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Application of response surface methodology (RSM) for optimization of leaching parameters for ash reduction from low-grade coal

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ABSTRACT

Coal is the world's most abundant energy source because of its abundance and relatively low cost. Due to the scarcity in the supply of high-grade coal, it is necessary to use low-grade coal for fulfilling energy demands of modern civilization. However, due to its high ash and moisture content, low-grade coal exerts the substantial impact on their consumption like pyrolysis, liquefaction, gasification and combustion process. The present research aimed to develop the efficient technique for the production of clean coal by optimizing the operating parameters with the help of response surface methodology. The effect of three independent variables such as hydrofluoric acid (HF) concentration (10–20% by volume), temperature (60–100 °C), and time (90–180 min), for ash reduction from the low-grade coal was investigated. A quadratic model was proposed to correlate the independent variables for maximum ash reduction at the optimum process condition by using central composite design (CCD) method. The study reveals that HF concentration was the most effective parameter for ash reduction in comparison with time and temperature. It may be due to the higher *F*-statistics value for HF concentration, which effects to large extent of ash reduction. The characterization of coal was evaluated by Fourier transform infrared spectroscopy (FTIR) analysis and Field-emission scanning electron microscopy with energy-dispersive X-ray (FESEM-EDX) analysis for confirmation of the ash reduction.

1. Introduction

Coal-fired thermal power sectors are the leading energy provider with significant growth in the world. India is the third largest country in the world for coal production, and approximately 85% of the total coal production is used in thermal power plants. Coal plays the primary dominant energy source for the generation of electricity, and it is expected to rise dramatically over the next few decades. It is very difficult to remove the mineral matter from the coal matrix by washing is due to the drift origin of Indian coal and wide variation of nature. Therefore, suitable beneficiation techniques are required for quality up-gradation of low-grade coal. India is endowed with the vast reserve of non-coking coal. It is hard to reduce the ash content to a certain limit by conventional coal washing technique because of the presence of the inherent minerals in the coal matrix. Indian coals are usually high ash and moisture content, which exerts a strong influence on their utilization, and such coals are neither acceptable for iron making nor suitable for power generation. Nowadays various beneficiation technologies are developed for upgrading the low-quality coal that may be appropriate for different fields. These beneficiation techniques are alternatively called as coal preparation, in which the undesirable impurities are physically separated from the coal [1]. The physical beneficiation techniques are based on the surface properties (wettability) and density of the mineral associated with the coal matrix [2]. It is mandatory to reduce the ash content from the coal for better utilization by required proper beneficiation technique to minimize the pollution in the environment. Similarly, the high sulfur bearing coals are not direct use, which also creates environmental pollution. Therefore, pre-desulfurization of the coal is the appropriate method to achieve the clean coal. The chemical leaching is an effective method to remove both organic and inorganic sulfur minerals from coal, as chemical solvents would easily diffuse through the coal matrix and dissolve the minerals [3]. Therefore, chemical beneficiation is one of the novel techniques that can remove inherent mineral and reduce ash-bearing mineral from the coal matrix. Several authors reported that the ash reduction was obtained by various leaching methods like acid leaching [4,5], alkali leaching under different operating conditions [6,7], leaching by molten caustic baths [8,9] and by leaching with

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Table 1
Chemical analyses of original coal sample.

Coal sample	Proximate analysis (wt.%, dry basis)			Ultimate analysis (wt.%, daf basis)				
	Volatile matter	Ash	Fixed carbon	Carbon	Hydrogen	Nitrogen	Sulfur	Oxygen (difference)
Ananta	30.6	39.9	29.5	39.37	3.21	0	0.37	57.05

calcium hydroxides [10,11]. The mineral matters in coal are categorized into carbonates, sulphates, phosphates, oxides, and sulphides. The presence of the major amount of silica and clay mineral content in the coal is mainly responsible for the formation of ash. The majority of silica and alumina found in the coal are in the form of kaolinite, dolomite, and quartz [12].

The effectiveness of the various demineralization depends on the composition and structure of the minerals in the coal matrix [13]. Steel et al., investigated the leaching behavior of the mineral matter in low-temperature ashing of Australian black coal by the effect HF and HCl. They reported HCl can dissolve the simple compounds like carbonates and phosphates, but it was less dissolute towards the clay mineral. The investigation result showed that HF could react with all types of mineral matter, except pyrite, and mostly all reaction products are water-soluble [14]. Steel and Patrick reported the production of ultra-clean coal (UCC) by chemical demineralization of high volatile UK coal. The coal was leached with HF at leaching temperature 65 °C and contact time 3 h, followed by HNO_3 in the same condition. The ash content of coal reduced from 7.9% to 2.6% and 67% demineralization was obtained by HF treatment alone and successive treatment of HF-treated coal with HNO_3. The ash content was decreased up to 0.63% by dissolution of residual mineral compounds such as CaF_2, MgF_2, AlF_3, $NaAlF_4$ and FeS_2 (pyrite) from HF treatment [15]. Rubiera et al. investigated the combustibility characteristics demineralized low ash bituminous coal with aqueous HF followed by HNO_3. The result was obtained from the investigation of over 95% of ash, and the SO_2 level was reduced by the leaching treatment. However, NO*x* emission was increased with nitric acid treatment [16]. Ning et al. studied the desulfurization of coal with a chemical reagent in combination with microwave irradiation [17]. By this investigation, the pyritic sulfur significantly decreased from 53.6% to 39.2% and organic sulfur reduced from 20.1% to 16.1% without the changes in coal properties.

Recently many statistical experimental design methods are used in the different chemical sector for optimization of process parameters. The optimization of a multivariable system in the conventional technique follows one-factor at a time. Many experiments are required for conventional techniques, and such methods do not represent the combined effect. It also requires more data to determine optimum level and take prolonged time, which is unreliable [18]. The primary purpose of the experimental design technique is to understand the interactions among the parameters, which could help in the optimization of experimental parameters and provide statistical models [19]. Choudhury studied the statistical and dimensional analysis for demineralized coal with NaOH and HCl by using fractional and factorial design method [20]. It has been found from the literature very limited work has been done on statistical optimization of process condition for ash reduction from low-grade coal by using HF acid treatment. Therefore, the present research aims to investigate the statistical optimization to determine the optimum leaching conditions for ash reduction from low-grade Indian coal by using aqueous HF leaching.

This paper investigates the combined effects of concentration, time and temperature on the leaching process, and the process parameters were optimized using central composite design (CCD) in conjunction with RSM method. A model was developed by using the design of experiment to determine the optimum leaching condition where the maximum ash reduction was obtained from the coal. In addition, this paper presents the comparison between the characteristic of raw and leached coal at the optimum condition with the help of FTIR and FESEM-EDS analysis.

2. Material and methods

2.1. Material

The coal sample was collected from Ananta open cast project of Mahanadi coalfield, Talcher, Orissa. It is one of the major coal producers in India, and a subsidiary of Coal India Limited. The lumped coal sample was crushed and sieved to −30+72 British standard sieve (BSS) mesh size required for the present study. Before the start of the experiment, the sample was dried in a hot air oven for 12 h at 105 °C. The proximate and ultimate analyses of the sample were determined in ASTM standard [21–24], and the results were summarized in Table 1. The analytical grade chemical reagent HF acid was used in this study. In this work, the coal sample was treated with aqueous hydrofluoric acid (HF) by varying the concentration at different operating parameter.

2.2. Methods

A series of experiments were conducted to find the maximum ash reduction from the coal where the optimum process condition maintained. For this purpose, the leaching experiments were carried out in a 1000 ml of Teflon reactor. A magnetic stirrer with a hot plate was used for the stirring and heating purpose. The experimental leaching set-up was shown in Fig. 1. The different concentration (10%, 12%, 15%, 18% and 20% by volume) of HF solutions were prepared for the leaching experiment. In each case, 10 g coal sample was treated with 100 ml HF solution. The reaction residence time varies from 60 min to 180 min, and temperature ranges from 60 °C to 100 °C. A plastic lid was covered on the top of the reactor to prevent the escape of vapours. After subsequent leaching, the treated sample is washed with distilled water to dilute the HF concentration and filtered in polypropylene funnel by using Whatman filter paper and dried in the hot air oven at 90 °C for 2–3 h. Finally, the ash reduction was calculated from Eq. (1).

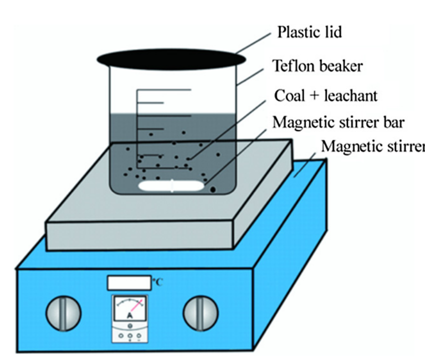

Fig. 1. Leaching experimental setup.