REE-Th bearing mineral is titanite which is highly resistant in arid-semi-arid conditions (Figure 6, Table 2).

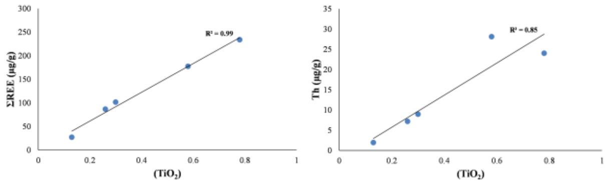

Figure 6. The correlations of TiO_2 contents vs. ΣREE and Th.

It can be concluded that the U-bearing minerals in lignite layers can be either phospate or vanadate group minerals such as torbernite, autinite or carnotite. These minerals are considered as secondary uranium minerals which can be found in sedimentary uranium deposits. The presence of $[PO_4]^{3-}$ and $[VO_4]^{3-}$ anions caused rapid precipitation of uranyl U^{6+} from low temperature hydrothermal or meteoric solutions (Figure 7, Table 2).

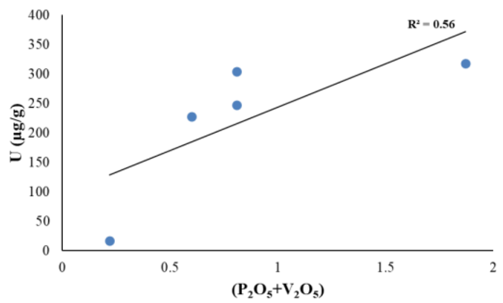

Figure 7. The correlations of U vs. $(P_2O_5+V_2O_5)$ contents.

In addition, the Th/U and U/Th ratio can be used as a paleoredox proxy (Myers and Wignall 1987, Nath et al. 1997). High concentrations of U suggest that the sediments were probably oxygen-deficient bottom water (Lezin et al. 2013). As shown in Table 1, Th/U ratios of coal samples are low, varying from 0.03 to 2.31, with an average of 0.35. Considering the Th/U ratios of 0–2 for mudstones suggest an anoxic environment, according to Wignall and Twitchett (1996), it can be concluded that coals were deposited in reducing environment. Furthermore, ratios of U/Th < 1.25 indicate oxidizing conditions, while values > 1.25 suggest suboxic and anoxic depositional conditions (Nath et al. 1997). Studied coals U/Th ratios are between 0.43 and 35.5 (average of 14.7), indicating that coal samples were deposited in anoxic depositional conditions.

Proximate analysis results of the samples are also given in Table 1. Considerably high moisture and volatile matter contents implies early stage of lignite rank. Produced fuel ratio values from volatile matter and fixed carbon contents supports this rank range as well. Moisture content of lignite seams decreases with increasing depth in a wide band (Figure 8). Therefore, impact of pressure is significant during coalification and immediate burial with peatification process probably took place.

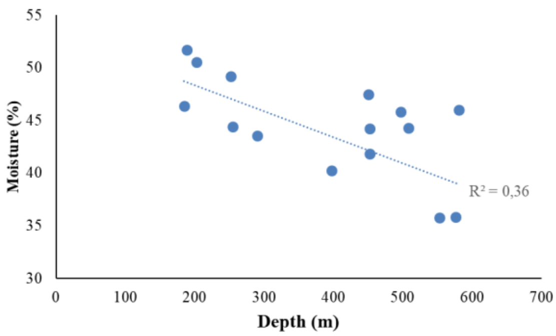

Figure 8. Moisture content vs. depth.

Table 1. Geochemical results (contents in µg/g) with original and dry-ash-free basis moisture, ash, volatile matter and fixed carbon (contents in %) values for coal samples from five different boreholes (named as A, B, C, D, E).

Element	A1	A2	B1	B2	B3	B4	B5	C1	C2	C3	C4	C5	D1	D2	D3	D4	E1
U	1065,8	17,4	65,1	228,3	135,1	247,5	84,7	14,5	318,3	101,7	304,4	117,4	8,31	98,0	198,4	259,5	124,3
Th	18,0	1,94	13,8	28,1	10,5	7,19	4,99	7,09	8,96	9,69	24,0	3,95	19,2	12,3	13,8	11,9	15,7
In	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Tl	1,53	0,52	0,04	4,72	3,18	0,23	0,24	0,32	0,17	2,71	3,81	0,23	1,21	4,22	2,93	2,50	5,90
Rb	78,7	25,0	56,1	143,2	111,9	34,9	26,1	37,6	47,7	70,6	138,5	17,8	145,3	124,2	69,9	65,2	78,5
Ga	29,0	6,04	20,5	22,1	19,1	8,88	4,58	12,1	29,6	12,0	37,5	7,47	31,7	23,6	19,9	19,6	18,1
Cd	8,20	7,03	6,74	2,56	4,29	1,94	7,87	9,87	8,54	3,48	2,22	3,77	3,39	3,70	1,84	4,03	3,38
Cs	15,0	3,77	5,76	20,0	15,1	5,79	5,33	3,66	9,62	9,95	20,3	3,17	10,0	15,0	12,0	10,9	14,4
Y	14,2	3,85	37,9	11,6	16,7	15,9	6,21	9,47	11,3	10,2	22,1	5,27	31,5	17,1	13,2	14,6	15,4
La	44,4	5,63	42,3	44,7	29,7	20,4	12,4	19,8	23,4	25,5	54,9	11,1	53,7	33,2	35,0	34,7	53,2
Ce	78,2	11,2	88,9	76,0	60,8	35,9	22,4	39,8	43,8	48,1	101,3	19,6	104,8	66,2	68,8	67,8	98,7
Pr	8,49	1,33	9,52	9,08	6,83	3,88	2,29	4,37	5,01	5,22	11,5	2,19	12,3	7,44	7,39	7,39	10,4
Nd	29,9	5,39	36,5	30,5	25,7	14,2	8,35	16,1	17,2	19,0	39,3	7,94	45,8	27,1	27,2	27,2	36,0
Sm	5,68	1,11	8,52	5,84	5,06	3,05	1,88	2,91	3,47	3,93	8,03	2,13	8,68	5,59	5,10	5,71	6,34
Eu	1,64	0,32	2,50	1,57	1,25	1,05	0,68	0,73	0,98	1,16	2,33	0,74	2,08	1,40	1,44	1,41	1,57
Gd	4,44	0,83	8,18	4,26	4,22	2,52	1,21	2,49	2,95	3,12	6,55	1,20	8,49	4,83	4,06	4,49	5,48
Tb	0,62	0,12	1,15	0,57	0,70	0,41	0,18	0,38	0,40	0,42	0,91	0,16	1,22	0,70	0,59	0,64	0,77
Dy	2,70	0,55	5,76	2,38	2,94	1,75	0,88	1,81	1,85	1,84	4,02	0,66	5,98	3,32	2,50	2,94	3,16
Ho	0,47	0,09	1,15	0,38	0,51	0,35	0,17	0,32	0,30	0,31	0,66	0,11	1,02	0,57	0,38	0,49	0,51
Er	1,45	0,34	3,50	1,31	1,63	1,43	0,57	0,95	1,08	1,03	2,29	0,43	3,18	1,77	1,25	1,45	1,54
Tm	0,17	0,04	0,45	0,12	0,24	0,20	0,08	0,12	0,11	0,11	0,27	0,07	0,39	0,25	0,16	0,18	0,17
Yb	1,26	0,31	2,94	0,93	1,50	1,41	0,56	0,96	0,96	0,89	1,85	0,36	2,71	1,49	1,27	1,21	1,31
Lu	0,16	0,03	0,40	0,12	0,20	0,22	0,08	0,13	0,13	0,10	0,25	0,05	0,40	0,22	0,11	0,16	0,15
ΣREE	179,6	27,3	211,8	177,7	141,3	86,7	51,7	90,8	101,7	110,7	234,1	46,8	250,6	154,1	155,2	155,8	219,3
LREE	168,3	25,0	188,3	167,6	129,4	78,5	47,9	83,7	93,9	102,9	217,3	43,8	227,2	140,9	144,9	144,2	206,2
HREE	11,3	2,31	23,5	10,1	11,9	8,29	3,73	7,16	7,79	7,83	16,8	3,04	23,4	13,2	10,3	11,6	13,1
LREE/HREE	14,9	10,8	8,00	16,7	10,8	9,47	12,8	11,7	12,1	13,1	12,9	14,4	9,72	10,7	14,0	12,5	15,8
Moisture	47,5	35,8	44,2	49,2	50,6	51,7	46,4	32,7	46,0	35,8	41,9	45,8	43,5	44,4	29,2	44,3	40,3
Ash	6,09	31,8	10,7	11,5	13,7	8,76	12,9	24,5	6,46	26,2	12,9	9,47	19,2	20,1	57,0	11,9	24,8
Volatile matter	27,9	31,2	25,9	23,4	23,4	24,8	26,7	30,7	28,0	28,4	28,0	27,4	25,1	24,4	11,4	25,6	24,6
Fixed carbon	18,5	1,30	19,2	16,0	12,4	14,7	14,0	12,1	19,6	9,60	17,2	17,3	12,1	11,1	2,45	18,3	10,3
Volatile matter (DAF)	60,2	96,0	57,4	59,4	65,4	62,8	65,5	71,6	58,8	74,7	61,9	61,3	67,4	68,8	82,3	58,3	70,5
Fixed carbon (DAF)	39,8	4,00	42,6	40,6	34,6	37,2	34,5	28,4	41,2	25,3	38,1	38,7	32,6	31,2	17,7	41,7	29,5
Fuel ratio	1,51	24,0	1,35	1,46	1,89	1,69	1,90	2,52	1,43	2,96	1,62	1,58	2,07	2,21	4,66	1,40	2,39
Average depth	450,2	553,0	452,6	252,4	202,3	188,5	184,6	847,4	581,5	576,9	452,8	497,5	290,2	255,2	490,9	508,2	397,7

Table 2 shows that the CaO is the most abundant element in lignite samples followed by SiO_2, Al_2O_3 and Fe_2O_3. Samples also have considerable amounts of V_2O_5 and P_2O_5. The high SO_3 values are probably caused by sulphur bearing minerals in reducing enviroment such as pyrite and marcasite.

Table 2. Contents of major-element oxides (in %) of selected coal samples.

Major Oxide	A2	B2	B4	C2	C4
SiO_2	7,22	22,5	8,58	15,6	30,2
Al_2O_3	3,23	11,4	4,56	7,42	13,8
Fe_2O_3	2,03	5,67	5,57	3,22	6,02
MgO	1,06	2,54	3,78	2,46	5,28
CaO	79,6	33,8	52,7	41,1	20,3
Na_2O	ND	0,24	0,32	0,34	0,57
K_2O	0,34	1,28	0,60	1,14	1,91
TiO_2	0,13	0,58	0,26	0,30	0,78
P_2O_5	0,22	0,38	0,49	1,21	0,49
MnO	ND	ND	ND	ND	ND
BaO	0,16	0,39	0,30	0,32	0,50
SrO	0,10	0,11	0,19	0,10	ND
V_2O_5	ND	0,22	0,32	0,66	0,32
SO_3	5,37	20,4	21,8	25,2	19,1

4.2 Geologic Factors Controlling The Accumulation of The Uranium

The writers believe an abundance of field evidence favors the epigenetic hypothesis, the third of three opposing concepts, numbered here as the (1) diagenetic, (2) syngeneic, and (3) epigenetic hypotheses of origin (Denson 1959). The uranium in these deposits is of stratal epigenetic origin, secondarily derived by leaching of the volcanic materials in the Miocene rocks that at one time covered most of the region. The uranium was a primary constituent of finely disseminated in volcanic ash and other glassy extrusive rocks and that the uranium was leached from them by ground water percolating downward or moving laterally along aquifers near the lignite beds during weathering & devitrification and extracted by the lignite after coalification (Denson 1959, Figure 9). This concept was based largely on the fact that beds of uranium-bearing lignite intercalated with the tuffaceous rocks of the Kumalar formation.

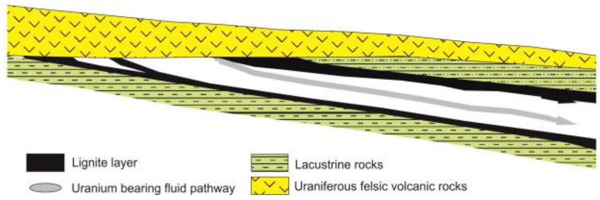

Figure 9. Accumulation modeling of the uranium in epigenetic-stratal deposits with stratigraphic and hydrologic settings. (after Galloway and Hobday, 1996).

In addition, it can be concluded that the greater concentrations of uranium occur in the upper parts of the stratigraphically highest lignite beds, which is compatible with other epigenetic lignite deposits.

5. Conclusion

- The uranium contents of studied coals from five boreholes at various depths were determined up to 1065 µg/g in ash.
- The determined uranium enrichments in study area can be considered as stratal epigenetic origin via volcanic intercalations within coals and epiclastic sedimentary rocks.
- The high correlations of U vs. (P_2O_5+V_2O_5) suggest that the U-bearing minerals in lignite layers can be either phospate or vanadate group minerals such as torbernite, autinite or carnotite.
- It can be stated that the groundwater enriched in oxidation agents causes the uranyl complexes to mobilize and hence a high concentration of uranium.
- The higher contents of uranium occurred in the upper parts of the stratigraphically highest lignite beds, which is compatible with other epigenetic lignite deposits

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