

*Araştırma Makalesi - Research Article*

## A New Chemical Method to Predict Grindability Index (HGI) for Limestones

### Kireçtaşları İçin Yeni Bir Kimyasal Öğütme İndeksi

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#### ABSTRACT

In this study, limestone samples (a total of 58 sample) were investigated in terms of their grindability and chemical composition. Grindability tests were carried out on standard HGI mill. Limestone samples were collected from two different limestone quarry and they were characterized in terms of their chemical composition. In the order of technological utilization, grindability nature of limestones is as significant as their chemical composition. Chemical composition of the limestone samples from the quarries under investigation differs and so does the grinding index, i.e. HGI (Hardgrove Grinding Index). In the context of this study, chemical composition data of limestone samples were correlated with the results of the grinding tests (HGI values). In addition, abovementioned correlations were provided as graphical demonstrations in this context. After these abovementioned graphical demonstration of the relationships between HGI values and chemical composition data, the role of the each chemical composition item in terms of grindability was understood. Based on this understanding, an empirical formula employing the chemical composition data was proposed to predict HGI.

**Keywords-** *Limestone Quarry, Chemical Composition, HGI, Modeling, Limestone Grinding*

#### ÖZ

Bu çalışmada, kireçtaşı numuneleri (toplam 58 numune) öğütülebilirlik ve kimyasal bileşim açısından incelenmiştir. Öğütülebilirlik testleri standart HGI değirmeninde gerçekleştirilmiştir. Kireçtaşı örnekleri iki farklı kireçtaşı ocağından alınmış ve kimyasal bileşimleri açısından karakterize edilmiştir. Teknolojik kullanım sırasına göre, kireçtaşının kimyasal bileşimi kadar öğütülebilirliği de önemlidir. İncelenen ocaklardan alınan kireçtaşı numunelerinin kimyasal bileşimleri ve öğütme indeks (Hardgro ve Öğütülebilirlik İndeksi) değerleri farklıdır. Bu çalışma kapsamında, kireçtaşı numunelerinin kimyasal bileşim verileri ile öğütülebilirlik test sonuçları (HGI değerleri) ile ilişkilendirilmiştir. Bunun yanında, belirtilen bu ilişkilendirmeler grafiksel gösterim olarak bu kapsamda verilmiştir. HGI değerleri ile kimyasal bileşim verileri arasındaki ilişkilerin grafiksel gösteriminden sonra, her bir kimyasal bileşim öğesinin öğütülebilirlik açısından rolü anlaşılmıştır. Bu çalışma kapsamında HGI'yi tahmin etmek için kimyasal bileşim verilerini kullanan ampirik bir formül önerilmiştir.

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## I. INTRODUCTION

Limestone which has calcite as dominant mineral is being utilized as a raw material by human beings for a long time. Limestone has wide field of utilization and it is mainly demanded by the construction industry. In addition to utilization in terms of construction purposes, it has also potential usage in metallurgy, agriculture. Flue gas desulfurization is also another field of industry which limestones are widely utilized as raw materials. In many industries abovementioned however, in addition to its specific chemical composition limestone should also be ground and have the form of powder.

Limestones are formed mainly as sedimentary rocks and the main formation conditions include climate and absence of clay or sandy material [1]. Limestones have mostly calcium carbonate or magnesium carbonate compounds and some impurities like iron, aluminum, silica, and sulfur presents at different amounts (depending on the type and formation) in their structures. Limestones are mostly classified in terms of their origin, geological formation, mineralogical structure, crystal form, chemical composition, color and hardness. Limestones are also classified depending on their carbonate amounts. In terms of carbonate classification, Folk [2,3] and Dunham [4] classification systems are the most common ones.

Grindability of a material is characterized by the amount of the work/energy required for a unit weight/volume of material to be ground to a specific size distribution [5]. Ore grindability is represented by the Bond work index value [6] and Mucsi et al. [5] have summarized the most widely known and utilized grindability tests as Bond, Hardgrove and Zeisel methods [7-9]. Many researchers [10-14] have employed BWI to address the limestone grindability. Musci et al. [5] have investigated the grindabilities of andesite, basalt, clinker, limestone and quartz. Referring back and forth to the study of Mucsi et al. [5], although it has not been addressed and emphasized, chemical composition along with the HGI values of abovementioned materials (andesite, basalt, clinker, limestone and quartz) were provided. As it is obvious and expected, change in chemical composition results in a change in grindability values, i.e. HGI in this case. According to the Seo et al. [15], difference in grinding efficiency depends on the chemical composition and crystal structure. Same authors [15] have employed HGI to understand the grindabilities of limestones and they have claimed the fact that HGI measurement is rather easier and not as much time consuming as compared to BWI (Bond Work Index) measurement. HGI method, although some disadvantages [16-18] and repeatability issues [19, 20] are associated with, is widely known and mostly addressed in terms of grindability specifications (especially for coal in terms of international trade) and mostly considered for design and optimization of grinding circuits [16].

In the study of Mendis et al. [21], chemical composition, moisture content and particle size are considered as the effective factors of raw material in terms of grindability and authors have claimed the fact that grindability is mainly affected by the chemical composition. According to Kural and Ozsoy [22], chemical and physical factors of raw material are the main reasons in terms of low grindability. Mendis et al. [21] have addressed the percentages of  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{Fe}_2\text{O}_3$ ,  $\text{CaO}$ ,  $\text{MgO}$ ,  $\text{Cl}$ ,  $\text{SO}_3$ ,  $\text{Na}_2\text{O}$ , and  $\text{K}_2\text{O}$  in limestones in order to assess relationships between grindability and chemical composition. In the study of same authors [21], it was shown that increase in  $\text{CaO}$  (%) resulted in an increase in grindability. Although some chemical composition data is interrelated to grindability, authors [21] have employed no.212 sieve residue rather than any grindability index values. Although employment of the method abovementioned to address the relationship between grindability and chemical composition is acceptable to some extent, assessment in terms of grindability index (HGI or BWI) could have been more reasonable.

Researchers [23] have investigated the effect of chemical composition on Portland cement clinker grindability. In addition, Ürünveren et al. [24] have tried to predict HGI values of Afşin-Elbistan (Turkey) Low-grade coals based on proximate analysis and ash chemical composition. Although prediction of HGI (for coal mostly) is more of concern by many researchers [25-27], some other researchers [28] have investigated the grindability behavior of clinker and colemanite. Although no recent research have investigated the relationship between chemical composition and grindability index values (HGI) as in the same context of this study, still some studies [23,29] have been conducted in terms of clinker composition and grindability.

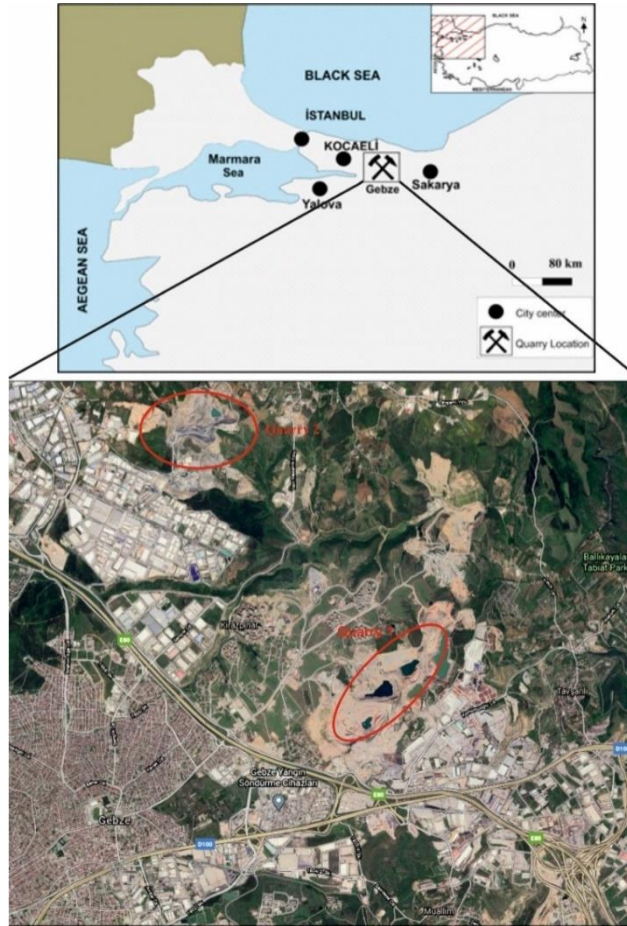
Grindability is most of the time significant issue in terms of energy requirements of milling, and pulverized limestones are generally desired for the further utilization of limestones. In this regard, not only chemical composition data of limestones are mostly referred but also the grindability is. While having chemical composition data on hand, if further utilization needs milling, field engineers should be aware of the grindability values of the corresponding limestones. So, in this context, chemical composition data can be interrelated with grindability index values, i.e. HGI, which is not only time consuming experimental procedure but also needs

expertise and significant amount of laboring. In this study, two different limestone quarries (Quarry-1 and Quarry-2) were taken into consideration and a total of 58 limestone samples, i.e. 22 samples from Quarry-1 and 36 from Quarry-2, were collected. Collected samples were analyzed in terms of their chemical composition and grindability tests (HGI) were performed. These analyzes abovementioned resulted in an understanding of the relationship between grindability values and chemical composition data for limestones. Having evaluated the relationships between each chemical component and HGI values, a new chemical grinding index for limestones was proposed.

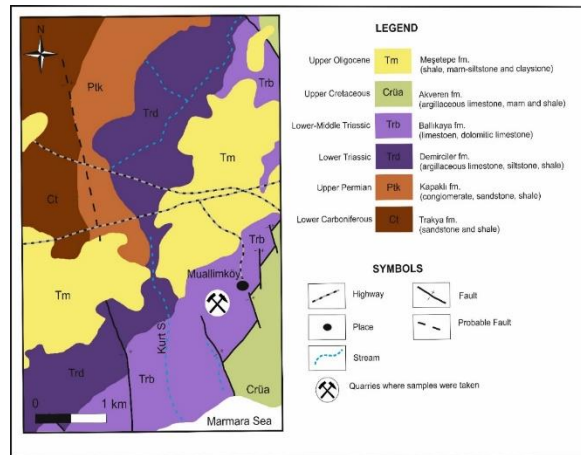
## II. MATERIAL AND METHOD

### A. Samples and Sample Preparation

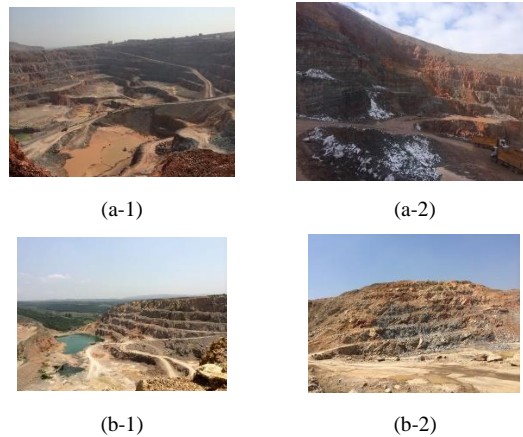
Limestone quarries under investigation are located in Gebze/Kocaeli, Turkey. Number of limestone quarries are being operated in the region where this study conducted. Location map of the quarries where samples are collected from is provided in Figure 1. Geological map of the study area (adapted from the study of Gedik et al. [30]) was provided in Figure 2. General view of limestone quarries studied is provided in Figure 3. Laboratory work along with sampling in the quarry is schematized in Figure 4.



**Figure 1.** The location map of the study area



**Figure 2.** Geological map of the study area (adapted from the study of Gedik et al. [30])



**Figure 3.** General view of the limestone quarries under investigation, refer to (a-1) and (a-2) for Quarry-1 & (b-1) and (b-2) for Quarry-2.



**Figure 4.** Schematic representation of the laboratory work and sampling in the quarry.

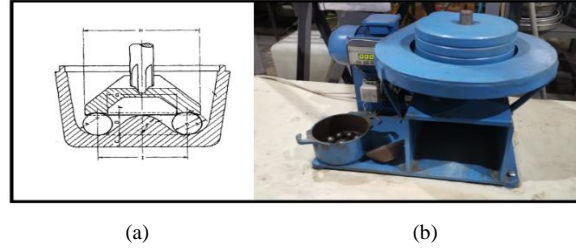
### B. Grinding Tests

Grinding tests were carried out with the standard HGI mill employment. All of the 58 samples were characterized in terms of their HGI values. HGI tests was carried out as in the standard described in ASTM D409-71[8]. In order perform HGI tests, samples were crushed step by step to the size fraction of  $-1.18+0.6$  mm, as standard implies for the feed size of HGI mill. After the grinding with HGI mill, final amount(s) ground under 75  $\mu$ m were noted. This final amount (ground under 75  $\mu$ m with HGI mill) was placed in the equation (1) and HGI value of that specific sample is determined accordingly.

$$\text{HGI}=13.6+6.93.w \quad (1)$$

where  $w$  is the weight of the test sample passing through 75  $\mu\text{m}$  sieve.

HGI tests were realized and corresponding HGI values for each limestone samples (a total of 58) were recorded. Representation of HGI testing environment is provided in Figure 5.



**Figure 5.** Representation of HGI testing environment, (a) Grinding elements of Hardgrove Machine (adapted from the study of Tichanek [16]), (b) HGI mill employed in this study.

### C. Chemical Analyzes

Representative samples of limestone (a total of 58) were collected from the quarries (Quarry-1 and Quarry-2). Collected samples were initially prepared for chemical analysis. This preparation includes size reduction and grinding. In terms of chemical analysis, standard method (ASTM C1271-99 [31]) was taken into consideration and the analysis was carried out with X-ray Fluorescence (XRF). A laboratory view of the XRF equipment (Philips PW-2404) employed for the chemical analyses is provided in Figure 6.



**Figure 6.** Laboratory view of the XRF equipment.

## III. RESULTS AND DISCUSSION

Limestone samples collected from the quarries were investigated in terms of their chemical composition and their grindability behavior. In order to understand the grindability behavior, HGI tests were performed. The chemical composition data and the corresponding HGI value of each sample were tabulated in Table 1 and Table 2, respectively for Quarry-1 and Quarry-2. As it is previously explained, a total of 58 limestone samples (22 of which is collected from Quarry-1 and 36 of which is collected from Quarry-2) were analyzed and corresponding data was tabulated.

**Table 1.** Chemical composition data and HGI values of the limestone samples from Quarry-1.

SAMPLE	HGI	MgO (%)	SiO <sub>2</sub> (%)	CaO (%)	Fe <sub>2</sub> O <sub>3</sub> (%)	Al <sub>2</sub> O <sub>3</sub> (%)	LOI (%)	TOTAL (%)
Q1-1	75.82	2.83	8.11	43.68	1.96	3.64	38.43	98.65
Q1-2	74.68	1.62	1.49	52.83	0.83	0.00	42.93	99.70
Q1-3	63.64	4.46	6.86	40.91	2.82	3.52	39.81	98.39
Q1-4	78.34	1.76	1.29	51.68	1.25	0.51	43.21	99.69
Q1-5	65.18	5.80	8.47	40.00	3.59	1.98	39.72	99.55
Q1-6	72.66	2.81	2.71	49.13	1.80	0.00	43.03	99.49
Q1-7	75.82	3.10	3.52	47.80	1.68	1.44	41.80	99.33
Q1-8	79.15	2.12	1.24	51.52	0.57	0.47	43.78	99.69
Q1-9	84.36	0.76	0.97	53.99	0.24	0.32	43.30	99.58
Q1-10	88.16	0.63	1.24	54.42	0.24	0.00	43.14	99.68
Q1-11	79.79	0.87	1.05	53.55	0.43	0.45	42.99	99.34
Q1-12	77.79	12.22	6.74	34.80	1.33	0.00	44.23	99.32
Q1-13	78.48	12.52	2.51	37.09	1.39	0.00	46.06	99.57
Q1-14	78.18	12.11	6.33	35.56	1.39	0.00	44.03	99.41
Q1-15	75.29	11.48	9.39	33.92	1.54	0.00	42.62	98.96
Q1-16	47.71	3.87	5.38	40.48	7.21	2.21	38.83	97.99
Q1-17	69.42	7.99	3.35	41.73	2.08	0.00	44.31	99.46
Q1-18	73.15	12.96	2.34	37.03	1.44	0.00	45.83	99.60
Q1-19	78.89	13.11	1.24	38.08	1.07	0.00	46.12	99.62
Q1-20	66.78	12.78	2.67	37.37	1.03	0.00	45.71	99.56
Q1-21	74.40	12.52	2.32	37.63	1.15	0.00	45.88	99.49
Q1-22	62.20	12.82	2.56	37.26	1.35	0.00	45.52	99.51

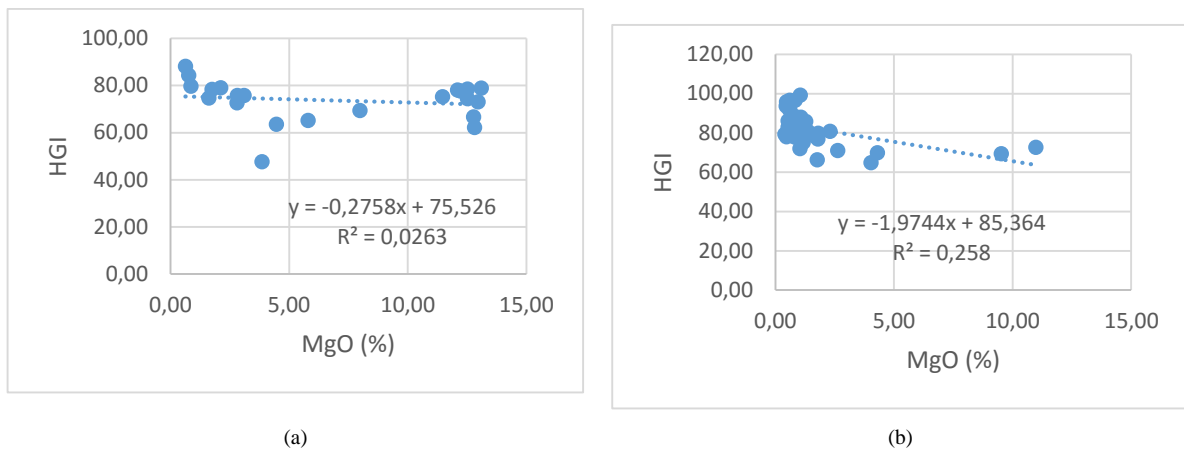
**Table 2.** Chemical composition data and HGI values of the limestone samples from Quarry-2.

SAMPLE	HGI	MgO (%)	SiO <sub>2</sub> (%)	CaO (%)	Fe <sub>2</sub> O <sub>3</sub> (%)	Al <sub>2</sub> O <sub>3</sub> (%)	LOI (%)	TOTAL (%)
Q2-1	96.69	0.83	4.71	54.24	0.73	1.97	36.71	99.20
Q2-2	95.94	0.46	4.23	54.47	0.68	1.67	37.93	99.44
Q2-3	78.18	0.46	5.23	53.75	1.23	1.86	36.85	99.38
Q2-4	86.75	0.80	11.67	46.59	1.11	2.73	36.19	99.09
Q2-5	66.30	1.76	30.42	27.95	3.39	12.86	19.30	95.68
Q2-6	83.07	0.54	3.81	54.36	0.61	1.33	38.90	99.55
Q2-7	72.14	1.03	14.30	46.58	2.20	6.65	27.18	97.95
Q2-8	86.30	0.53	2.83	54.60	0.85	1.26	39.50	99.57
Q2-9	79.49	0.40	4.70	53.95	0.97	1.45	38.04	99.50
Q2-10	76.91	1.79	6.57	49.87	1.42	2.51	36.56	98.72

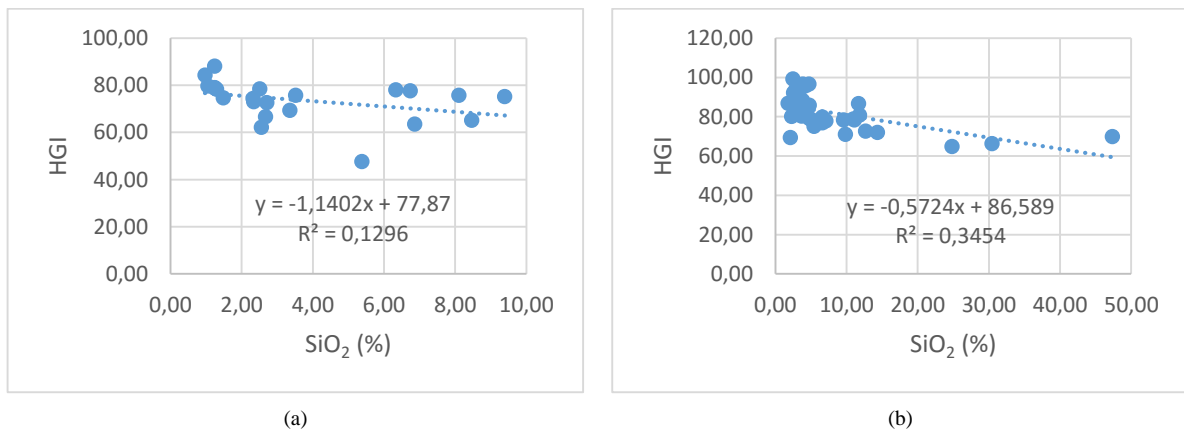
Table 2. Continues

Q2-11	82.78	0.94	4.35	53.79	0.78	1.48	38.05	99.38
Q2-12	72.78	10.98	12.64	35.21	1.98	5.43	32.84	99.07
Q2-13	70.01	4.29	47.33	9.94	2.02	22.28	10.06	95.93
Q2-14	69.55	9.51	2.07	42.57	1.98	1.22	42.40	99.75
Q2-15	80.34	1.41	3.65	53.48	0.65	1.26	39.22	99.67
Q2-16	92.04	0.59	3.28	54.43	0.34	0.59	40.54	99.77
Q2-17	77.91	0.81	7.10	52.63	1.30	3.54	33.46	98.84
Q2-18	93.84	0.45	2.85	54.78	0.28	0.56	40.83	99.75
Q2-19	78.46	0.90	11.09	47.69	1.88	3.71	33.52	98.79
Q2-20	64.84	4.03	24.81	30.77	3.46	8.04	25.95	97.06
Q2-21	78.97	0.90	4.87	53.12	0.83	1.73	37.86	99.31
Q2-22	78.46	1.67	9.55	50.04	1.56	4.03	31.66	98.50
Q2-23	92.33	0.60	2.55	55.37	0.62	0.98	39.56	99.68
Q2-24	96.65	0.60	3.84	54.59	0.72	1.46	38.27	99.47
Q2-25	88.39	0.84	3.81	54.14	0.70	1.64	38.26	99.40
Q2-26	82.59	1.25	4.13	53.01	0.82	1.73	38.21	99.15
Q2-27	88.13	1.07	3.31	54.95	0.61	1.44	38.07	99.46
Q2-28	79.87	1.80	6.59	50.02	1.05	2.41	37.08	98.95
Q2-29	99.23	1.04	2.46	55.25	0.56	1.01	39.26	99.58
Q2-30	75.19	1.16	5.43	52.66	1.51	2.22	36.18	99.15
Q2-31	86.97	1.02	1.78	55.94	0.55	0.64	39.51	99.44
Q2-32	85.88	1.28	4.73	53.34	0.71	1.87	37.39	99.30
Q2-33	78.85	1.66	10.95	47.21	1.65	3.86	33.23	98.55
Q2-34	80.23	0.99	2.27	54.04	0.64	0.77	40.91	99.63
Q2-35	71.17	2.62	9.81	45.89	1.50	2.48	36.64	98.94
Q2-36	80.81	2.30	11.81	45.30	1.59	4.28	33.01	98.30

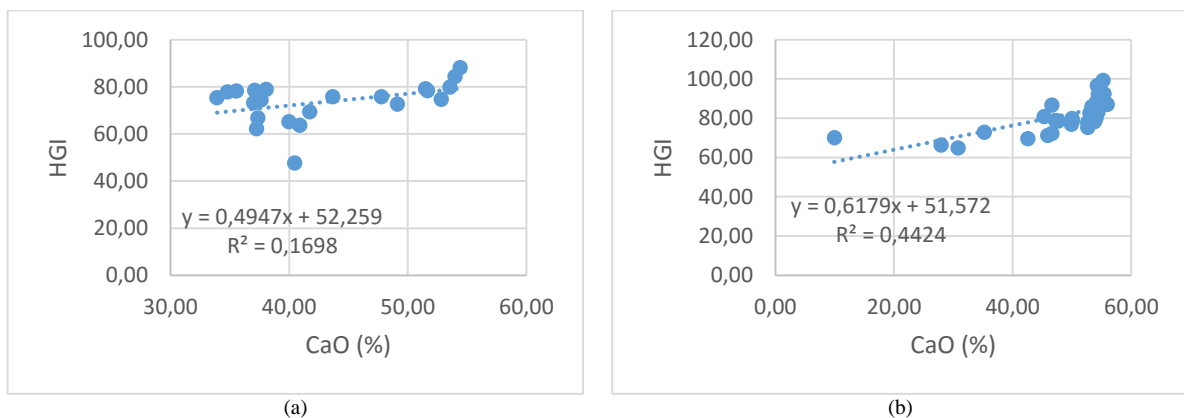
As it is presented in Table 1 and Table 2, chemical composition data for both quarries includes major oxide percentages (MgO, SiO<sub>2</sub>, CaO, Fe<sub>2</sub>O<sub>3</sub>, Al<sub>2</sub>O<sub>3</sub>) and LOI (%). Although chemical composition data for both quarry is available for other element oxides like K<sub>2</sub>O, TiO<sub>2</sub>, P<sub>2</sub>O<sub>5</sub>, Na<sub>2</sub>O, BaO, MnO and etc, tabulated data only includes the abovementioned major oxides, i.e. MgO, SiO<sub>2</sub>, CaO, Fe<sub>2</sub>O<sub>3</sub>, Al<sub>2</sub>O<sub>3</sub>. This is because of the fact that total percentages of these major oxides and LOI (%) is approximately 100 (See Table 1 and Table 2) for all samples and so the rest of the element oxides can be neglected to count in this case. In order to evaluate the change in grindability (HGI) with respect to the chemical composition data, graphical demonstrations of the relationships were provided between Figure 7 and Figure 12 [Figure 7(a)-12(a) for Quarry-1 and Figure 7(b)-12(b) for Quarry-2].



**Figure 7.** Relationship between HGI values and MgO (%) content of limestone samples (a) from Quarry-1, (b) from Quarry-2.

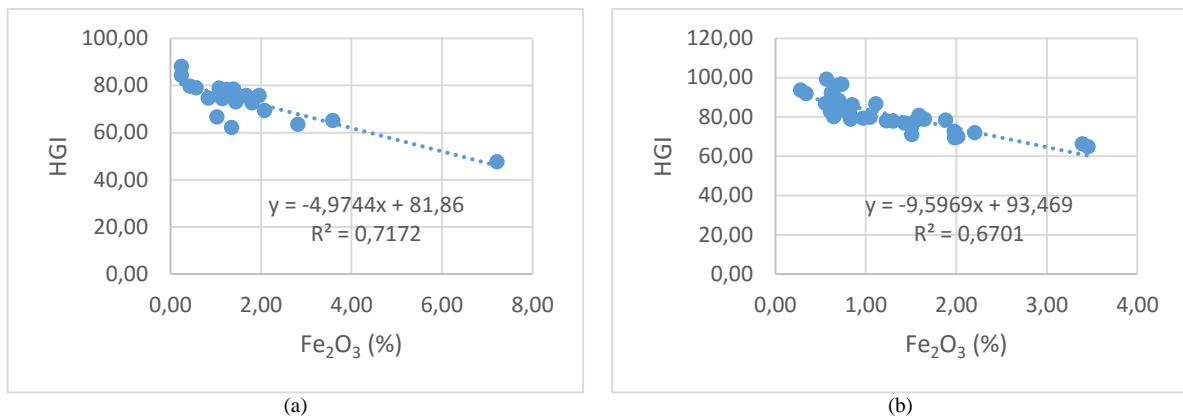


**Figure 8.** Relationship between HGI values and SiO<sub>2</sub> (%) content of limestone samples (a) from Quarry-1, (b) from Quarry-2.

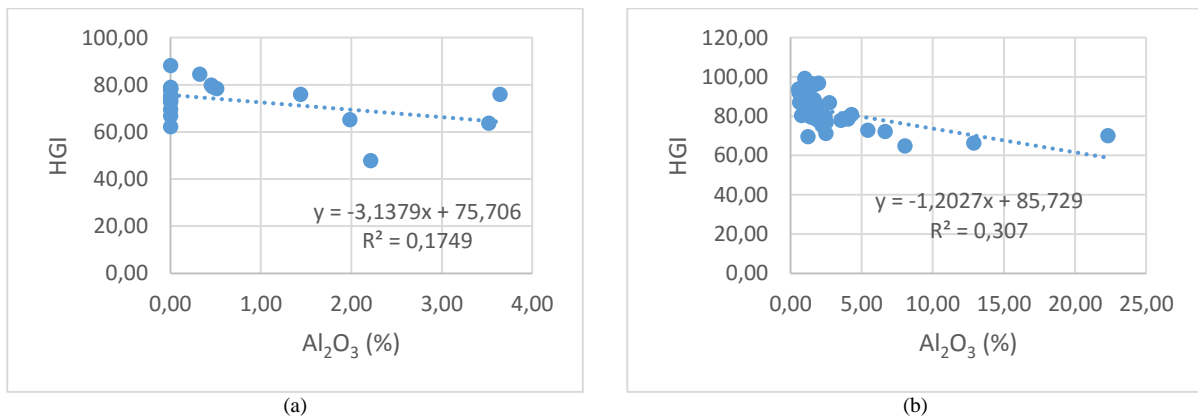


**Figure 9.** Relationship between HGI values and CaO (%) content of limestone samples (a) from Quarry-1, (b) from Quarry-2.

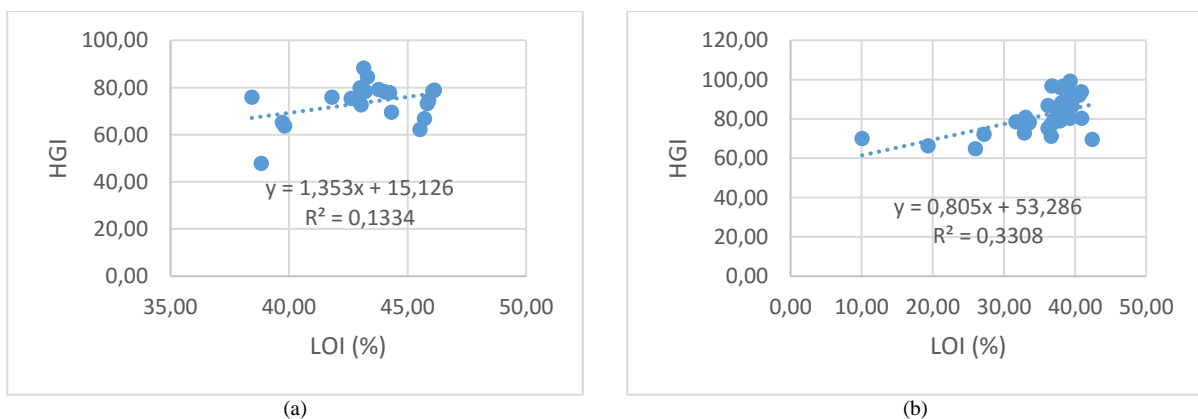




**Figure 10.** Relationship between HGI values and  $\text{Fe}_2\text{O}_3$  (%) content of limestone samples (a) from Quarry-1, (b) from Quarry-2.



**Figure 11.** Relationship between HGI values and  $\text{Al}_2\text{O}_3$  (%) content of limestone samples (a) from Quarry-1, (b) from Quarry-2.



**Figure 12.** Relationship between HGI values and LOI (%) content of limestone samples (a) from Quarry-1, (b) from Quarry-2.

Initial evaluations of the relationships (Figure 7 - Figure 12) are as following: i.  $\text{Fe}_2\text{O}_3$  content (%) is the most meaningful chemical composition parameter in terms of HGI for both quarries, ii. relationships obtained for the samples from Quarry-2 resulted in higher  $R^2$  values (coefficient of determination) except  $\text{Fe}_2\text{O}_3$ (%). In terms of these initial understandings abovementioned, one can claim the fact that any empirical equation to be proposed for the prediction of HGI should include  $\text{Fe}_2\text{O}_3$  (%) content.

Limestone grindability is a significant issue since limestone as raw material has a wide range of utilization, mostly requires powder form. In order to improve the efficiency of grinding circuits of limestones, iron content should be taken into consideration in the beginning. Graphical demonstrations of the relationship of HGI dependency on  $\text{Fe}_2\text{O}_3$  [Refer to Figure 10 (a) and (b)] simply includes the understanding as following: higher the

$Fe_2O_3$  results in lower the HGI value and so harder (more difficult) is the grinding operation and vice versa, respectively. This actually makes sense due to the fact that easier milling with any pre-treatment (microwave) would end up less wear of the mill, mill liner, and milling medium, summarized by Kumar et al. [32]. Here, in order to have higher grinding efficiency for limestone grinding, pre-treatment including magnetic separation could be employed.

In addition to the abovementioned interrelations, a new empirical equation for the prediction of HGI in terms of chemical composition data was proposed in the context of this study. In order to have less number of parameters contributing to HGI and to obtain the highest  $R^2$ , several attempts were carried out on a statistical software, i.e. XLSTAT. In this context, “Gamma Regression” in the body of “Log-Linear Regression” of that abovementioned software was taken into consideration. The model proposed (See (2) and (3)) has only 4 parameters employing and it has the corresponding  $R^2$  of 0.74.

$$\text{Prediction Method of HGI} = e^{(4.19+0.004xSiO_2+0.0056xCaO-0.084xFe_2O_3-0.0022xratio)} \quad (2)$$

Here, “ratio” is defined as (3):

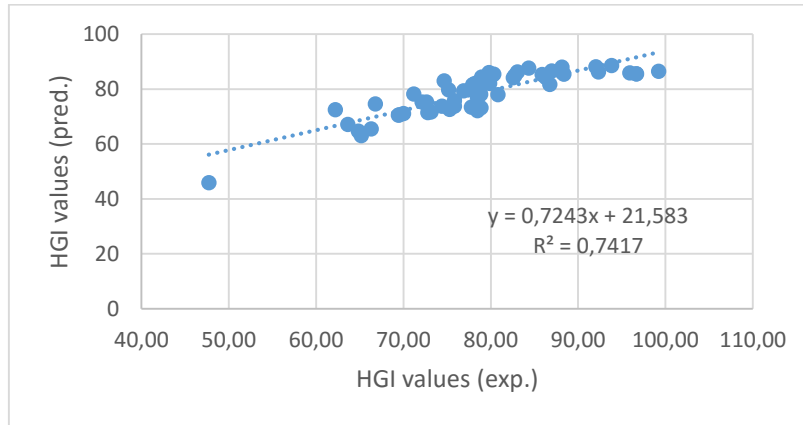
$$ratio = \frac{Fe_2O_3 + Al_2O_3 + MgO}{SiO_2} \quad (3)$$

Note that all chemical composition items, i.e.  $SiO_2$ ,  $CaO$ ,  $MgO$ ,  $Fe_2O_3$ ,  $Al_2O_3$  are in percentages. Trial error processing of linear and nonlinear regressions have only resulted as either too many number of parameters involving or less  $R^2$  obtained at the end. So this equation presented in (2) is regarded as the best equation in terms of easier evaluation and better prediction. In this context, graphical representation of the comparison between experimentally obtained HGI values and the predicted (Eqn.2 is employed) HGI values (See Table 3 and See Figure 13).

**Table 3.** Predicted (Eqn.2 is employed) and experimentally obtained HGI values.

Sample	Experimentally Obtained HGI values	Predicted HGI values	Sample	Experimentally Obtained HGI values	Predicted HGI values
Q1-1	75.82	73.71	Q2-1	96.69	85.57
Q1-2	74.68	82.95	Q2-2	95.94	85.94
Q1-3	63.64	67.11	Q2-3	78.18	82.06
Q1-4	78.34	79.34	Q2-4	86.75	81.74
Q1-5	65.18	63.03	Q2-5	66.30	65.50
Q1-6	72.66	75.25	Q2-6	83.07	86.24
Q1-7	75.82	75.69	Q2-7	72.14	75.32
Q1-8	79.15	83.95	Q2-8	86.30	84.27
Q1-9	84.36	87.63	Q2-9	79.49	83.79
Q1-10	88.16	88.03	Q2-10	76.91	79.36
Q1-11	79.79	86.00	Q2-11	82.78	84.93
Q1-12	77.79	73.41	Q2-12	72.78	71.42
Q1-13	78.48	72.15	Q2-13	70.01	71.12
Q1-14	78.18	73.17	Q2-14	69.55	70.59
Q1-15	75.29	72.59	Q2-15	80.34	85.43
Q1-16	47.71	45.92	Q2-16	92.04	88.10
Q1-17	69.42	70.51	Q2-17	77.91	81.63
Q1-18	73.15	71.67	Q2-18	93.84	88.57
Q1-19	78.89	73.19	Q2-19	78.46	76.89
Q1-20	66.78	74.62	Q2-20	64.84	64.71
Q1-21	74.40	73.76	Q2-21	78.97	84.41
Q1-22	62.20	72.48	Q2-22	78.46	79.52
			Q2-23	92.33	86.18
			Q2-24	96.65	85.55
			Q2-25	88.39	85.41
			Q2-26	82.59	84.15
			Q2-27	88.13	86.27
			Q2-28	79.87	82.02
			Q2-29	99.23	86.46
			Q2-30	75.19	79.69
			Q2-31	86.97	86.59
			Q2-32	85.88	85.33
			Q2-33	78.85	78.13
			Q2-34	80.23	85.23
			Q2-35	71.17	78.14
			Q2-36	80.81	77.96

Based on the data provided in Table 3, it can be easily noticed that experimentally obtained or predicted HGI values are higher for the samples collected from Quarry 2. This abovementioned difference between each quarry can be associated with the quarry location, quarry altitude difference, heterogeneous structure of the samples, alteration differences on each quarry, proximity to underground water supply & fault zone, meteorological differences, mineralogical and crystallographic differences and effects of freezing & thawing and etc.



**Figure 13.** Relationship between HGI values (exp.) and HGI values (pred.), “exp.” stands for experimentally obtained and “pred.” stands for predicted.

Referring to Figure 13, the correlation between experimentally obtained and predicted HGI values is significant, which can be interpreted as the achievement of the model proposed. In terms of linear regression models, proposed empirical equation (2) and (3) is the most significant, which is obtained with number of iterations. Prediction of HGI based on the chemical composition data has not been considered for limestones, which means it can be improved. Still, the proposed model represents a quite significant understanding of the parameters contributions and it can be employed as a guide for the initial estimates of HGI for limestones. Accordingly, one can refer to the coefficients of the each parameter of the proposed model (Eqn.2). In this context, it can be claimed that, increase in percentage of CaO results in an increase in HGI, which means easier grindability and increase in “ratio” results in a decrease in HGI, i.e. more difficult is the grinding. The contribution by SiO<sub>2</sub> and Fe<sub>2</sub>O<sub>3</sub> is kind of complicated in this case, since “ratio” does also depend on these parameters. Increase in Fe<sub>2</sub>O<sub>3</sub> content results in an increase in “ratio” and it therefore results in a duplicated decrease in HGI, which means harder grinding operation likely to be occurring. Increase in SiO<sub>2</sub> content results in an increase in HGI in some limits only. In addition to this abovementioned parametric evaluation, one can easily have initial estimates of HGI based on chemical composition data of limestones by avoiding the time consuming and laboring intense downside of the HGI method itself.

#### IV. CONCLUSION

In this study, a total of 58 limestone samples were studied in terms of their chemical composition and grindability value (HGI). Limestone samples were collected from two different quarry, i.e. Quarry-1 and Quarry-2. Analysis results of chemical composition were interrelated to the results of HGI tests for the corresponding samples. Further evaluations were made in this context with the referrals back and forth to the graphical representations of the relations (relationships between chemical composition data and HGI values). Major oxides (%) and LOI percentages for all samples from each quarry were tabulated. Tabulated data of chemical composition of the limestone samples was restricted to major oxides (SiO<sub>2</sub>, CaO, MgO, Fe<sub>2</sub>O<sub>3</sub>, Al<sub>2</sub>O<sub>3</sub>) since XRF analysis resulted as negligible for some oxides like Na<sub>2</sub>O, K<sub>2</sub>O and etc. In this context, graphical representations of the each chemical composition data (included) and the HGI values resulted with the understanding of the Fe<sub>2</sub>O<sub>3</sub> percentage significance in terms of HGI. Initial evaluations are summarized as following: i. Fe<sub>2</sub>O<sub>3</sub> content (%) is the most meaningful chemical composition parameter in terms of HGI for both quarries, ii. relationships obtained for the samples from Quarry-2 resulted in higher R<sup>2</sup> values (coefficient of determination) except Fe<sub>2</sub>O<sub>3</sub>(%). Latter in the context of this study, a new chemical grinding index (an empirical equation) was proposed to predict HGI. This model proposed includes 4 parameters (5 major oxides) and it has R<sup>2</sup> of 0.74 between HGI exp. and pred. values. The equation proposed includes a parameter stated as “ratio” which takes SiO<sub>2</sub>, MgO, Al<sub>2</sub>O<sub>3</sub> and Fe<sub>2</sub>O<sub>3</sub> into account. A correct evaluation in terms of the HGI values dependency on chemical composition data is as following: “i. increase in percentage of CaO results in an increase in HGI, which means easier grindability, ii. increase in Fe<sub>2</sub>O<sub>3</sub> content results in an increase in “ratio” and it therefore results in a duplicated decrease in HGI, which means harder grinding operation likely to be occurring, iii. increase in SiO<sub>2</sub> content results in an increase in HGI in some limits only”. By this method presented in the scope of this study, grindability index values for

limestones can be easily predicted for initial estimates without employing the time consuming and laboring intense HGI testing procedure.

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#### REFERENCES

- [1] Šiler, P., Kolarova, I., Bednarek, J., Jana, M., Musil, P., & Opravil, T. (2018). The possibilities of analysis of limestone chemical composition. *IOP Conf. Series: Materials Science and Engineering*.
- [2] Folk, R. L. (1959). Practical petrographical classification of limestones. *Amer. Ass. Petrol. Geol. Bull.*, 43(1), 1–38.
- [3] Folk, R. L. (1962). Spectral subdivision of limestone types. *Amer. Ass. Petrol., Geol. Mem.*, 1, 62–84.
- [4] Dunham, R. J. (1962). Classification of carbonate rocks according to depositional texture. *Mem. Amer. Ass. Petrol. Geol.*, 1, 108–121.
- [5] Mucsi, G., Rác Á., Mag G., Antal G., & Csóke B. (2019). Volume based closed-cycle Hardgrovegrindability method. *The Mining-Geology-Petroleum Engineering Bulletin*, 34(4), 9-17.
- [6] Todorovic, D., Trumic, M., Andric, L., Milosevic, V., & Trumic, M. (2017). A quick method for bond work index approximate value determination. *Physicochem. Probl. Miner. Process.*, 53(1), 321–332.
- [7] Bond, F.C., & Maxson, W. L. (1943). Standard grindability tests and calculations. *Trans. Soc. Min. Eng. AIME*, 153, 362–372.
- [8] ASTM D409 (1991). Standard Test Method for Grindability of Coal by the Hardgrove Machine Method.
- [9] Zeisel, H. G. (1953). Schriftenreihe der Zementindustrie. 14, *Verein Deutscher Zementwerke, Düsseldorf*, 51.
- [10] Hoşten, Ç. & Gülsün, M. (2004). Reactivity of limestones from different sources in Turkey. *Minerals Engineering*, 17(1), 97-99.
- [11] Yin, L. & Guo J. (2011). Study on Wet FGD Limestone Quality, *Third International Conference on Measuring Technology and Mechatronics Automation*, Shangshai, 605-608.
- [12] Deniz, V. (2014). Relationships between Bond's grindability (Gbg) and breakage parameters of grinding kinetic on limestone. *Powder Technology*, 139(3), 208-213.
- [13] Altun, N.E. (2014). Assessment of marble waste utilization as an alternative sorbent to limestone for SO<sub>2</sub> control. *Fuel Processing Technology*, 128, 461-470.
- [14] Ozkahraman, H.T. (2005). A meaningful expression between bond work index, grindability index and friability value. *Minerals Engineering*, 18(10), 1057-1059.
- [15] Seo, J.H., Baek, C. S., Cho, J.S., Ahn, Y.J., Ahn, J.W., & Cho, K.H. (2019). Evaluation and application of grinding index of domestic desulfurization limestone. *Journal of Energy Engineering*, 28(1), 1-9.
- [16] Tichanek, F. (2008). Contribution to determination of coal grindability using Hardgrove Method. *GeoSci. Eng.*, LIV (1).
- [17] Chelgani, S. C., Hower, J. C., Jorjani, E., Mesroghli, Sh., & Bagherieh, A. H. (2008). Prediction of coal grindability based on petrography, proximate and ultimate analysis using multiple regression and artificial neural network models. *Fuel Proc. Technol.*, 89, 13–20.
- [18] Sahoo, R.K. (2006). Review: An investigation of single particle breakage tests for coal handling system of the gladstone port, *Powder Technology*, 161(2), 158-167.
- [19] Ozbayoglu, G., Ozbayoglu, A. M., & Ozbayoglu, M. E. (2008). Estimation of Hard grove grindability index of Turkish coals by neural networks. *Int. J. Miner. Proc.*, 85, 93–100.
- [20] Sengupta, A.N. (2002). An assessment of grindability index of coal, *Fuel Processing Technology*, 76(1) 1-10.

- [21] Mendis, B.S.M., Jayathunga, T.H.G.S., Madurapperuma, H.H., Rohitha, L.P.S., Dharmarathna, P.G.R., & Hemalal, P.V.A. (2017). Determination of existing relationship among grindability, chemical composition and particle size of raw material mix at Aruwakkalu Limestone.
- [22] Kural, A., & Ozsoy, C. (2004). Identification and control of the raw material bending process in cement industry. *International Journal of Adaptation Control Signal Processing*, 18(5), 427-442.
- [23] Frigione, G., Zenone, F. & Esposito, M. V. (1983). The effect of chemical composition on Portland cement clinker grindability. *Cement and Concrete Research*, 13(4), 483-492.
- [24] Ürünveren, A., Altın, M., Kuvvetli, Y., Ural, O. B., & Ural, S. (2020). Prediction of Hard grove grindability index of Afsin-Elbistan (Turkey) Low-grade Coals based on proximate analysis and ash chemical composition by neural networks. *International Journal of Coal Preparation and Utilization*, 40(10), 701-711.
- [25] Bilen, M., Kızgut, S., Cuhadaroglu, A., Yılmaz, S., & Toroglu, İ. (2017). Coal grindability and breakage parameters. *International Journal of Coal Preparation and Utilization*, 37(5), 279-284.
- [26] Bilen, M., Kızgut, S., Yılmaz, S., Baris, K., & Cuhadaroglu, D. (2018). Grindability of coal changing with burial depth. *International Journal of Coal Preparation and Utilization*, 38(2), 75-87.
- [27] Yılmaz, S., & Bilen, M. (2016). Empirical Relationships of HGI in terms of proximate analysis of coal. In *XVIII International Coal Preparation Congress*, 953-957.
- [28] Cuhadaroglu, A. D., Kızgut, S., Yılmaz, S., & Zorer, Y. (2013). Characterization of the grinding behavior of binary mixtures of clinker and colemanite. *Particulate Science and Technology*, 31(6), 596-602.
- [29] Gouda, G. R. (1979). Effect of clinker composition on grindability. *Cement and Concrete Research*, 9(2), 209-218.
- [30] Gedik, İ., Duru, M., Pehlivan, Ş., & Timur, E., (2005). 1/50.000 ölçekli Türkiye Jeoloji haritaları, İstanbul-F22c Paftası, *MTA Raporu*, No:11, Ankara.
- [31] ASTM C1271-99. (2012). Standard Test Method for X-ray Spectrometric Analysis of Lime and Limestone.
- [32] Kumar, P., Sahoo, B.K., De, S., Kar, D.D., Chakraborty, S., & Meikap, B.C. (2010). Iron ore grind ability improvement by microwave pre-treatment. *Journal of Industrial and Engineering Chemistry*, 16(5), 805-812.