Fuel 165 (2016) 425–431

Contents lists available at ScienceDirect

Fuel

journal homepage: www.elsevier.com/locate/fuel

Organo-refining of high-ash Indian coals at bench scale

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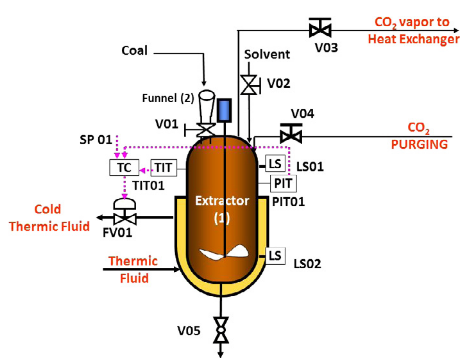
HIGHLIGHTS

- Production of <10% ash clean coal from 30% to 40% feed ash coal at bench scale.
- 6–12% points moisture reduction by decanter centrifuge over basket centrifuge.
- Mixing of two raw coals give 6% more yield rather than mixing their extracts.
- Free Swelling Index (FSI) improved from 1–5 range for feed coal to 7–8 for products.

GRAPHICAL ABSTRACT

This process has been demonstrated at bench scale. Organic matters were extracted from the coal using organic solvents. Clean coal with <10% ash was produced from feed having 30% to 40% mineral matter.

Piping & Instrumentation diagram of bench scale extractor



ARTICLE INFO

Article history:
Received 28 May 2015
Received in revised form 11 August 2015
Accepted 14 October 2015
Available online 23 October 2015

Keywords:
Solvent extraction
Clean coal
Swelling
Organo-refining
Bench scale

ABSTRACT

Organo-refining is a process of solvent extraction through which organic matters are extracted from coal using organic solvents. Indian coals have high mineral matter content ranging from 25 to 45 wt.% (dry basis). The ash in coal is highly disseminated, which makes it difficult to remove by physical beneficiation. Thus, chemical beneficiation is needed to remove the mineral matter. This study was conducted for high ash Indian coals at bench scale. The capacity of the bench scale plant was 40 kg/batch feed basis. Coking coal, washery discards and thermal coals of bituminous origin were used as feed material. The mineral matter of these coals varied in the range of 27–36 wt.% (db). The feed size of the particle was below −0.5 mm. N-methyl-2-pyrrolidone (NMP) and ethylenediamine (EDA) were used as main solvent and co-solvent respectively. The extraction was done at 200 °C with 1:6 coal to solvent ratio (by mass, wt./wt.) for 60 min. Extraction was followed by solid–liquid separation, solvent recovery and washing of products. The feed and product materials were subjected to proximate analysis, ultimate analysis, and Free Swelling Index (FSI) tests. Organo-refining process produced clean coal yield in the range of 30–45% and clean coal mineral matter was less than 10% for coking as well as non-coking coals. Free Swelling Index (FSI) also improved from 1–5 range for feed coals to 7–8 for product clean coals. Two types of filter (basket and centrifuge) were also tried for separation of solid and liquid from different types of slurries. It was observed that cake moisture can be reduced by 6–12% points for these slurries using decanter centrifuge. The cake moisture reduced to 70% from 76% for clean coal slurry and 60% to 48% for reject slurry. The clean coal yield increased by 5 units (from 35% to 40%) with decanter centrifuge filter. In order to have better filtration and precipitation, the extraction was carried out by mixing of two different raw coals (TL and TC) in a certain ratio (1:1 to 1:3). Also, the extraction was done separately for TL and TC coals and then the clean coal extracts were mixed in 1:1 to 1:3 proportions. Mixing of

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http://dx.doi.org/10.1016/j.fuel.2015.10.048
0016-2361/

two raw coals gives better results (36 wt.% yield with 6.4% ash clean coal) than mixing of their extracts (30 wt.% yield with 6.7% ash clean coal).

1. Introduction

The demand for clean coking coal in India is increasing due to government's new policy to increase steel production from the current level of around 90 million ton per annum (mtpa) to 200 mtpa by 2020. A number of new steel projects have been announced to increase the steelmaking capacity in India, and the demand for coking coal in India will rise by more than 100 mtpa [1]. Indian coal industry is able to meet only <30% of total coking coal requirements. This is mainly due to high mineral matter content and limited reserve of good coking coal in India. High mineral matter is undesirable. It decreases blast furnace productivity and increases energy consumption. It also contributes to more CO_2 emission and more solids wastes generation. In general, more than 60% of the raw coal mined in Tata Steel's collieries is discarded as middling/tailings due to high mineral matter. If ash issue is resolved, these discards and non-coking coals are potential source for its application in coke making with innovative solutions. India had coal demand of about 731 MT (million tonne) for the period of 2013–14, out of which 565 MT was met by indigenous production and the remaining gap of 165 MT was fulfilled by import [1]. Coal demand will reach up to 980 MT by 2016–17 at CAGR (compound annual growth rate) of 8.9%, and domestic supply will reach up to 795 MT. Similarly, at CAGR of 7% the demand is expected to reach 1373 MT by 2021–22 with a domestic coal supply of 1100 MT. The ministry of coal is planning indigenous coal production of 1.5 billion tonne by 2019–20 [1]. Although, mineral matter content of about 10–12% in the clean coal is most suitable for steel industry, the same could not be exercised because of drop in yield of clean coal during beneficiation leading to the process becoming uneconomical. Hence, most of the steel companies in India are importing metallurgical coal from Australia, New Zealand, Canada, etc. [1]. However, due to increased cost of imported coals, the Indian companies are falling back in their cost competitiveness in the global market. Coal security is also a major issue for these companies. A fresh look is, therefore, essential to look into indigenous coals, especially to drive further lowering mineral matter.

The cleaning of coal, generally means removal of mineral matter, can be done by both chemical and physical treatments [2]. Physical cleaning of coal, although a cheaper solution than chemical treatment, has limited yield. On the other hand chemical treatment gives substantially higher yield with lesser ash content clean product at the cost of higher energy consumption and cost. The gap between cost and yield is still a major constraint and drives the people to adopt the technology accordingly. The present study uses chemical beneficiation of coal (termed as "Organo-refining") to remove the mineral matter of Indian coals.

Coal is an organic sedimentary rock which contains varying amounts of carbon, hydrogen, nitrogen, oxygen and sulphur as well as trace amounts of other elements, including mineral matter [2,3]. The physical constitution of Indian coal and macromolecular network structure of bituminous coals were reported by Navale [4] and Larsen et al. [5]. Coal is a complex material and shows a wide range of physical properties. There exist different physical forces, such as, Hydrogen bonds, London forces, and Van der Wall's forces, as intermolecular forces among these molecules [6]. The solvent molecules disrupt these interactions and penetrate into the solid coal. During extraction of coal, the solvents must get through the solid coal matrix.

Efficient contact between the coal and solvent is also an important physical development. Contact between the coal and solvent is increased by using fine coal; use of coarse coal is always ineffective [6,7]. Miura et al. [8] from Kyoto University reported that bituminous coals can be separated into different molecular size fractions without decomposition through extraction at below 350 °C. It has been reported that N-methyl-2-pyrrolidinone (NMP) is one of the most effective solvent for coal extraction [9–14]. Renganathan et al. [9] reported that extraction yield was 74% with 0.1% ash yield at 202 °C temperature and atmospheric pressure. Iino et al. [10,11] reported that carbon disulphide (CS_2) and NMP mixture (in same ratio) gave more than 60% extraction yield for some bituminous coals at room temperature. Recently, Miura et al. [15] reported the extraction of coal using tetralin or 1-methylnaphthalene (1-MN) solvent at 200–400 °C and 10 MPa, the extraction yield was 65–85 wt.% for the bituminous coals at 350 °C. Investigation of associated structure of Upper Freeport coal, effect of solvent basicity on the kinetics, macromolecular structure study and effect of binary solvent mixtures on solvent swelling of coal was reported by some researchers [16–19].

Yoshida et al. [20] have been working since 90s on production of hyper-coal. They reported that crude methylnaphthalene oil (CMNO) gives much higher extraction yield than light cycle oil (LCO). They used several organic solvents under hot filtration at 360 °C using a flow type reactor. They reported that high extraction yield was obtained due to the relaxation of coal molecules by the solvent, in addition to the thermal-induced relaxation. Okuyama et al. [21] developed the process for ash-free coal production. They found that ash removal using solvent is an assuring way to produce hyper-coal in a competitive price. Research on hyper-coal characterization, effects of pretreatment with carbonic acid and the addition of polar compounds was reported by Japanese scientists [22–25]. Renganathan and Zondlo [26] reported extraction mechanism of bituminous coals using NMP. Kinetic studies and shrinking core model on solvolytic extraction of coal was reported by Giri and Sharma [27]. They found that extractions were first order rate law. Choudhury et al. [28,29] reported results on high-ash non-coking coals and washery middlings. Their aim was to prepare a potential coking additive and solvent-refined coal from high-ash Indian coals in a bench scale unit under hydrogen pressure.

R&D, Tata Steel started a project on chemical beneficiation of coal due to limitation of physical beneficiation in terms of deashing of coal. Broadly, chemical beneficiation is possible by chemical demineralization of mineral matter present in coal with hydrochloric acid, nitric acid, hydrofluoric acid, caustic soda etc., or through solvent extraction, by dissolving organic matter of coal in various organic solvents for production of low ash coal. Coal demineralization study of Indian coals was done by Dash et al. [30]. They conducted leaching experiments at high temperature and pressure. Extensive work on chemical demineralization was reported by Steel et al. [31–34]. They used hydrochloric and hydrofluoric acids for production of ultra clean coal by chemical demineralization. Coal structure and reactivity changes caused by chemical demineralization were reported by Rubiera et al. [35]. Similarly, organo-refining process was developed in the laboratory with optimization of different parameters. After successful completion in the lab, it was scaled up to 40 kg/batch coal bench scale plant. We [36] reported on optimization of solvent extraction process parameters. It was found that maximum yield was obtained