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MINERALOGICAL AND GEOCHEMICAL COMPOSITION OF THE ORES PROCESSED IN THE METALLURGICAL FACILITY IN KAVADARCI

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Abstract: *In the metallurgical facility in Kavadarci, Fe-Ni ores originating from several locations are processed, namely: Rzhanovo (Macedonia), Albania, Turkey, Indonesia, Guatemala, Africa. All Fe-Ni ores that are processed represent lateritic types of nickel-bearing ores that have a similar mineralogical composition, with the exception of the ores from Rzhanovo and Albania, which also contain minerals that are not only of lateritic origin but also of metamorphic origin. From the perspective of the impact on the quality of the environment in the Tikvesh region, it is important to take in to account the geochemical composition of these ores in terms of the presence of trace elements that can be risky for the environment, such as arsenic, thallium, cadmium, lead, and others.*

Keywords: *ores, lateritic, metamorphic, environment,*

МИНЕРАЛОШКИ И ГЕОХЕМИСКИ СОСТАВ НА РУДИ КОИ СЕ ОБРАБОТУВААТ ВО МЕТАЛУРШКИОТ ОБЈЕКТ ВО КАВАДАРЦИ

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Апстракт: *Во металургискиот објект во Кавадарци се преработуваат Fe-Ni руди кои потекнуваат од неколку локации и тоа: Ржаново (Македонија), Албанија, Турција, Индонезија, Гватемала, Африка. Сите Fe-Ni руди кои се преработуваат претставуваат латеритски типови на николносни руди кои имаат сличен минералошки состав, со исклучок на рудите од Ржаново и Албанија кои содржат и минерали кои немаат само латертитско потекло туку и метаморфно потекло. Од аспект на влијанието врз квалитетот на животната средина во регионот Тиквеш важно е да се води сметка за геохемискиот состав на овие руди од аспект на присуство на елементи во траги кои можат да бидат ризични за животната средина, како што се арсен, талиум, кадмиум, олово, и други. Со досегажните испитувања кои се направени во текот на 2015-2017 година е докажано дека нема значителна варијација во геохемискиот фон во почвите од регионот Тиквеш и дека самата преработка на овие руди во значителна форма не го промени локалниот геохемискиот фон.*

Клучни зборови: *руди, латерит, метаморфизам, животна средина,*

1. INTRODUCTION

Namely, in 1980, a Nickel production plant is built in Tikveš valley which started with the production in 1982 with an annually processing of about 1.5 million tons of latherite types of nickelous ore. Starting in 2005, in total, about 900,000 tones of ore come from the Ržanovo mine annually (southern parts of Kozuf Mountain in Tikveš Valley), and since then the smelter plant has begun processing ore originating from Albania, Turkey, Indonesia and Guatemala. Data concerning the composition (chemical and mineralogical) of these types of ores are presented in several publications (Boev et al., 1998, 2009; Boev and Bermanec, 2005). The operation of this plant in any case effects changes in the composition (mineralogical) of urban dust in the Tikveš valley.

The factory was built in the period of 1976-1980 with pyrometallurgical instalations for the processing of latherite nickelous ores, with a yearly capacity of 2 million tons of ore. The capacity of the smelter plant is about 16,000 tons of nickel in the form of ferro-nickel with a Ni content of 25 to 40 %. (Boev et al, 1996, Boev et al, 1999, Serafimovski et al, 2013).

Based on studies of mineral associations as well as the major mineral phases, the major nickel bearing minerals in the ores include magnetite, hematite, clinochlore, talc, sepiolite, magnesioriebeckite, lizardite, antigorite, actinolite, tremolite, chrysotile, dolomite, phlogopite, stilpnomelane, muscovite, quartz, albite, pyrite, maghemite, pirotine, digenite and millerite. Only five of the mentioned minerals are constantly present, including magnetite, hematite, clinochlore, talc and magnesioriebeckite (Boev, 1982, Boev et al, 1994, Boev et al, 1992, Maksimovic, et al, 1982, Boev et al. 2009).

A simplified scheme of technological process is as follows: the ore comes by transport belt from Rzanovo mine (as well as by rail or truck transportation from the port of Thessaloniki, Greece, for ore originating from mines in Indonesia), or from Albania or Turkey. The ore is then processed by crushing, grinding and homogenization. During these processes a certain amount of dust is generated due to the structure of the ore and its very fine grinding. From this plant the milled and homogenized ore is either inserted into the peletization plant or goes directly to another section for prereduction (rotary kilns). In these plants a certain amount of dust is also generated and emitted into the atmosphere, despite the built-in filtering equipment and chimneys. Then, the prereduced material is inserted into the electric furnaces were the material is melted and after additional reduction the liquid ferro-nickel is produced. The ferro-nickel is then refined into the refining plant and finally the granulated ferro-nickel is produced.
Based on this processes, it can be concluded that, during processing of the nickel ore, a certain amount of dust is generated and emitted into the air in the Tikveš region. Legal norms existing in the Republic of Macedonia specify 50 μg m-3 or less. It must be noted that the emission of dust, is observed from the factory for the producing of nickel in fact exceeds this limit (Baceva et al., 2011, 2012; Stafilov et al., 2012), (Boev et al, 1996).

2. METHODOLOGY

Mineralogical characterization

The mineralogical content of the collected dust samples was determined using an X-Ray Siemens D 500 equipped with an automated computer and a Cu-monochromatic lamp working at 40 KV and 30 mA. Quantitative analysis of the mineral phases present

was performed using the DIFRAC-11 software package and program support by EVAL and IDR.
The values given for the quantitative composition of the analyzed samples represent an average of 3 replicates. For QA and QC of the measurements referent materials and standards from various mineralogical compositions were used: BDS 17385/96 (standard for ore and ore concentrates for X-Ray diffraction quantitative phase analysis), ST SEV 3534-82 (SpS-quartz sand), ST SEV 2981-81 (KN-2, limestone), ST SEV 2980-81 (MpA-copper ore), USZ 47-2008 (granite “MGT-1”). In several cases standard addition method was applied by using some of the aforementioned RM and satisfactory values for the recoveries were obtained.
Differential thermal measurements were performed using the Q-1500-D instrument produced by MOM, Hungary, under the following conditions: sampling mass of 500 mg, sensitivity of DTA - 250 µV and DTG - 500 µV, heating rate, 10oC/min, temperature interval of measurement, 15-20oC to 1000oC and furnace ambient – air without turbulences.
The minerals present in the collected attic dust samples were determined by X-Ray diffraction while the quantitative analysis of the mineral phases present was performed using the software package DIFRAC-11 with program support from EVAL and IDR.

Laboratory methods

- Preparation of ores samples for determination of the presence of macroelements and trace elements (ISO-14507);
- Determination of macroelements and trace elements using the method of ICP-AES and ETAAS.

Instrumentation

The investigated elements are analyzed by applying atomic emission spectrometry with inductively coupled plasma (AES-ICP) and electrothermal atomic absorption spectrometry (ETAAS). By the application of AES-ICP the following elements were analyzed: Al, As, Ba, Ca, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Na, Ni, Pb, Sr, V and Zn. The instruments used for these analyzes were: Varian 715-ES Series ICP Optical Emission Spectrometer (Varian, USA) and Zeeman ETAAS Varian SpectrAA-640Z. Optimal instrument parameters for the two used techniques are given in Tables 1 and 2.

Table 1. Optimal instrumental parameters for ICP-AES system (Varian, 715ES)

RF Generator	
Operating frequency	40.68 MHz free-running, air-cooled RF generator
Power output of RF generator	700–1700 W in 50 W increments
Power output stability	Better than 0.1%
Introduction Area	
Sample Nebulizer	V- groove
Spray Chamber	Double-pass cyclone
Peristaltic pump	0-50 rpm
Plasma configuration	Radially viewed
Spectrometer	
Optical Arrangement	Echelle optical design
Polychromator	400 mm focal length
Echelle grating	94.74 lines/mm
Polychromator purge	0.5 L min^{-1}
Megapixel CCD detector	1.12 million pixels

Wavelength coverage			177 nm to 785 nm		
Conditions for program					
RFG Power		1.0 kW	Pump speed		25 rpm
Plasma Ar flow rate		15 L min^{-1}	Stabilization time		30 s
Auxiliary Ar flow rate		1.5 L min^{-1}	Rinse time		30 s
Nebulizer Ar flow rate		0.75 L min^{-1}	Sample delay		30 s
Background correction		Fitted	Number of replicates		3
Element	**Wavelength**	**Element**	**Wavelength**	**Element**	**Wavelength**
Al	396.152 nm	Cr	267.716 nm	Na	589.592 nm
As	188.980	Cu	324.754 nm	Ni	231.604 nm
Ba	455.403 nm	Fe	238.204 nm	Pb	220.353 nm
Ca	370.602 nm	K	769.897 nm	Sr	407.771 nm
Cd	226.502	Mn	257.610 nm	Zn	213.857 nm
Co	230.786 nm	Mg	279.553	V	292.401

Table 2. Optimal instrumental parameters за ETAAS одредување од Varian SpectrAA-604Z

Parameter	As	Cd	Co	Cr	Ni	Pb
Wavelength, nm	193.7	228.8	242.5	357.9	232.0	283.3
Spectral width slit, nm	0.2	0.5	0.2	0.2	0.2	0.5
Calibration mode	Peak area					
Lamp current, mA	10.0	4.0	7.0	7.0	4.0	5.0
DRY						
Temperature, ºC	120	120	120	120	120	120
Ramp time, s	55	55	55	55	55	55
Hold time, s	-	-	-	-	-	-
PYROLYSIS						
Temperature, ºC	1400	250	750	1000	900	200
Ramp time, s	10	5	5	10	5	5
Hold time, s	37	22	22	30	22	22
ATOMIZING						
Temperature, ºC	2600	1800	2300	2600	2400	2100
Ramp time, s	0.6	0.8	1.1	1.2	1.1	1
Hold time, s	2	2	2	2	2	2
CLEANING						
Temperature, ºC	2600	1800	2300	2600	2400	2100
Time, s	2	2	2	2	2	2
Ramp time, s	-		-	-	-	-
SHEATH GAS	Argon					

3. RESULTS AND DISCUSION (Mineralogical and geochemical implications)

RZANOVO ORES

The ores from the Rzhanovo ore deposit have a complex mineralogical composition and it is distinguished by the presence of the following minerals:

Table 3. Content of the main minerals in the Rzhanovo deposit (%)

Sample ID	R-1	R-2	R-3	R-4	R-5	Average values:
Hematite	58.00	55.00	51.00	56.00	58.00	*55.60*
Magnetite	0.00	0.00	4.00	0.00	0.00	*0.80*
Maghemite	0.00	4.00	0.00	0.00	0.00	*0.80*
Clinochlore	7.00	6.00	7.00	8.00	7.00	*7.00*
Talc	21.00	18.00	23.00	21.00	19.00	*20.40*
Mg-ribeckite	7.00	8.00	9.00	8.00	7.00	*7.80*
Sepiolite	0.00	4.00	3.00	3.00	4.00	*2.80*
Calcite	0.00	3.00	2.00	2.00	0.00	*1.40*
Dolomite	2.00	1.00	0.00	0.00	0.00	*0.60*
Kaolinite	3.00	0.00	0.00	0.00	3.00	*1.20*
Kutnachorite	1.00	0.00	0.00	0.00	1.00	*0.40*
Total per sample	**99.00**	**99.00**	**99.00**	**98.00**	**99.00**	**98.80**

From the data shown in the Table 3, it can be concluded that the main minerals in the Rzhanovo ore deposit are: hematite, magnetite, clinochlore, talc, magnesioribeckite, maghemite, serpiolite, kaolinite, calcite, dolomite, etc. (I fought). The content of nickel in these ores is shown in the Table. From the results shown, it can be concluded that the nickel content ranges from 0.86 to 0.93% Ni.
The Table 4, shows the chemical characteristics of the ores from the Studenata Voda ore deposit. From the results shown, it can be concluded that these are low percentage nickel-bearing ores.

Table 4. Chemical characteristics of nickel-bearing ores from Studena Voda

Lab. ID	Samp le ID	Ni, g/t	Cr, g/t	Fe, %	Al, %	Mg, %
7987	A-1	9319	6289	51.15	2.21	4.30
7988	A-2	9499	6145	50.61	2.24	4.14
7989	A-3	9473	6411	45.66	2.03	3.84
7990	A-4	9497	6412	50.82	2.24	4.22
7991	A-5	9370	6431	42.27	1.96	3.54
7992	R-1	9290	4656	41.51	0.86	8.41
7993	R-2	8620	3907	37.51	0.89	8.10
7994	R-3	8939	4865	39.70	0.86	7.96
7995	R-4	9241	4699	40.06	0.84	8.63
7996	R-5	9002	4905	41.56	0.87	7.43
7997	T-1	12140	2913	42.36	0.73	1.78
7998	T-2	11795	2498	36.56	0.58	1.77
7999	T-3	12653	2984	38.52	0.78	2.03
8000	T-4	12057	3100	37.04	0.83	1.80
8001	T-5	12767	2951	37.64	0.73	2.04

The geochemical association of the elements of these ores is typical for ophiolitic sequences and is represented by Ni, Fe, Co, Cr, Mg, and as a consequence of the young alpine metamorphic processes, an association of Pb, Zn, Cu, As, Sb, Mo,

ORES FROM ALBANIA

Table 5. Mineralogical characteristics of nickel-bearing ores from Albania

Sample ID	A-1	A-2	A-3	A-4	A-5	Average values:
Hematite	10.00	3.00	0.00	4.00	0.00	*3.40*
Clinochlore	0.00	0.00	0.00	6.00	8.00	*2.80*
Talc	5.00	10.00	5.00	3.00	0.00	*4.60*
Lizardite	10.00	6.00	9.00	8.00	0.00	*6.60*
Calcite	6.00	5.00	6.00	5.00	6.00	*5.60*
Kaolinite	2.00	3.00	0.00	0.00	0.00	*1.00*
Quartz	0.00	0.00	0.00	0.00	3.00	*0.60*
Muscovite+Ilite	5.00	5.00	8.00	6.00	7.00	*6.20*
Albite	0.00	0.00	0.00	0.00	3.00	*0.60*
Clinochrysotyle	0.00	0.00	0.00	0.00	9.00	*1.80*
Polydymite	0.00	1.00	0.00	0.00	0.00	*0.20*
Goethite+amorph.Fe	61.00	66.00	71.00	67.00	63.00	*65.60*
Total per sample	**99.00**	**99.00**	**99.00**	**99.00**	**99.00**	**99.00**

From the results shown in Table 5, it can be concluded that as the main minerals in the nickelifeorous, ores from Alabia are; hematite, clonochlor, talc, lizardite, calcite, kaolinite, quartz, chrysotile, etc. The presented results clearly indicate the close mineralogical characteristics of ores from Rzhanovo and ores from Albania.

ORES FRFOM TURKEY

Table 6. Mineralogical characteristics of nickel-bearing ores from Turkey

Sample ID	T-1	T-2	T-3	T-4	T-5	Average values:
Clinochlore	0.00	0.00	0.00	0.00	4.00	*0.80*
Lizardite	4.00	6.00	4.00	8.00	9.00	*6.20*
Mg-ribeckite	0.00	0.00	0.00	0.00	0.00	*0.00*
Sepiolite	0.00	0.00	0.00	0.00	0.00	*0.00*
Calcite	14.00	16.00	15.00	16.00	18.00	*15.80*
Dolomite	0.00	4.00	2.00	0.00	0.00	*1.20*
Kaolinite	0.00	0.00	4.00	0.00	0.00	*0.80*
Quartz	2.00	3.00	3.00	7.00	6.00	*4.20*
Muscovite+Ilite	10.00	9.00	7.00	7.00	0.00	*6.60*
Goethite+amorph.Fe	68.00	60.00	63.00	60.00	61.00	*62.40*
Total per sample	**98.00**	**98.00**	**98.00**	**98.00**	**98.00**	**98.00**

From the results shown in Table 6, one can notice a very great similarity in the mineralogical composition of the ores from Rzhanovo, Albania with the ores from Turkey. The main minerals in nickel-bearing ores from Turkey are: clinochlore, lizardite, magnesioribeckite, sepiolite, calcite, dolomite, kaolinite, quartz.

ORES FROM AFRICA

Lately, nickel-bearing ores from Africa (Ivory Coast) have also been brought to the metallurgical complex in Kavadarci, and the mineralogical composition of these ores is as follows; quartz, antigoreitg, lizardite, chloritg, chloritoid, magnesiopherite, goethite. This mineralogical composition points to the fact that it is a question of classical lateritic types of nickel-bearing ores. The geochemical elemental matrix of these ores is shown in the Table, and from the same it can be concluded that these ores are rich in nickel, and that these ores do not contain significant concentrations of elements that can represent a significant risk to the environment.

Table 7. Geochemical characteristics of nickel-bearing ores from Africa

Al	mg/kg	20571
Sb	mg/kg	60.3
As	mg/kg	1,11
Cu	mg/kg	40.9
Ba	mg/kg	98.3
Be	mg/kg	1.00
Bi	mg/kg	<1.00
B	mg/kg	<10.0
V	mg/kg	105
S	mg/kg	<500
Ga	mg/kg	8,02
Ge	mg/kg	<1.00
Fe	mg/kg	152773
Cd	mg/kg	<1.00
K	mg/kg	1169
Ca	mg/kg	12657
Sn	mg/kg	12,60
Co	mg/kg	318
Li	mg/kg	<10.0
Mg	mg/kg	62524
Mn	mg/kg	2125
Mo	mg/kg	732
Na	mg/kg	2015
Ni	mg/kg	15163
Pb	mg/kg	12.0
Se	mg/kg	2,33
Pd	mg/kg	<1.00
Ag	mg/kg	<1.00
Sr	mg/kg	177
Tl	mg/kg	<1.00
Ti	mg/kg	808
Cr	mg/kg	1872
P	mg/kg	158
Zn	mg/kg	68.5

Table 8. Geochemical characteristics of nickel-bearing ores from Rzhanovo, Albania and Turkey

Lab. ID	Sample ID	Mo, g/t	W, g/t	Pb, g/t	Zn, g/t	Cu, g/t	Co, g/t	Ni, g/t	Cd, g/t	Bi, g/t	Ag, g/t	As, g/t	Sb, g/t
7987	A-1	4	<20	30	196	77	394	9025	1	39	<1	22	52
7988	A-2	4	<20	30	199	88	425	9018	2	37	<1	22	52
7989	A-3	4	<20	29	201	76	421	8587	2	38	<1	23	51
7990	A-4	3	<20	30	198	84	412	9323	2	38	<1	19	50
7991	A-5	3	<20	30	202	78	423	8193	2	40	<1	21	52
7992	R-1	3	<20	20	53	18	155	4126	<1	32	<1	15	53
7993	R-2	3	<20	20	51	25	160	3987	<1	28	<1	15	27
7994	R-3	3	<20	20	52	18	164	4265	<1	32	<1	14	50
7995	R-4	3	<20	20	51	16	161	4192	<1	32	<1	20	48
7996	R-5	3	<20	22	53	19	161	4509	<1	34	<1	17	48
7997	T-1	11	<20	28	248	37	581	9200	5	26	<1	583	17
7998	T-2	10	<20	27	236	34	559	10791	5	23	<1	545	9
7999	T-3	11	<20	26	246	36	611	9652	5	22	<1	542	6
8000	T-4	10	<20	29	257	36	626	10056	5	26	<1	567	6
8001	T-5	11	<20	25	266	37	649	10053	5	25	<1	557	<5

Lab. ID	Sample ID	Ba, g/t	Sr, g/t	Cr, g/t	Li, g/t	V, g/t	Be, g/t	P, g/t	Fe, %	Al, %	Mn, g/t	Ti, g/t	Ca, %
7987	A-1	25	17	4996	8	163	<1	356	41.42	1.79	3169	315	2.22
7988	A-2	24	17	5182	8	163	<1	358	41.74	1.84	3154	311	2.16
7989	A-3	27	17	5196	8	166	<1	360	43.43	1.84	3153	325	2.29
7990	A-4	27	18	5248	9	164	<1	355	44.05	1.87	3238	325	2.37
7991	A-5	27	18	4736	8	166	<1	364	41.35	1.89	3284	324	2.27
7992	R-1	14	66	3736	17	93	<1	20	33.74	0.77	1757	184	0.54
7993	R-2	30	140	3499	14	81	<1	18	32.05	0.70	2197	179	1.10
7994	R-3	14	66	2298	17	94	<1	19	34.16	0.80	1764	180	0.54
7995	R-4	14	70	4227	17	92	<1	23	33.46	0.80	1820	176	0.64
7996	R-5	19	52	4517	16	93	<1	20	34.72	0.82	1673	200	0.46
7997	T-1	108	16	2674	6	71	3	31	37.34	0.61	4005	57	6.30
7998	T-2	86	15	2234	5	61	3	40	35.57	0.47	3703	57	6.83
7999	T-3	99	14	2144	6	61	3	29	35.60	0.65	4137	50	6.80
8000	T-4	99	16	2708	7	68	3	29	35.86	0.66	4124	50	6.71
8001	T-5	100	16	2445	6	62	3	39	31.98	0.57	4498	50	6.85

Lab. ID	Sample ID	Mg, %	K, g/t	Na, g/t	Tl, g/t	Ga, g/t	Sn, g/t	La, g/t	Y, g/t	Zr, g/t
7987	A-1	3.41	242	114	<5	<1	2	<50	<5	<5
7988	A-2	3.27	219	180	<5	<1	2	<50	<5	<5
7989	A-3	3.53	237	112	<5	<1	2	<50	<5	<5
7990	A-4	3.64	221	162	<5	<1	2	<50	<5	<5
7991	A-5	2.72	234	165	<5	<1	2	<50	<5	<5
7992	R-1	3.69	249	127	<5	<1	2	<50	<5	<5
7993	R-2	4.27	638	227	<5	<1	2	<50	<5	<5
7994	R-3	3.70	244	105	<5	<1	2	<50	<5	<5
7995	R-4	3.72	250	98	<5	<1	2	<50	<5	<5
7996	R-5	3.93	457	160	<5	<1	2	<50	<5	<5
7997	T-1	1.31	58	120	<5	<1	2	<50	<5	<5
7998	T-2	1.40	65	114	<5	<1	3	<50	<5	<5

7999	T-3	1.57	57	114	<5	<1	2	<50	<5	<5
8000	T-4	1.50	61	107	<5	<1	3	<50	<5	<5
8001	T-5	1.63	75	103	<5	<1	2	<50	<5	<5

4. CONCLUSIONS

Taking into account all the data stated in this paper, which refer to the mineralogical and geochemical characteristics of the nickel ores from Rzhanovo, Albania, Turkey and Africa, the following can be concluded: - all processed ores have a similar genetic origin, i.e. all the ores that are processed in the metallurgical complex in Kavadarci are originally lateritic ores, with some being primary (Africa) and others being lateritic reprecipitated type of ores that have subsequently been metamorphically changed. - all ores have a similar geochemical matrix, which is characteristic of laterite types of ores (increased concentrations of Ni, Mg, Fe, Si) and low concentrations of sulfur. - the concentrations of trace elements from the group of arsenic, antimony, lead, zinc, cadmium, are within the limits that are characteristic for these types of ores, with the exception of the ores from Turkey (which have increased concentrations of arsenic). However, it should be mentioned that nickel-bearing ores from Turkey are no longer used in the metallurgical process. The similar elemental matrix in the ores will not significantly affect the change of the local geochemical background in the region of Tikvesh

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