Optimisation of process parameters for coal flotation using statistical technique

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Abstract
The ash content present in coal plays an important role in determining the quality or grade of the coal for its utilization in different industries. The maximum ash content allowed for steel grade I is 15%, while steel grade II requires ash content ranging from 15% to 18%. A sample of coking coal analysing 26.32% ash was subjected to froth flotation to reduce its ash content to below 18%. Optimization of flotation process parameters such as collector, frother dosages and airflow rate, was carried out using factorial design of experiments.

It was observed that the interaction of collector and frother dosages had the most significant impact on achieving the desired ash rejection, with collector dosage also playing an important role. Optimum process parameters identified are collector dosage of 0.0348 kg/t, 0.005 kg/t frother dosage and 2 lpm airflow rate, wherein the ash content of the sample was reduced to 14.58% from 26.32%.

Keywords: Coking coal, froth flotation, collector, frother, airflow rate, design of experiment.

Introduction

The grading of coking coal is determined by its ash content. As per the notification from the Ministry of Coal, steel grade I coal requires ash content not exceeding 15%, whereas ash content in steel grade II coal ranges between 15-18%. Coal fines generally contain 20-30% ash. Hence, the processing of coal is becoming more important. The mineral content associated with coking coal is typically made up of hydrophilic minerals. These minerals are primarily clays, including kaolinite and montmorillonite, as well as quartz, pyrite and carbonate minerals.

To enhance the quality of the coal for use in combustion or industrial processes, impurities are often removed using froth flotation when liberation occurs in fine size. The performance of a flotation unit could be influenced by various factors including the quantity and type of chemicals added[2,4,8,19], the size of the bubbles[6], the configuration of the stator and rotor[5] and the residence time[15]. Leja-Schulman's theory states that frothers exhibit a preference for accumulation at the interfaces of air and water. During particle-to-bubble collision and attachment, they interact with collector molecules that are adsorbed onto solid particles[9]. Air bubbles capture hydrophobic coal particles and leave hydrophilic ash minerals behind, streamlining separation and optimizing efficiency.

The mineral processing techniques have shown the advantages of using statistical design of experiments over the traditional approach of testing one variable at a time[1,3,12,14]. One commonly used statistical technique is the factorial design test, which examines the main effects as well as interactions of multiple factors[13]. The objective of this study is to optimize the dosage of collector, frother and air flow rate to achieve a maximum yield while reducing ash content below 18% using statistical techniques.

Material and Methods

Materials: A coking coal sample received from Jharkhand, India, was subjected to proximate analysis which revealed an ash content of 26.32% and a fixed carbon content of 47.83% (Table 1).

Table 1
Results of proximate analysis

Constituents	Percentage, %
Moisture	0.91
Volatile matter	24.94
Ash	26.32
Fixed carbon	47.83

The coal sample was subjected to wet sieve analysis using 500µm, 300µm and 106µm sieves. Table 2 provides the size and size-wise ash content of the sample. The ash content was higher (above 26%) across all size ranges except +500 µm.

Table 2
Size and size-wise ash content.

Size, µm	Ash Content, %		
	Weight, %	Ash, %	Ash Distribution, %
+500	0.24	21.03	0.19
-500+300	30.82	28.55	32.06
-300+106	36.90	26.50	35.64
-106	32.04	27.51	32.12

The ash content was higher (above 26%) across all size ranges except +500 µm.

The powdered feed coal sample was subjected to X-ray diffraction studies for mineralogical phase analysis. The crystallographic data was obtained from XRD patterns of Bruker D8 Advance diffractometer for the 2θ angles between 10 and 80° with CuKα radiation (λ = 1.5418 Å). The XRD analysis of the coal sample (Figure 2a) shows quartz and kaolinite as the predominant gangue mineral phases followed by pyrite and muscovite.

Customized collector and frother, namely, collector ‘C’ and frother ‘F’, were employed in the flotation process. Collector ‘C’ and frother ‘F’ are synthetic, organic and proprietary reagents. The FTIR absorbance spectra of collector ‘C’ (Figure 1a) and frother ‘F’ (Figure 1b) were recorded, ranging from 4000 – 500 cm^{-1}, using Perkin Elmer-FTIR. The peaks at 2921, 2854, 1461, 1371 and 720 cm^{-1} indicate the presence of -C-C-, CH_3 and CH_2 functional groups (Figure 1a). The presence of the -C=O functional group is evidenced by a strong absorbance peak at 1741 cm^{-1} and the peak at 1168 cm^{-1} confirms the presence of the ether linkage (-C-O-C-)[10].

The presence of -C-O- ether and ester functional groups is confirmed by 1018 cm^{-1} peak[17]. The strong absorbance band at 3336 cm^{-1} (Figure 1b) denotes the NH stretching vibration[17] and the peaks at 2956, 2925, 2872, 1459, 1377 and 763 cm^{-1} indicate the presence of –C-C-, CH_3 and CH_2 functional groups[10]. The strong band at 1036 cm^{-1} is due to the presence of –C-O- ether and ester functional groups[17]. The functional groups present in collector ‘C’ and frother ‘F’ as revealed by the FTIR study such as oxygen-containing groups and alkane groups (-CH, CH_3 and CH_2), resemble that of traditionally used reagents/surfactants in coal flotation indicating promising role as collector and frother respectively.

Methods: A 2^3 factorial design of experiment was implemented (Table 3) to assess the impact of collector, frother dosage and airflow rate on flotation performance responses such as final concentrate yield, ash and fixed carbon content of the concentrate. Based on the factorial design of experiment, 8 sets of tests were designed using MINITAB software with $N=2^n$ equation, where N is the number of tests and n is the number of variables. A 2-liter Denver cell was used for bench-scale flotation tests at 10% pulp density and a natural pH of 7, where the pulp was agitated for 2 minutes followed by conditioning with reagents for 2 minutes. The concentrates and tailings were collected, dried and then analysed for ash.

Results and Discussion

A table showing the yield, ash and fixed carbon content of the clean coal (concentrate) for 3 variables is shown in table 4. It could be observed from the results that the clean coal of steel grades I and II was obtained from feed coking coal with 26.32% ash. Clean coal of steel grade II with ash content of 16-18% was obtained with yield ranging from 67-74%, whereas a clean coal of steel grade I was obtained with 14.58% ash and 57.35% yield. The XRD analysis of the coal sample, concentrate and tailings (Figure 2) reveals that the intensity of the ash-forming gangue minerals in the concentrate is less as compared to that of the head coal sample and tailings which indicate the enrichment of the clean coal in concentrate and rejection of gangue minerals into the tailings.

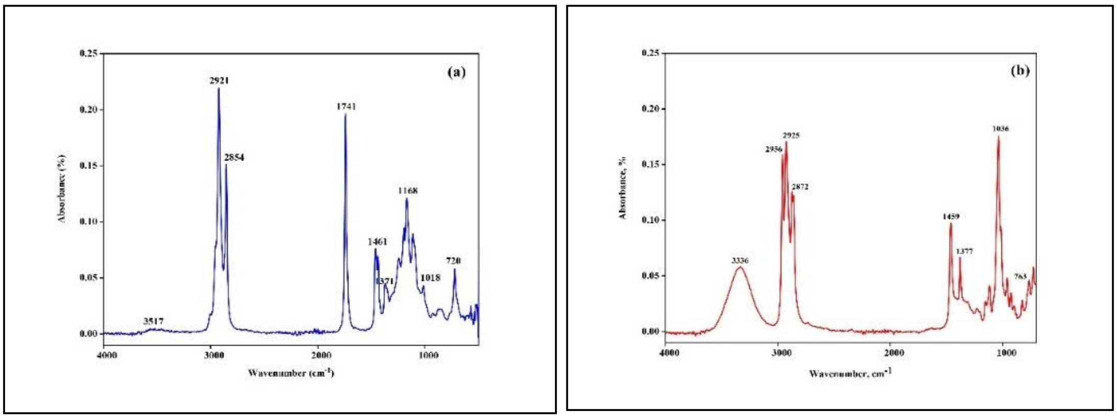

Fig. 1: FTIR spectrum of (a) collector ‘C’ and (b) frother ‘F’

Table 3
Ranges of input variables and their levels in factorial design

Input	Codes	Levels and Range	
		1	2
Collector dosage, kg/t	C	0.0348	0.0522
Frother dosage, kg/t	F	0.0033	0.0050
Air flow rate, lpm	A	1	2

Table 4
Factorial design of experiment matrix (L8).

Run No.	Collector Dosage, kg/t	Frother Dosage, kg/t	Airflow rate, lpm	Yield, %	Ash, %	Fixed Carbon, %
1	0.0348	0.0033	1	73.6	17.83	51.92
2	0.0522	0.0033	1	80.4	19.03	54.34
3	0.0348	0.005	1	67.47	16.22	56.04
4	0.0522	0.005	1	82.76	19.66	54.14
5	0.0348	0.0033	2	73.16	17.33	55.36
6	0.0522	0.0033	2	83.84	19.84	53.19
7	0.0348	0.005	2	57.35	14.58	57.29
8	0.0522	0.005	2	86.87	20.55	53.27

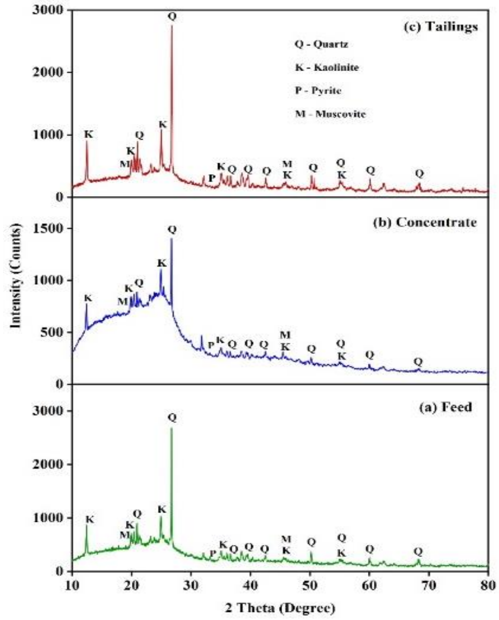

Fig. 2: XRD patterns of a) feed coal sample, b) concentrate) and c) tailings

Optimization using Analysis of Variance (ANOVA): Analysis of variance was conducted to assess the significance of the effect of factors and interactions among factors. An effect is deemed significant if p-value is less than the significance level (α). To evaluate the significance of effects on yield, ash and fixed carbon content in the final concentrate, an ANOVA analysis was conducted at a significance level (α) of 0.10. The ANOVA tables for yield, ash and fixed carbon content are shown in table 5(a-c).

In the analysis of variance table (Table 5a-c), the p-values for frother dosage and airflow rate, as well as the interactions, are more than 0.1, which indicates that the effects are statistically not significant. The p-values for collector dosage for ash% (Table 5b) is less than 0.1 indicating that the main effect of collector quantity for ash content is statistically significant at the significance level of 0.1.

Comparing the F-values given in ANOVA tables, the degree of significance of input variables is in the following order:

Yield, % and Ash, %: Collector dosage > Frother dosage > Airflow rate

Fixed carbon, %: Frother dosage > Collector dosage > Airflow rate

Collector dosage is the most significant parameter in both cases of % yield and ash content whereas frother dosage is found to be more significant than collector dosage in the case of fixed carbon% of the clean coal concentrate.

Regression model equations: The regression equation has been used to describe the relationship between the responses such as yield, ash and fixed carbon content and the input parameters such as collector dosage, frother dosage and airflow rate.

Model equations were derived using regression analysis as follows:

$$\text{Yield, \%} = 148.8 - 1803C - 18554F - 12.4A + 461968C*F + 520C*A - 2650F*A \quad (1)$$

$$\text{Ash, \%} = 34.59 - 377C - 4168F - 3.62A + 96349C*F + 110.3C*A - 312F*A \quad (2)$$

$$\text{Fixed Carbon, \%} = 18.5 + 641C + 6251F + 11.39A - 104293C*F - 192.8C*A - 562F*A \quad (3)$$

where C is Collector Dosage, kg/t, F is Frother Dosage, kg/t and A is Airflow rate, lpm.

Analysis of the above equations reveals that the term C*F holds the largest absolute value, indicating the highest effect of the interaction of factor C (collector quantity) and F (frother quantity) on yield, ash and fixed carbon content of the final concentrate. The equations (1) and (2) show that increasing both collector and frother dosage at the same time leads to a rise in yield and ash percentage of the concentrate. This is indicated by the positive coefficients of C*F. On the other hand, equation (3) suggests the opposite, as the negative coefficient of C*F shows that an increase in both collector and frother dosage leads to a decrease in the fixed carbon percentage of the concentrate.

Table 5(a)
ANOVA table for yield%.

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Model	6	664.886	110.814	8.28	0.260
Linear	3	520.376	173.459	12.95	0.201
Collector Dosage, kg/t	1	485.006	485.006	36.22	0.105
Frother Dosage, kg/t	1	34.238	34.238	2.56	0.356
Airflow rate, lpm	1	1.133	1.133	0.08	0.820
2-Way Interactions	3	144.510	48.170	3.60	0.365
Collector Dosage, kg/t * Frother Dosage, kg/t	1	93.366	93.366	6.97	0.230
Collector Dosage, kg/t * Airflow rate, lpm	1	40.997	40.997	3.06	0.331
Frother Dosage, kg/t * Airflow rate, lpm	1	10.148	10.148	0.76	0.544
Error	1	13.390	13.390		
Total	7	678.276			

Table 5(b)
ANOVA table for ash%.

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Model	6	28.7260	4.7877	25.73	0.150
Linear	3	22.6811	7.5604	40.64	0.115
Collector Dosage, kg/t	1	21.5168	21.5168	115.65	0.059
Frother Dosage, kg/t	1	1.1400	1.1400	6.13	0.244
Airflow rate, lpm	1	0.0242	0.0242	0.13	0.780
2-Way Interactions	3	6.0449	2.0150	10.83	0.219
Collector Dosage, kg/t*Frother Dosage, kg/t	1	4.0612	4.0612	21.83	0.134
Collector Dosage, kg/t*Airflow rate, lpm	1	1.8432	1.8432	9.91	0.196
Frother Dosage, kg/t*Airflow rate, lpm	1	0.1404	0.1404	0.75	0.545
Error	1	0.1860	0.1860		
Total	7	28.9120			

Table 5(c)
ANOVA table for fixed carbon%.

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Model	6	20.1480	3.3580	4.40	0.349
Linear	3	9.3053	3.1018	4.07	0.346
Collector Dosage, kg/t	1	4.0186	4.0186	5.27	0.262
Frother Dosage, kg/t	1	4.3956	4.3956	5.76	0.251
Airflow rate, lpm	1	0.8911	0.8911	1.17	0.475
2-Way Interactions	3	10.8426	3.6142	4.74	0.323
Collector Dosage, kg/t*Frother Dosage, kg/t	1	4.7586	4.7586	6.24	0.242
Collector Dosage, kg/t*Airflow rate, lpm	1	5.6280	5.6280	7.38	0.225
Frother Dosage, kg/t*Airflow rate, lpm	1	0.4560	0.4560	0.60	0.581
Error	1	0.7626	0.7626		
Total	7	20.9106			

The negative coefficients of C, F and A (Eq. 1 and 2) indicate that increase in airflow rate, collector or frother quantity leads to decrease in yield and ash% of the concentrate while the positive coefficient of C, F and A (Eq. 3) suggest that increase in airflow rate, collector or frother quantity leads to increase in the fixed carbon percentage of the concentrate. The predicted values for yield, ash and fixed carbon (FC) were obtained using eq. (1-3) and the plots for actual vs. predicted responses are given in figures 3a-c.

The R^2 values for concentrate yield, ash and fixed carbon content were found to be 98.03, 99.36 and 96.35% respectively. This shows that the model explains 98.03%, 99.36% and 96.35% of the variance in yield, ash and fixed carbon content respectively which indicates the model fits the data well. The adjusted determination coefficients (Adj. R^2 = 86.18, 95.50 and 74.47% for yield, ash and fixed carbon content respectively) were also satisfactory and confirmed the significance of the models.

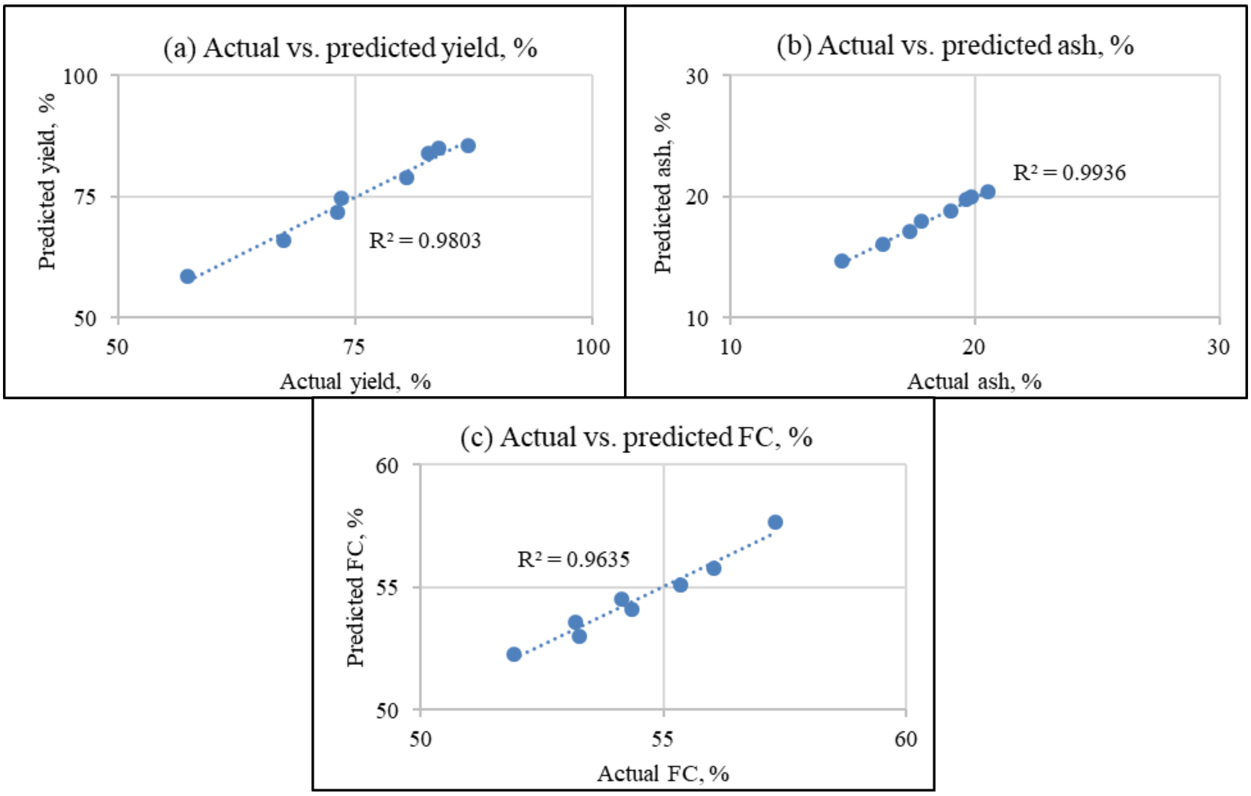

Fig. 3: (a) Actual vs. predicted yield% (b) Actual vs. predicted ash% (c) Actual vs. predicted fixed carbon% (FC)

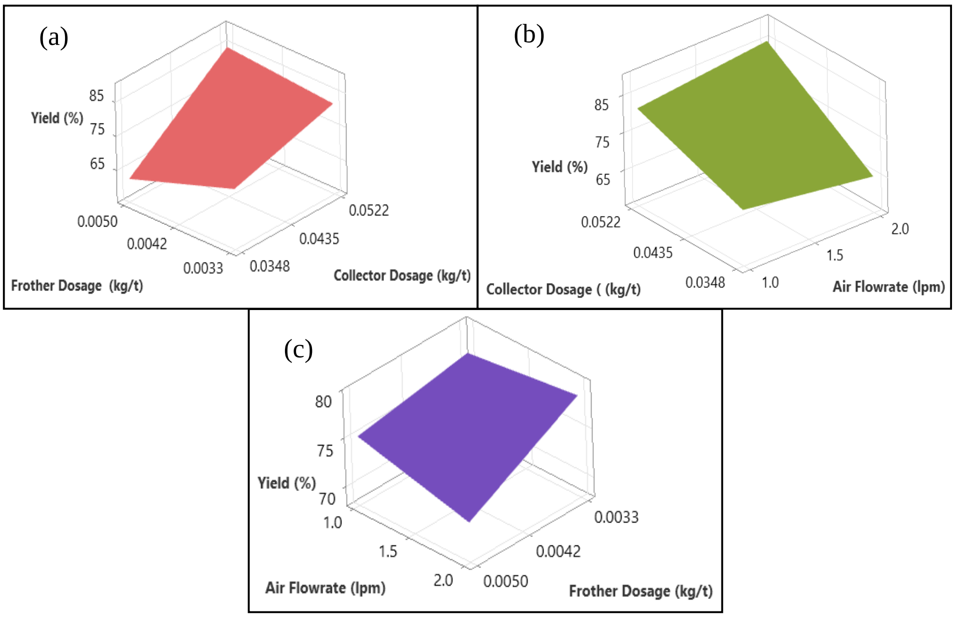

Fig. 4: Surface plot of yield% vs. (a) collector dosage, kg/t and frother dosage, kg/t; (b) airflow rate, lpm and collector dosage, kg/t; (c) airflow rate, lpm and frother dosage, kg/t

Response surface plots for yield, ash and fixed carbon content of final concentrate: Regression equations can be graphically represented using 3D surface plots. They help to understand the association between the output and input variables and the interactions between them to determine the optimal conditions[7,11,16,18]. From figure 4a, it is observed that when collector dosage is higher and frother dosage is lower, the yield% of concentrate is also minimum, but it reaches maximum when both the reagent dosages are higher. When collector dosage is low and airflow rate is high, the yield is minimum, however the response is maximum when both the parameters are higher (Figure 4b). When both frother dosage and airflow rate are high, the yield is minimum, however it reaches maximum when frother dosage is low and airflow rate is higher (Figure 4c).

When collector quantity is high and frother quantity is low, the ash% of concentrate is minimum, but it reaches maximum when both the reagent quantities are higher (Figure 5a). When collector quantity is low and airflow rate is high, the ash is minimum, however the response is maximum when both the parameters are higher (Figure 5b). When both frother dosage and airflow rate are high, the concentrate ash% is minimum, however it reaches maximum when frother quantity is low and airflow rate is higher (Figure 5c).

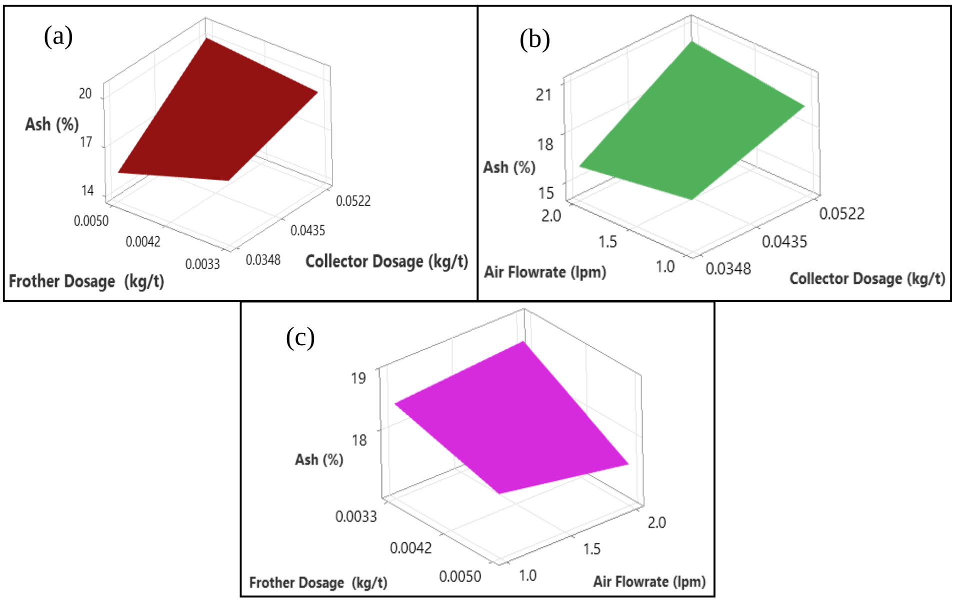

Fig. 5: Surface plot of ash% vs. (a) collector dosage, kg/t and frother dosage, kg/t; (b) airflow rate, lpm and collector dosage, kg/t; (c) airflow rate, lpm and frother dosage, kg/t

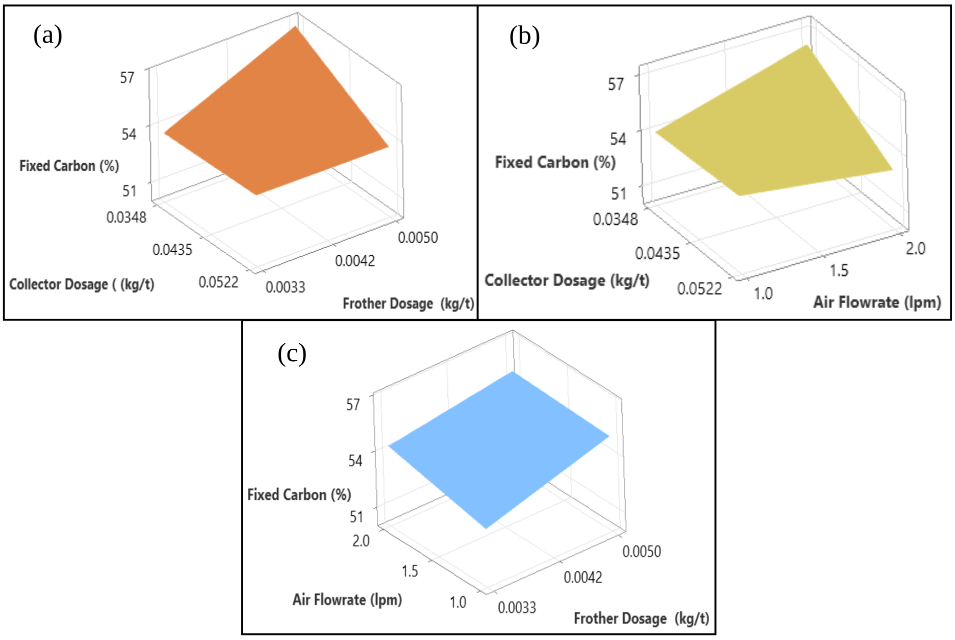

Fig. 6: Surface plot of fixed carbon% vs (a) collector dosage, kg/t and frother dosage, kg/t; (b) airflow rate, lpm and collector dosage, kg/t; (c) airflow rate, lpm and frother dosage, kg/t

When the collector quantity is high and the frother quantity is low, the fixed carbon content in the final concentrate is minimum, but it reaches maximum when the collector quantity is low and the frother quantity is high (Figure 6a). When both the collector quantity and airflow rate are high, the fixed carbon content in concentrate is minimum. However, the response is maximum when collector quantity is low and airflow rate is high (Figure 6b). When both frother quantity and airflow rate are low, the concentrate fixed carbon content is minimum; however, it reaches a maximum when both the parameters are higher (Figure 6c).

At higher collector and frother quantity, the increase in both yield and ash may be contributed to increased recovery of both coal and gangue into the concentrate[3]. The increase in yield and ash content in concentrate with the increase in airflow rate may be due to increased bubble surface area flux resulting in increased collection rate of coal particles along with interlocked gangue into the froth. Flotation is a complex process that involves solid, liquid and gaseous phases. Different phases interact with one another and also with the molecules of surfactants. Hence, the output responses are a function of the individual and synergistic effects of the contributing factors. From this study, it is evident that the interaction of the collector and frother dosages affects majorly the output parameters.

Conclusion

A low-rank coking coal analyzing 26.32% ash and 47.83% fixed carbon, comprising of quartz as well as kaolinite as major mineral matters was subjected to flotation to reduce the ash content in the range of 15-18%. Laboratory-synthesized reagents (collector 'C' and frother 'F') were used for flotation experiments. The statistical analysis of experimental results indicated that the interaction of collector and frother dosages had the most substantial impact in attaining the desired result, with the dosage of the collector being the next most influential factor. Coking coal concentrates of steel grade-II with ash content of 16-18% were obtained with the yield ranging from 67-74% whereas a concentrate of steel grade-I was obtained with 14.58% ash and 57.35% yield from the coking coal analysing 26.32% ash.

The optimum process conditions were identified as 0.0348 kg/t collector 'C', 0.005 kg/t frother 'F' and 2 lpm airflow rate. Thus, the tailor-made laboratory synthesized reagents could reduce the ash content of the coal to below 15% thereby producing a clean coal suitable for steel grade-I and II.

References

1. Aslan N. and Fidan R., Optimization of Pb flotation using statistical technique and quadratic programming, *Separation and Purification Technology*, **62(1)**, 160-165 **(2008)**

2. Bonner C.M. and Aplan F.F., The influence of reagent dosage on the floatability of pyrite during coal flotation, *Separation Science and Technology*, **28(1-3)**, 747-764 **(1993)**

3. Cilek E.C. and Yılmazer B.Z., Effects of hydrodynamic parameters on entrainment and flotation performance, *Minerals Engineering*, **16(8)**, 745-756 **(2003)**

4. Erol M., Colduroglu C. and Aktas Z., The effect of reagents and reagent mixtures on froth flotation of coal fines, *International Journal of Mineral Processing*, **71(1-4)**, 131-145 **(2003)**

5. Forrester S.E., Rielly C.D. and Carpenter K.J., Gas-inducing impeller design and performance characteristics, *Chemical Engineering Science*, **53(4)**, 603-615 **(1998)**

6. Gorain B.K., Franzidis J.P. and Manlapig E.V., Studies on impeller type, impeller speed and air flow rate in an industrial scale flotation cell—Part 1: Effect on bubble size distribution, *Minerals Engineering*, **8(6)**, 615-635 **(1995)**

7. Haider M.A. and Pakshirajan K., Screening and optimization of media constituents for enhancing lipolytic activity by a soil microorganism using statistically designed experiments, *Applied Biochemistry and Biotechnology*, **141**, 377-390 **(2007)**

8. Jia R., Harris G.H. and Fuerstenau D.W., An improved class of universal collectors for the flotation of oxidized and/or low-rank coal, *International Journal of Mineral Processing*, **58(1-4)**, 99-118 **(2000)**

9. Leja J. and Schulman J.H., Flotation theory: molecular interactions between frothers and collectors at solid–liquid–air interfaces, *Transactions of the Metallurgical Society of AIME*, **199**, 221-228 **(1954)**

10. Li Y., Xia W. and Zhang N., Efficiency and mechanism analysis of the flotation of anthracite coal using soybean oil as an alternative sustainable collector, *Energy Sources, Part A: Recovery, Utilization and Environmental Effects*, **43(18)**, 2210-2217 **(2021)**

11. Liu G.Q. and Wang X.L., Optimization of critical medium components using response surface methodology for biomass and extracellular polysaccharide production by Agaricus blazei, *Applied Microbiology and Biotechnology*, **74**, 78-83 **(2007)**

12. Martínez-L A. and Ortiz J.C., Study of celestite flotation efficiency using sodium dodecyl sulfonate collector: factorial experiment and statistical analysis of data, *International Journal of Mineral Processing*, **70(1-4)**, 83-97 **(2003)**

13. Naik P.K., Sukla L.B. and Das S.C., Aqueous SO_2 leaching studies on Nishikhal manganese ore through factorial experiment, *Hydrometallurgy*, **54(2-3)**, 217-228 **(2000)**

14. Rao G.V. and Mohanty S., Optimization of flotation parameters for enhancement of grade and recovery of phosphate from low-grade dolomitic rock phosphate ore from Jhamarkotra, India, *Mining, Metallurgy & Exploration*, **19**, 154-160 **(2002)**

15. Rubio J., Souza M.L. and Smith R.W., Overview of flotation as a wastewater treatment technique, *Minerals Engineering*, **15(3)**, 139-155 **(2002)**

16. Sayyad S.A., Panda B.P., Javed S. and Ali M., Optimization of nutrient parameters for lovastatin production by Monascus purpureus MTCC 369 under submerged fermentation using response surface methodology, *Applied Microbiology and Biotechnology*, **73**, 1054-1058 **(2007)**

17. Suma A., Ashika B.D., Roy C.L., Naresh S., Sunil K.S. and Sathyamurthy B., GCMS and FTIR analysis on the methanolic extract of red Vitis Vinifera seed, *World Journal of Pharmaceutical Sciences*, **6(8)**, 106-113 **(2018)**

18. Tanyildizi M.S., Özer D. and Elibol M., Optimization of α-amylase production by Bacillus sp. using response surface methodology, *Process Biochemistry*, **40(7)**, 2291-2296 **(2005)**

19. Vazifeh Y., Jorjani E. and Bagherian A., Optimization of reagent dosages for copper flotation using statistical technique, *Transactions of Nonferrous Metals Society of China*, **20(12)**, 2371-2378 **(2010)**.

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