THE EFFECTS OF THE DISTRIBUTION COEFFICIENT ON THE PROPERTIES OF THE REFRACTORY CASTABLES FROM THE CHAMOTTE WASTE AND CLAY "RAPAJLO"

Marina Jovanović, Adnan Mujkanović, Nadira Bušatlić
University of Zenica, Faculty of Metallurgy and Technology
Travnička cesta 1, Zenica
Bosnia and Herzegovina

Keywords: conventional castables, chamotte waste, clay, particle size distribution

ABSTRACT
The paper examined the conventional castables composed of a mixture of high aluminate cement with 70% Al_2O_3, chamotte waste with 42% Al_2O_3 and brick clay ,,Rapajlo". The influence of particle size distribution on density, porosity, water absorption, modulus of rapture and cold crashing strength was investigated. Particle size distribution was determined by the Dinger and Funk equation, using four different coefficient q. With the reduction of coefficient q, the amount of fine fractions is increased and the amount of larger fractions is decreased. Two series of samples were investigated. In the first series as the smallest fraction of all mixtures with different factor q was used high aluminate cement and water-cement ratio 0.485. Increasing the amount of cement has led to increasing the density and strength of castable. The increased amount of water has led to an increase in porosity. In the second series, the amount of cement and water remained the same, a part of the superfine fraction is replaced by clay. Replacing chamotte waste with clay leads to a slight increase in the density and strength of the material. When a part of the cement is also replaced with clay it slight reduce the density and strength of the material, because part of the water is placed between sheets of clay minerals and leads to an increase in volume, and at the same time reducing the amount of water needed for the reaction with cement.

1. INTRODUCTION
Refractory materials are an important component for all high-temperature processes. Thanks to its properties, refractory materials are widely used in many industries such as metallurgy, chemical, ceramic and cement industries, glass industry, petroleum industry, iron and steel industry, non-ferrous metal industry, as well as numerous other indutrial branches. Nowadays unshaped refractory materials, especially refractory castables and mortars, are suppressing classic shaped refractory products. Refractory castables are composite materials of complex blends of refractory aggregates, binders, fillers, additives and water. Physical, mechanical and thermal properties of refractory castables depend on the density of components packing in concrete mix [1, 2, 3, 4]. Naturally, the properties of refractory castables depend on the share of cement and aggregate in the blend, as well as the content of reactive components. The effect of additives is mainly focused on their influence on water content, fluidity, time of handling and strength. Conventional refractory castables contain 15 - 30% of calcium-alumina cement which introduces 3 to 8% CaO as one of the most destructive refractory compounds in castables [5]. In order to reduce the amount of cement or water intake, a new group of refractory castables has been developed, called defloated refractory castables. In this

group some cement fractions have been replaced by fine particles such as microsilicon (or silicate dust), chromium oxide and reactive alumina [6].
Conventional refractory castables of different granulometric composition was studied in this paper. In addition to the influence of the granulometric composition, the influence of the replacement the cement part and the smallest fraction of chamotte waste with clay was investigated.

2. EXPERIMENTAL WORK

In this experimental work the effect of the distribution of grain size on the properties of refractory castables on the basis of chamotte waste and the influence of the replacement the part of the cement and the finest fraction of chamotte waste with the clay was investigated. Particle size distribution was obtained by the Dinger - Funk equation [1, 2, 3, 4]:

$$CPFT = \frac{D^q - D_{min}^q}{D_{max}^q - D_{min}^q} \times 100 \qquad (1)$$

Where is:
D – particle size,
D_{min} – minimum particle size,
D_{max} – maximum particle size,
q – distribution coefficient.

Coefficients q values of 0.37, 0.34, 0.31 and 0.28 are selected which, according to literary sources, provide conventional vibratory refractory castables with a cement content of over 20%. For the minimum grain size the value of 0.3 μm was taken and the maximum was 4000 μm because the test was performed on 4 × 4 × 16 cm prisms, where the larger grain size would be unfavorable. Seven fractions (0.3 to 75 μm, 75 to 150 μm, 150 to 300 μm, 300 to 500 μm, 500 to 1000 μm, 1000 to 2000 μm, 2000 to 4000 μm) were used for the study. Two series of tests were carried out. In the first series, only cement and chamotte waste were used, where the fraction from 0.3 to 75 μm consisted only of cement, since according to the declaration the used cement has 88% particles below 63 μm. For other fractions, a chamotte waste was used.
In the second series of tests, cement was used in the amount of 20.6% and the remainder of the fraction 0.3 - 75 μm was clay "Rapajlo", sived on a 75 μm sive. A fraction of 75 to 150 μm was also clay "Rapajlo". Other factions were from chamotte waste. The mixtures were made according to JUS B.C8.022 standard with a water-cement ratio of 0.485, corresponding to a water content of 10%. Sodium hexametaphosphate was used as a deflocculant in an amount of 0.2%.
For the preparation of castable mixtures the following raw materials were used: high alumina cement Gorkal 70, chamotte waste (“Šamoter” d.o.o. Zenica) and clay “Rapajlo” with chemical composition shown in table 1. DTA/TG analysis and approximate minaralogical composition of the clay are presented in Figure 1 and Table 2. The clay is an illite-kaolinite type with a large proportion of quartz and muscovite and a smaller amount of goethite. The micrographs of the clay and chamotte waste particles recorded on the binocular microscope OLYMPUS BX60 Mare shown in the Figure 2 and on the PCE-MM200 Digital Microscope in Figure 3. It can be seen that the grains of chamotte are coarser than clay grains. The grain size distributions of clay and chamotte waste obtained by laser method are shown in Figure 4 and Figure 5. The types and calculated quantities of materials for selected values of *q* are given in table 3.

Table 1. The chemical composition of the raw materials

Component	Chemical composition (wt. %)		
	Cement	Chamotte waste	Clay
SiO_2	< 0.5	49.75	67.6
Al_2O_3	69 - 71	42.8	12.9
Fe_2O_3	< 0.3	1.93	6.86
TiO_2	-	1.35	0.55
CaO	-	< 0.01	0.03
MgO	-	0.12	0.76
MnO	-	0.04	-
$Na_2O + K_2O$	< 0.3	2.84	3.87
L.O.I.	-	-	7.28

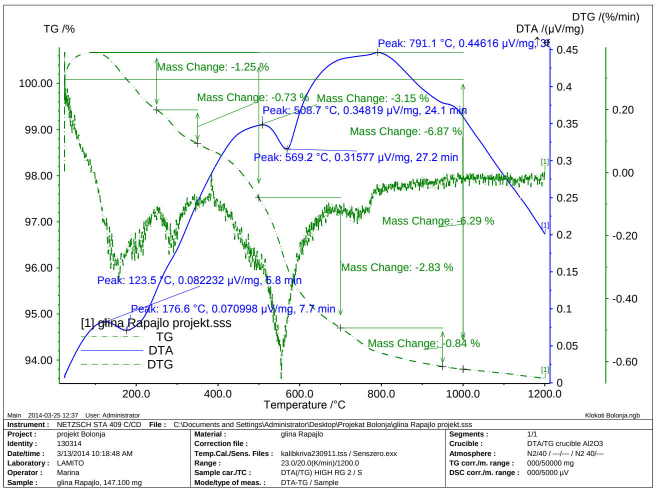


Figure 1. DTA/TG of the clay [7]

Table 2. Approximate mineralogical composition of clay [7]

Mineral	Illite	Muscovite	Kaolinite	Goethite	Quartz
Mineral content (%)	29.5	10.8	22.6	6.8	36.6

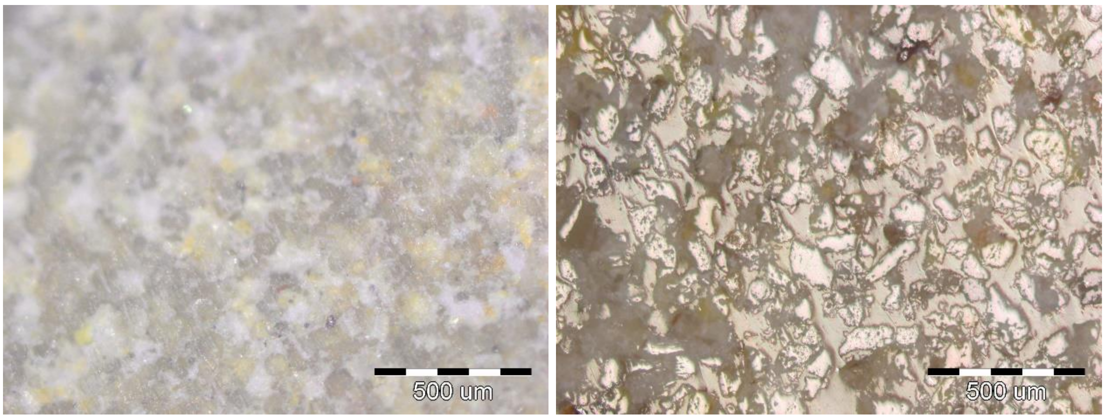


Figure 2. Optical micrographs of the clay (left) and chamotte waste (right)

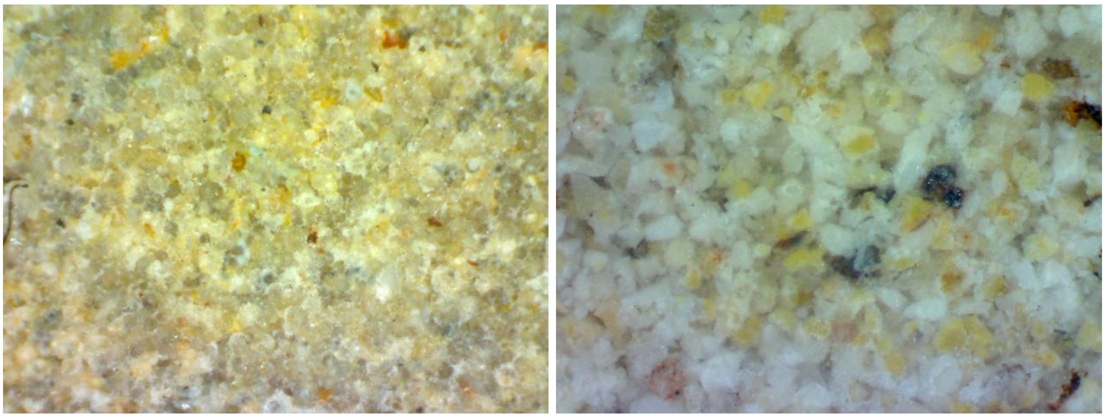


Figure 3. Optical micrographs 200X magnitude of the clay (left) and chamotte waste (right) PCE-MM200 Digital Microscope

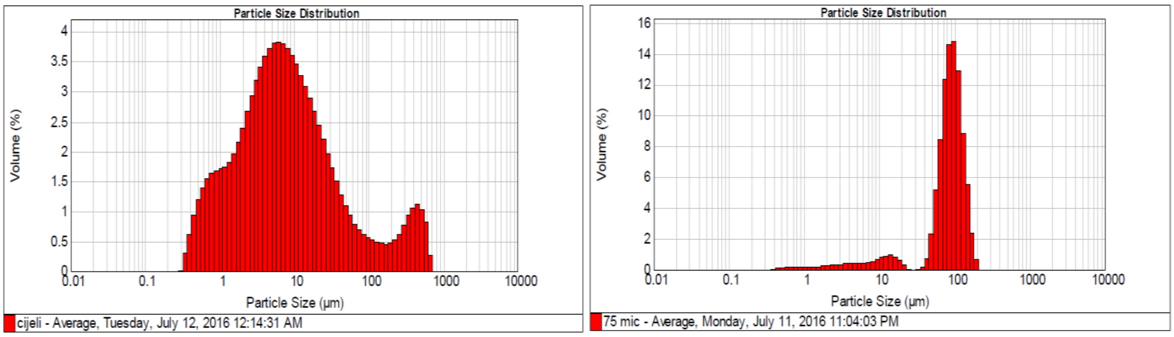


Figure 4. Particle size distribution of clay "Rapajlo": all fractions (left) and fraction 75 – 150 μm (right)

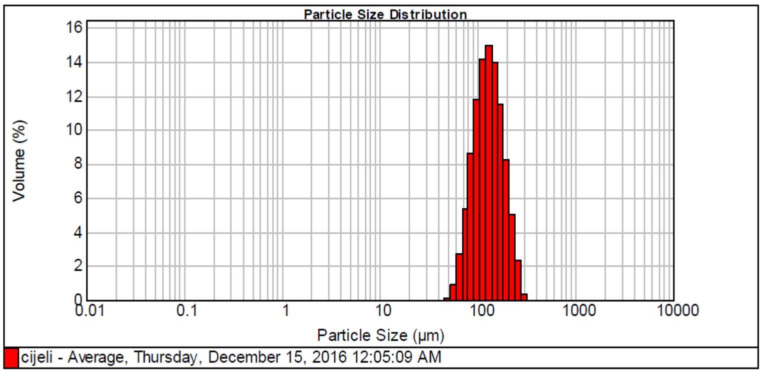


Figure 5. Particle size distribution of chamotte waste fraction 75 – 150 μm

Table 3. The type and amount of materials

Fraction [μm]	Amount of fraction [%]				Raw material	
	q=0.37	q=0.34	q=0.31	q=0.28	1st series	2nd series
0.3 – 75	20.6	20.6	20.6	20.6	Cement	Cement
0.3 – 75	0	2.22	4.61	7.19	Cement	Clay
75 – 150	6.92	7.15	7.38	7.56	Chamotte	Clay
150 – 300	8.94	9.07	9.14	9.19	Chamotte	Chamotte
300 – 500	8.22	8.18	8.12	8.00	Chamotte	Chamotte
500 – 1000	13.96	13.65	13.28	12.87	Chamotte	Chamotte
1000 – 2000	18.04	17.27	16.46	15.62	Chamotte	Chamotte
2000 – 4000	23.32	21.86	20.41	18.97	Chamotte	Chamotte

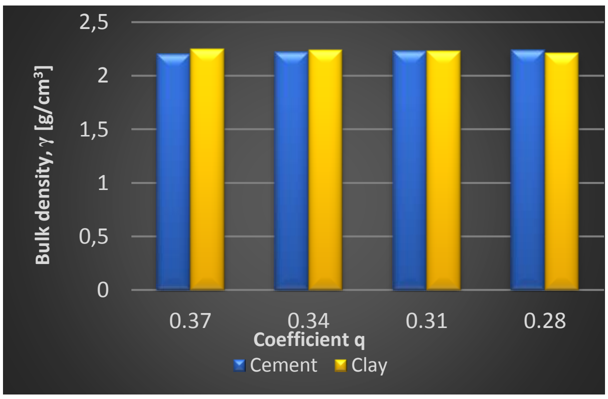
Mixing the prepared materials and filling metal molds was carried out in a laboratory mixerfor cement paste. Afterwards the samples were left in the molds for 24 hours, and after 24 hours were demolded and placed in a container with water for 3 days. After 3 days they were removed from the water and allowed to dry first in air for one day, and then in an oven for another day at 110 ± 5 °C. The following properties of refractory castableswere investigated: apparent density, apparent porosity, water absorption, cold crashing strength, flexural strength and structure on optical microscope.

3. RESULTS AND DISCUSSION

Table 4. The results for tested castables

Coefficient q	Property of castable									
	Bulk density γ[g/cm^3]		Apparent porosity P_p[%]		Water absorbtion U_v[%]		Modulus of rapture σ_s [MPa]		Cold crashing strength σ_p [MPa]	
	I	II	I	II	I	II	I	II	I	II
0.37	2.2	2.25	9.2	10.05	4.3	4.46	11.06	14.7	78	80
0.34	2.22	2.24	9.4	10.17	4.64	4.53	11.35	13.7	83	79
0.31	2.23	2.23	9.7	9.99	4.71	4.47	11.93	12.9	85	64
0.28	2.24	2.21	10.5	10.9	5.0	4.95	12.6	12.2	87	61

I – castable with cement only, II –castable with clay



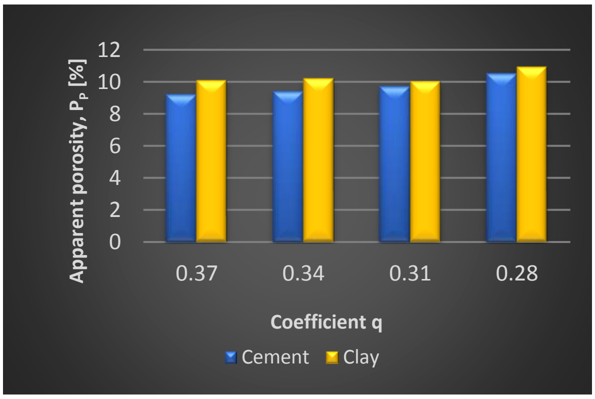


Figure 6. Bulk density and apparent porosity vs coefficient q

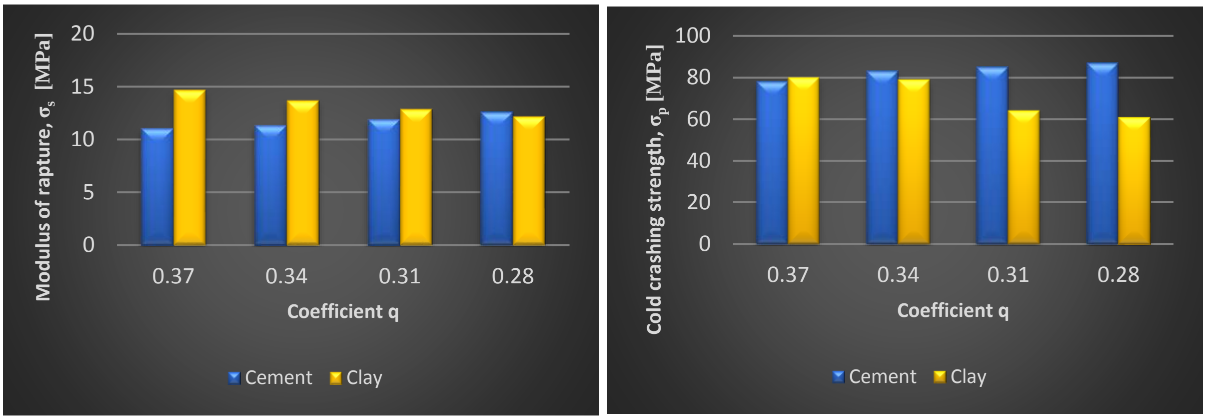


Figure 7. Modulus of rapture and cold crashing strength vs coefficient q

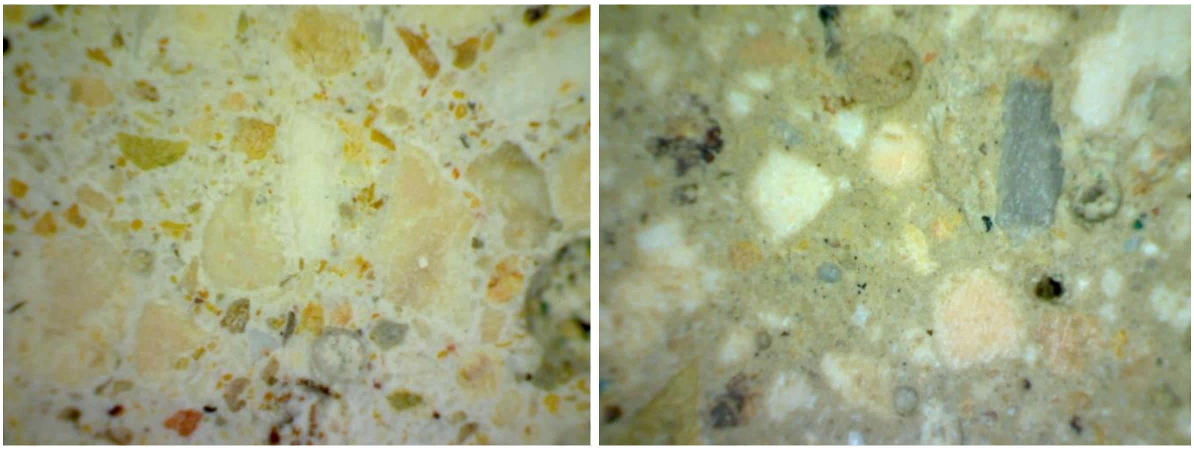


Figure 8. Optical micrographs 60X magnitude of the castable with cement and chamotte (left) and castable with cement, chamotte and clay (right) for q 0.31 by PCE-MM200 Digital Microscope

By reducing the coefficient q in the equation for the calculation of the granulometric composition of castable, the quantities of large fractions above 300 μm are reduced and the fractions below 300 μm are increased (Table 3). These changes also affect the tested properties, so that the density of the obtained castables increases slightly (Figure 6 left, blue bars), and also increases the strength of the castables (Figures 7 blue bars). However, the apparent porosity and absorption of water also increases. Strength increase can be explained by increasing the amount of hydraulic binder i.e. cement.

Increasing the amount of small fraction improves the packing of the particles, and thus the density. With the reduction of coefficient q increases the addition of cement, and also the amount of added water. It is known that the porosity of concrete is higher the greater the amount of water added, which is manifested by the increase in apparent porosity and absorption of water with the reduction of coefficient q.

When the clay replaces chamotte in the fraction 75 - 150 μm (q = 0.37), a small increase in density is obtained, because the clay in this fraction contains a lot of small particles (Figure 4 right) which provide better packing and give higher density. Fractions of clay and chamotte were obtained by dry sieving. However, clay is very hard to sieve dry, because it is easy to moisten while standing in the air, so its tiny particles agglomerate. Therefore, the results of porosity and water absorption are disproportionate to the change in coefficient q.

With the addition of the clay fraction below 75 μm, the reduced density and cold crashing strength of castables is obtained (Figures 6 and 7). The structure of clay minerals is like sheets and when clay minerals come into contact with water, the water is placed between the sheets and leads to increased volume or swelling so that samples with an increased amount of clay fraction below 75 μm show a decrease in density. Reduced material density leads to reduced cold crashing strength.

The density and modulus of rapture of the samples with clay are generally higher in relation to non-clay samples (Figures 6 and 7). Clay below 75 μm contains more clay minerals that with water are most likely to create a structure with a smaller number of internal defects affecting modulus of rapture. This can be evidenced by the fact that increased apparent porosity has increased density, probably due to decreased closed porosity. Clay bands part of the water so that the amount of water for the reaction with the cement is reduced, which is reflected by decreasing the cold crashing strength with increasing clay content (Figure 7).

Figure 8 shows the morphology of castables with q 0.31 and it can be seen that the matrix in castable with clay more homogenized.

The cold crashing strength of the tested castables ranges from 61 to 87 MPa and the bulk density from 2.2 to 2.25 g/cm^3 which is better compared to some commercial castables of similar composition (Table 5). The best results in terms of cold crashing strength, modulus of rupture and density gives a castable in which only a fraction of 75 to 150 μm of chamotte is replaced by clay of the same fraction.

Table 5. Strength and density of castables

Data source	**Property of castable**		
	Bulk density γ[g/cm^3]	**Cold crashing strength** σ_p [MPa]	**Modulus of rapture** σ_s [MPa]
RHI [8]	2.26	60	-
KSKM [9]	2.0 – 2.1	25	-
AGC [10]	2.05 – 2.15	-	4
SKG [11]	2.1	25	-
Tested castables	2.2 – 2.25	61 - 87	11.06 – 14.7

4. CONCLUSION

It has been shown that the decrease of factor q increases the amount of fine fractions and decreases the amount of coarse fractions leading to an increase of density and strength of the refractory castables. At the same time, there has been an increase in the apparent porosity and water absorpition. Increased strength was due to the increasing the amount of cement and increasing porosity was caused by increase in the amount of added water. The clay can be used instead of part of the cement and the finest fraction of chamotte waste, because it increases the modulus of rapture and density. Although the clay reduces cold crashing strength it is still in the same range or even better than the castables which can be found on the market.

5. REFERENCES

[1] Myhre B.: *The effect of particle size distribution on flow of refractory castables*, 30^{th} Annual Refractories Symposium, St. Louis, 1994.,

[2] Myhre B., Hundere A. M.: *The use of particle size distribution in development of refractory castables*, XXV ALAFAR Congress, San Carlos de Bariloche, 1996.,

[3] Myhre B.: *Particle size distribution and its relevance in refractory castables*, 2ndIndia International Refractory Congress, New Delhi, 1996.,

[4] Parija A.: *Low cement high alumina castables: effects of distribution coefficients*, diplomski rad, National institute of technology Department of ceramic engineering, Roukela, 2013.,
[5] Petrovski P., Bušatlić I.: *Cementi i druga neorganska mineralna veziva*, HIJATUS, Zenica 2006.,
[6] Samadi H.: *The effect of microsilica addition on Iranian based chamotte castables*, Pars Refractories Co., Tehran (www.researchgate.net) (11/2016), 2002.,
[7] Tutić E. et al.: Approximate mineralogical composition of clays, 1st International Student Conference on mining, metallurgy, chemical engineering, material science and related fields, Bor Lake, Bor, Serbia October 3rd 2014.,
[8] Prospekt firme RHI: Lining concepts for the nonferrous metal industry.,
[9] Prospekt firme Konya Selçuklu Krom Magnezit: Lining concepts for the nonferrous metal industry.,
[10] Prospekt firme AGC Plibrico.,
[11] www.skgrl.com (11/2016).