iJRASET
International Journal For Research in
Applied Science and Engineering Technology

INTERNATIONAL JOURNAL FOR RESEARCH

IN APPLIED SCIENCE & ENGINEERING TECHNOLOGY

Volume: 5 **Issue:** III **Month of publication:** March 2017

DOI: http://doi.org/10.22214/ijraset.2017.3134

www.ijraset.com Volume 5 Issue III, March 2017
IC Value: 45.98 ISSN: 2321-9653
International Journal for Research in Applied Science & Engineering Technology (IJRASET)

Desulfurization of High-Sulfur Lignites (Çan, Çanakkale-Turkey) by the Meyers Method

Seda Demirci [1], Zehra Altınçelep [2], Oktay Bayat [3]
[1,3]*Cukurova University, Vocational Schools of Aladag, Adana, Turkey*

Abstract**: *The uses of high-sulfur coals in burning processes are limited because of environmental pollution. In the world, the reserves of high quality coal are decreasing as well as Turkey where coal reserves are mainly lignite and almost 60% of the known reserves contain high sulfur coals therefore it is important to evaluate the low-quality lignites. Most of the produced lignite is utilized at the coal power stations where low-sulfur content is required. The polluting gases containing sulfur dioxide (SO^2) after burning of low quality coals are the main reason for air pollution, acid rain and some respiratory systems diseases. For instance, a typical un-controlled coal plant emits 14.000 tons of SO_2 per year. In this study, the Meyers (Fe^{3-} salts) method was applied to Çan lignite with high sulfur content. In the first stage, the leaching experiments using Fe^{3-} salts were carried out following Yates experimental design. The effects of particle size, temperature, time and $Fe(NO_3).9H_2O$ concentration were investigated for the removal of sulfur. Later, optimization tests were applied taking into consideration effective parameter. Optimum conditions were 0.5 M of Fe^{3-} salts concentration, 70 ^oC temperature, d100 = 75 µm particle size and 30 minutes of reaction time.***
Keywords**: *Lignite, Meyers method, Desulfurization, Coal yield, Reaction Time***

I. INTRODUCTION

Turkey has important lignite reserves (7.38 billion tons) and Çan (Çanakkale) lignite deposit is an important place in the north-western Turkey. Turkey lignite is not suitable nor as a fuel for home and neither for industry but very suitable for thermal power plants. The quality of Çan lignite reserves is not appropriate for the sulfur content. High amounts of sulfur cause problems, particularly environmental pollution. For this reason, it is extremely important to reduce sulfur from coal prior to application in power plants. Sulfur removal involves physical, biological and chemical methods. In the experimental test procedure, chemical method preferred due to more effective in removing both organic sulfur and inorganic sulfur than other methods. One of the most powerful practices for studying operate attitude is the factorial-design test with analysis of variance (ANOVA) from which information can be used for optimization aims. Statistical design is a significant of tool in understanding process behavior. The statistical-design general notion has been used successfully in leaching studies [1-6].

In this study, removing sulfur from Çan lignite was carried out by Meyers method. The effects of different parameters were investigated by 24 factorial design using the Yates method for leaching tests..

II. MATERIALS AND METHODS

A. Materials

The representative sample (approximately 100 kg) was taken from Çan – TKİ (Turkish Coal Enterprises) Çanakkale, Turkey. Proximate and ultimate analyses of the representative sample were defined according to standard methods. The total sulfur of the coal sample was measured using an ELTRA CS 580 model instrument. The air-dried sample was ground to -125 µm size then it was dried in an oven at 105 ^oC for 4 h to use for the experimental study. Typical properties of the lignite sample were given in Table 1.

Table I

Proximate And Ultimate Analysis Of Lignite Sample (Dry Basis) (Wt%)

	Particle size		
	| < 75 µm	**75-106 µm**	**106-125 µm**
Proximate analyses (dry basis)			
Ash	48.19	47.14	44.22
Volatile matter	33.02	33.01	33.07

International Journal for Research in Applied Science & Engineering Technology (IJRASET)

Fixed carbon	18,79	19.85	22.71
Ultimate analysis (dry basis)			
Carbon	30.45	31.86	33.45
Hydrogen	3.35	3.37	3.62
Nitrogen	2.12	2.15	2.19
Oxygen	15.89	15.48	16.52
Ash	48.19	47.14	44.22
Total sulfur	4.99	5.07	5.10

B. Methods

The impact of factors including a kind of dimensional fractions, temperature, time, and reagent concentration on the degree of desulfurization were researched using the ANOVA Yates test technique. Several tests were performed for estimation the effects of operating conditions. The Yates algorithm [7] for 2^4 experiments was used for statistical design and analysis of the experimental results. Combining the Yates technique with ANOVA (Tables 2 and 3) eases the determination of considerable differences. Experiments were done in a beaker with a magnetic and temperature was measured by a thermometer. In these experiments 5 g of the coal sample were soaked and mixed with distilled water to 50 mL volume. The glass cover was used to avoid evaporation in leaching experiments. The desulfurization of coal by Meyers process with acidic $Fe(NO_3)_3.9H_2O$ solution. The leaching test were performed at d_{100} = 75 µm, -106+75 µm, -125+106 µm particle sizes, concentration of $Fe(NO_3)_3.9H_2O$ (0.10, 0.25, 0.5 M), leaching time (30, 60, 90 min.) and temperature (30, 50, 70 ^oC).

Table II Leaching Parameter Levels

Description	Factor	Low level	Base level	High level
Particle size (µm)	A	<75	75-106	106-125
Temperature (^oC)	B	30	50	70
Time (min)	C	30	60	90
$Fe(NO_3).9H_2O$ (M)	D	0.10	0.25	0.50

Table III

Leaching experimental design of anova-yates test technique

Yates arrangements	A Particle size (µm)	B Temperature (^oC)	C Time (min.)	D $Fe(NO_3)_3.9H_2O$ (M)
1	< 75	30	30	0.10
a	75-106	30	30	0.10
b	< 75	70	30	0.10
ab	75-106	70	30	0.10
c	< 75	30	90	0.10
ac	75-106	30	90	0.10
bc	< 75	70	90	0.10
abc	75-106	70	90	0.10
d	< 75	30	30	0.50
ad	75-106	30	30	0.50
bd	< 75	70	30	0.50
abd	75-106	70	30	0.50
cd	< 75	30	90	0.50
acd	75-106	30	90	0.50
bcd	< 75	70	90	0.50
abcd	75-106	70	90	0.50
Medium 1	106-125	50	60	0.25
Medium2	106-125	50	60	0.25
Medium3	106-125	50	60	0.25

Volume 5 Issue III, March 2017
ISSN: 2321-9653

International Journal for Research in Applied Science & Engineering Technology (IJRASET)

The statistical design of leaching test is used when it can be desired to learn fundamental and impact of coaction used experimental parameters [8-13]. The leaching test was conducted to determine mostly the effect of the studied factor on the desulfurization process. Four parameters were taken into account in the experimental tests particle such as size (A), temperature (B), time (C) and concentration of $Fe(NO_3)_3.9H_2O$ (D) were set at their highest levels. Four parameters were determined and analysis of variance (ANOVA, Yates method) was employed using 2^n design (n = number of parameters, 4). The 2^k adverts to designs with k factors where each factor has just two levels. In the present state, it was $2^4 = 16$ where k = 4.

The three factors, considered important to the operation, were particle size (X_1), temperature (X_2), reaction time (X_3) and $Fe(NO_3)_3.9H_2O$ concentration (X_4). The particle size, temperature, reaction time and $Fe(NO_3)_3.9H_2O$ concentrations can all be changed continuously along their respective scales, from a low to a high setting.

Besides the leaching tests that composed the central point of the experimental design, assigned by arithmetic means of the high and low levels. The higher level term was described as '+' and lower level term as '−'. The four experiments were performed at the base level to estimate error and standard deviation. The stirring speed was kept constant during the leaching tests. The ANOVA experimental parameter levels and experimental design were given in Tables 4 and 5. High-level and low-level are typically terms used to classify, describe and point to specific goals of a systematic operation.

The all possible factor combinations mean that we can estimate all the main and interaction effects. There are four main effects, four-three factor interactions, and a four-factor interaction, all of which appear in the full model as follows:

The regression equation with interactive terms can be composed as:

$$Y = b_0 + b_1X_1 + b_2X_2 + b_3X_3 + b_4X_1X_2 + b_5X_1X_3 + b_6X_2X_3 + b_7X_1X_2X_3$$

The model created for removal of sulfur using the impact of changeable significance at the greater reliance interval is given bellows:

C. For Removal Sulfur

$$Y= 26.81 + 2.216258X_1 + 7.87625X_2 + 8.20625X_4 + 6.98X_2X_4$$

The correlation coefficient, R^2, is used to control the model's ability to predict the answer completely. The empirical models were discovered to estimate accurately the response variable, as define by the R^2 value, which was 0.9895. The residual analysis for removal of sulfur was given in Figure 2.

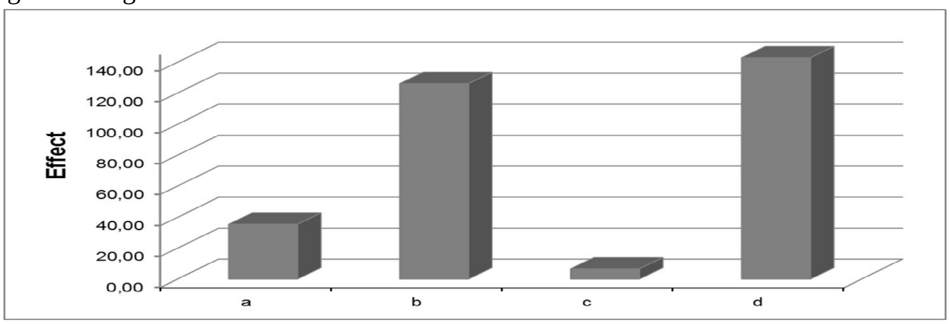


Fig.1 Main effects of variables active ratio. a = particle size (μm); b = temperature (°C); c = reaction time (min); d = $Fe(NO_3)_3.9H_2O$ concentration (M).

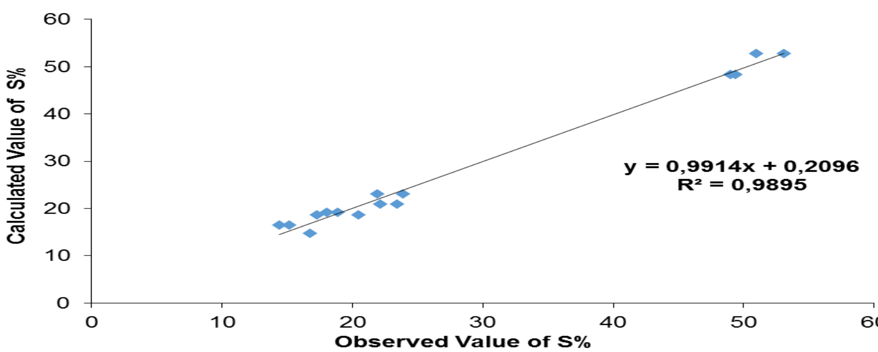


Fig. 2 Removal of sulfur values between observed and fitted responses for Meyers Method.

Volume 5 Issue III, March 2017
ISSN: 2321-9653
International Journal for Research in Applied Science & Engineering Technology (IJRASET)

Table IV

$Fe(no_3)_3.9h_2o$ leaching results for desulfurization

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
Yates arrangements	Experimental results (S% removal)	I	II	III	IV	MS	DF	F (Calc.)	F (Table) 1;16;0.05	Decision	X_1	X_2	X_3	X_4	Y (Calculations)	Coal yield
1	14.33	32.32	68.85	143.03	428.88	464.34	1	32.52014679	4.49	Active	-1	-1	-1	-1		89.24
a	17.99	36.53	74.18	285.85	35.46	78.59	1	5.503942398	4.49	Active	1	-1	-1	-1	19.19875	89.57
b	14.42	35.61	142.31	21.75	126.02	992.57	1	69.51449437	4.49	Active	-1	1	-1	-1	16.55875	88.44
ab	22.11	38.57	143.54	13.71	8.04	4.04	1	0.282949229	4.49	Inactive	1	1	-1	-1	20.99125	87.06
c	16.77	42.38	11.35	7.17	6.56	2.69	1	0.188366686	4.49	Inactive	-1	-1	1	-1	14.76625	87.02
ac	18.84	99.93	10.40	118.85	5.86	2.15	1	0.150311306	4.49	Inactive	1	-1	1	-1	19.19875	87.03
bc	15.12	41.12	3.45	10.29	2.50	0.39	1	0.027357502	4.49	Inactive	-1	1	1	-1	16.55875	84.94
abc	23.45	102.42	10.26	-2.25	-1.24	0.10	1	0.006730383	4.49	Inactive	1	1	1	-1	20.99125	85.30
d	20.48	3.66	4.21	5.33	142.82	1274.85	1	89.28417193	4.49	Active	-1	-1	-1	1	18.65875	85.33
ad	21.90	7.69	2.96	1.23	-8.04	4.04	1	0.282949229	4.49	Inactive	1	-1	-1	1	23.09125	83.44
bd	48.95	2.07	57.55	-0.95	111.68	779.53	1	54.59429073	4.49	Active	-1	1	-1	1	48.37125	91.63
abd	50.98	8.33	61.30	6.81	-12.54	9.83	1	0.688321746	4.49	Inactive	1	1	-1	1	52.80375	96.80
cd	17.28	1.42	4.03	-1.25	-4.10	1.05	1	0.073580737	4.49	Inactive	-1	-1	1	1	18.65875	85.27
acd	23.84	2.03	6.26	3.75	7.76	3.76	1	0.263584495	4.49	Inactive	1	-1	1	1	23.09125	86.28
bcd	49.36	6.56	0.61	2.23	5.00	1.56	1	0.109430007	4.49	Inactive	-1	1	1	1	48.37125	87.51
abcd	53.06	3.70	-2.86	-3.47	-5.70	2.03	1	0.142215237	4.49	Inactive	1	1	1	1	52.80375	78.82

a: particle size (µm), b: temperature (^oC), c: time (min), d: $Fe(NO_3)_3.9H_2O$ (M)

F (Calculation): (MS/DF)

Y: Calculated value of S% extra

III. OPTIMIZATION DESIGN

Optimization tests were carried out the steepest ascent method in which the set of value sequentially chose to correspond to the product of the factor. The objective function was estimated as much as optimum period was achieved that values on the way to destination were incremented and decreased [12].

The significance of each parameter on the efficiency of desulfurization was assessed using a, b, c and d exponents. ANOVA- Yates test technique showed that temperature and $Fe(NO_3)_3.9H_2O$ concentration was the least influential parameter when compared with other parameters for removal sulfur of the high – sulfur lignite. The optimization test terms were elected with respect to Table 4.

As a result of the optimization process, the removal of sulfur from the coal was further reduced from 5.10 %S to 2.10 %S. The removal of sulfur of the coal increased to 60.16% same as the yield of the coal increased to 94.36% (Table 5).

Table V

Optimization test conditions

Variable	$Fe(NO_3)_3.9H_2O$ (M)	Temperature (^oC)
Principal level Z_{jo}	0.50	70.00
Increment, delta Z_j	0.20	20.00
Coefficient, b_j	8.21	7.87
Delta Z_j * b_j	1.64	157.40
Normal steps	0.10	5.00

Volume 5 Issue III, March 2017
ISSN: 2321-9653
International Journal for Research in Applied Science & Engineering Technology (IJRASET)

Table VI
Results of optimization parameters

Experiments	Temperature (^oC)	$Fe(NO_3)_3.9H_2O$ (M)	Sulfur removal (%)	Coal yield (%)
1	60	0.3	26.25	87.08
2	60	0.4	42.10	89.71
3	60	0.6	45.53	87.91
4	60	0.7	46.65	88.60
5	65	0.3	42.01	92.50
6	65	0.4	41.63	88.62
7	65	0.6	47.86	88.60
8	65	0.7	51.27	87.86
9	75	0.3	36.88	94.43
10	75	0.4	39.21	95.75
11	75	0.6	51.87	84.28
12	75	0.7	50.91	85.52
13	80	0.3	44.50	91.67
14	80	0.4	42.50	95.80
15	80	0.6	51.42	92.10
16	80	0.7	60.16	94.36

IV. RESULTS AND DISCUSSION

The parameters investigated in the experimental work were: $Fe(NO_3)_3.9H_2O$ concentration (0.5, 0.25, 0.10 M), temperature (30, 50 and 70 ^oC), reaction time (30, 60 and 90 min.) and particle size. The ANOVA-Yates technique for 24 experimental test was used for statistical evaluation of the tests result. The most sulfur removal was acquired by reaction time and $Fe(NO_3)_3.9H_2O$ concentration. The effect of each variable for desulfurization was ("bd" of ANOVA factor) where d100 = 75 µm, 70 ^oC, 30 min. reaction time and 0.50 M $Fe(NO_3)_3.9H_2O$. The optimization step of experimental conditions was selected with respect to using the steepest ascend method. The highest sulfur removal value was obtained at 80 oC and 0.7 M $Fe(NO_3)_3.9H_2O$ concentration with 60.16% sulfur removal and 94.36% coal yield.

A. The Influence of Experimental Tests

To understand the effect of the particle size on sulfur removal from the coal, d_{100} = 75 µm, -106+75 µm and -125+106 µm particle sizes were used in the leaching experiments. The results showed that the removal of sulfur increased with decreasing particle size.
The reaction of $Fe(NO_3)_3.9H_2O$ concentration was investigated using 0.10, 0.25, 0.30, 0.4, 0.50, 0.6 and 0.6 M $Fe(NO_3)_3.9H_2O$ liquor. The results of ANOVA and optimization tests show that the removal sulfur increased with increasing chemical concentration. The effect of reaction time was altered from 30 to 90 min. It was observed that leaching time had no effect. The leaching temperature were studied in the range of 30 - 70 ^oC. The desulfurization increased as the temperature was increased and achieved its peak sulfur removal at 80 ^oC**.**

V. CONCLUSIONS

The experimental work showed that the removal of sulfur was 60.16% for the coal taken from Çan-TKİ, Çanakkale, Turkey at the optimum conditions of chemical leaching where 0.70 M of $Fe(NO_3)_3.9H_2O$, 80 ^oC, 30 min. of leaching time and d100 = 75 µm.

REFERENCES

[1] Cochran, W.G.; Cox, G.M. Experimental Designs, 2nd edition. Wiley, New York, 1957
[2] Daniel, C. Application of Statistics to Industrial Experimentation, Wiley, New York, 1976S. Zhang, C. Zhu, J. K. O. Sin, and P. K. T. Mok, "A novel ultrathin elevated channel low-temperature poly-Si TFT," IEEE Electron Device Lett., vol. 20, pp. 569–571, Nov. 1999.
[3] Box, G.E.P.; Hunter, W.G.; Hunter, J.S. Statistics for Experiments. Wiley, New York, 1978..

[4] Davis, O.L. The Design and Analysis of Industrial Experiments. 2nd edition. Longman Group, London, 1978. (2002) The IEEE website. [Online]. Available: http://www.ieee.org/

[5] Veglio, F.; Pagliarini, A.; Toro, L. Factorial experiments for the development of kaolin bioleaching process. International Journal of Mineral Processing. 1993, 39, 87-99. FLEXChip Signal Processor (MC68175/D), Motorola, 1996.

[6] Datta, P.; Ray, H.S.; Tripathy, A.K. Application of statistical design of experiments in process investigation, In: Quantitative Approaches in Process Metallurgy. 1995, Proceeding Volume, 243-253

[7] Yates, F. Design and Analysis of a Factorial Experiment. Imperial Bureau of Soil Science. 1976, 485.

[8] Çiftçi, H.; Alandağ, S. Leaching of Electric Arc Furnace Dust by Using Sodium Hydroxide. Proceedings of 14th International Mineral Processing Symposium, Kuşadası, Turkey, 2014, 513-520.

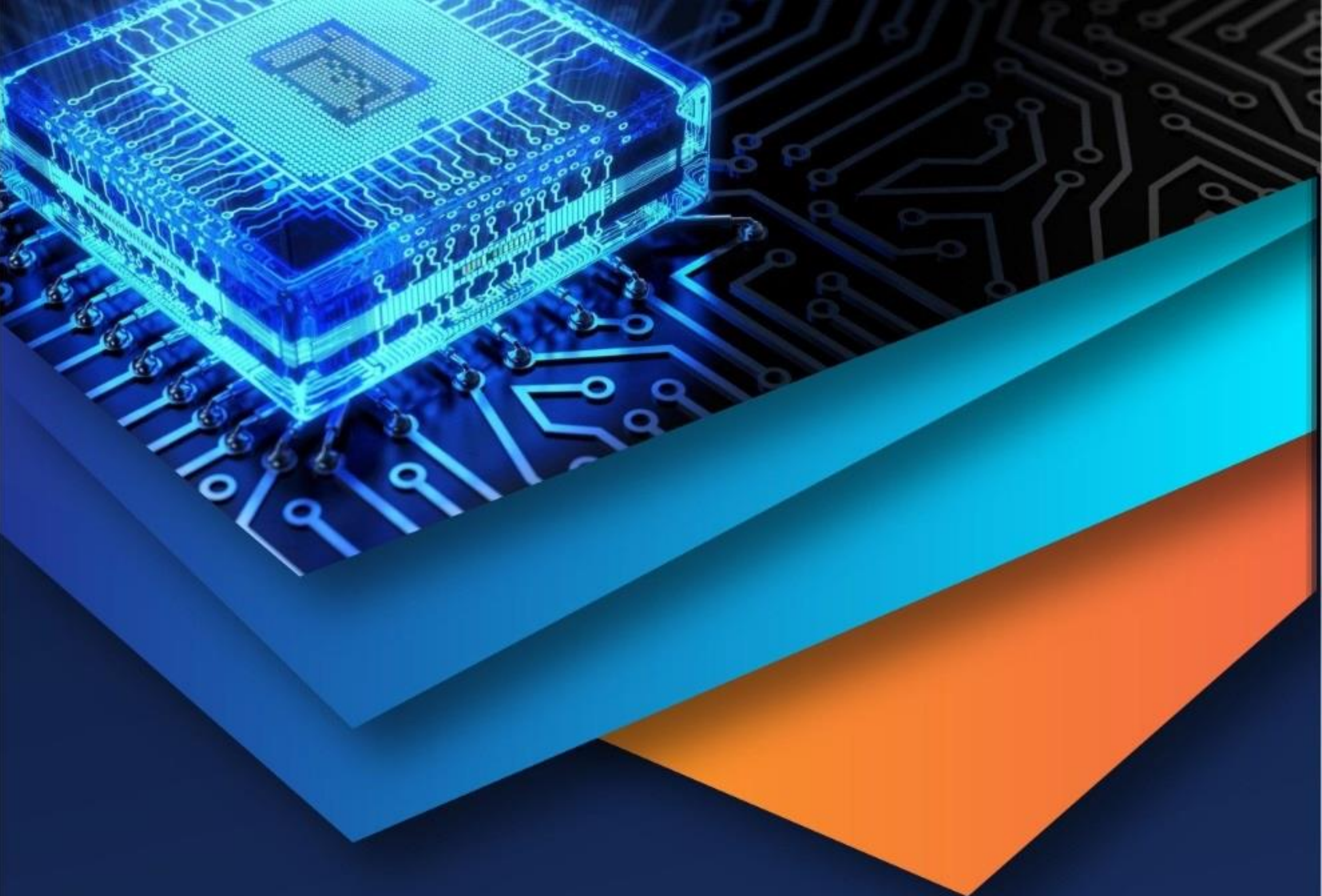
[9] Uçurum, M.; Arslan V.; Vapur H.; Bayat O.; Arslan, V. Recovery of Unburned Carbon from Lime Calcination Process Using Statistical Technique, Fuel Processing Technology. 2006, 87, 117-1121.

[10] Naik, K.P.; Reddy, R.S.P.; Misra, N.V. Interpretation of Interaction Effects and Optimization of Reagent Dosages for Fine Coal Flotation, Int. J. Miner. Process. 2005, 75, 83-90.

[11] Davies, L. The Statistical Design and Analysis of Chemical Experiments, The Royal Society of Chemistry. Cambridge, UK, 1993, 180.

[12] Naik, P.K.; Reddy, P.S.R.; Misra, V.N. Optimization of coal flotation using statistical technique, Fuel Process. Technol. 2004, 85, 1473–1485

[13] Montgomery, D.C. Design and Analysis of Experiments, 3rd ed., Wiley, Singapore, 1991, 615



10.22214/IJRASET

INDEX COPERNICUS

45.98

IMPACT FACTOR: 7.129

IMPACT FACTOR: 7.429

TOGETHER WE REACH THE GOAL

INTERNATIONAL JOURNAL FOR RESEARCH

IN APPLIED SCIENCE & ENGINEERING TECHNOLOGY