A Review on Desulphurization of Liquid Fuel by Adsorption

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Abstract: *Sulfur compounds create impurities in the liquid oil. Because of the requirement for low sulfur content of fuel oils, it is necessary to develop methods for removal of sulphur from liquid oil. Adsorption is widely used to remove sulphur from liquid fuel. The Sulphur removal from liquid fuel by adsorption is very important area of research. Many researchers have done their research on this area. This review paper presents the idea about the removal of sulphur from liquid fuel by various researchers. Adsorption is one of the easily and fast method to remove sulfur from liquid fuel.*

Keywords: Adsorption, Sulphur, Liquid fuel

1. Introduction

Sulphur causes environmental pollution if present in the liquid fuels. It is therefore needed to develop efficient and affordable processes for desulphurization of liquid fuels. Present review paper is aimed and studying the research carried out in order to investigate the mechanism of removal of sulphur from liquid fuel by adsorption using different adsorbent. Adsorption is a low cost and important physical process for the removal of sulphur from liquid fuel.

1.1 Adsorption
Adsorption is a process of accumulation of one substance over another. It occurs between liquid-liquid, gas-liquid, gas-solid, or liquid solid interface. Typical liquid separation includes the removal moisture dissolve din gasoline decolourization of petroleum products and fractionations of mixtures of aromatic and paraffinic hydrocarbons .Adsorption may be physical or chemical. In physical adsorption intermolecular forces of attraction between molecules of the solid and the substance adsorbed. Chemisorptions or activated adsorption is an interaction between the solid and adsorbed substance.

1.2 Adsorbents
Adsorbent solids are used in granular form varying in size. They must have adequate strength and hardness. There is different type of adsorbents in general use for removal of sulphur from liquid fuel. A porous Zeolite catalytic support mainly composed of Alumina and Silica was synthesized, and the said catalytic support loaded with Single walled carbon Nanotubes (SWNT's) to enhance the catalyst adsorption capability [1] the commercial powder activated carbon and acid activated bentonite got the highest sulfur removal efficiency and the sulfur content. monotmorollonite clay as adsorbent in desulphurization of kerosene and diesel and adsorption study carried out in1 hr time at room temp 25^0c. [23]

2. Literature Review

Khan et al., [1] used oxidative desulphurization followed by catalytic adsorption method. The author studied oxidative desulphurization followed by catalytic adsorption for desulfurization of diesel oil. The investigator reported that, effectiveness of the process mainly depends on efficient decomposition of H_2O_2 in the diesel to oxidize various organic sulfur compounds (sulfones, sulfoxides) and to extract them via polar solvent and developing and analyze the catalytic adsorbent for the adsorption of unextracted aromatic sulphur compound (DBT). Author also concluded that a porous Zeolite catalytic support mainly composed of Alumina and Silica was synthesized, and the said catalytic support loaded with Single walled carbon Nanotubes (SWNT's) to enhance the catalyst adsorption capability.

Unsteady state adsorption column studies were carried out by Patil [2]. Author used fixed-bed adsorber for the desulfurization of hydrocarbon liquid fuel by an activated carbon. Runs were carried out, with continuous flow of feed solution through a column packed with activated carbon as an adsorbent. Investigators studied effect of parameters like, feed flow rate, feed solution concentration, and adsorbent bed height on rate of adsorption. Increase in feed flow rate decreases adsorption zone height Za, at all concentration studied. Author also concluded with increasing amounts of fluid are passed through such a bed, the solid adsorbs increasing amounts of solute, and an unsteady state prevails.

Comparative analysis of adsorptive desulphurization of crude oil by manganese dioxide and zinc oxide studied by Adeyi and Adekanmi [3] .The investigators used selective adsorption for removing sulphur for separating a less than 1 wt. % of fuel mass and leave the 99 wt. % of non sulphur-containing crude oil mass untouched. The authors reported that comparative study was made of the desulphurization potentials of two metal oxides-activated manganese dioxide, activated zinc oxide. The investigators also reported that data was found to follow both Langmuir and Freundlich models for the balance analysis of the adsorptive desulphurization.

Hosseini et.al [4], studied novel methods for desulfurization of fuel Oils. The investigators used ultrasound-assisted oxidative desulfurization method for the oxidization of compounds such as benzothiophene and dibenzothiophene .The Author reported that the removal of sulfur compounds is reach about 99%.

Bhattacharyulu [5] studied desulphurization of hydrocarbon liquid fuels by adsorption. The investigator used batch reactor for sorption of sulfur onto activated carbons which prepared from black liquor and Phosphoric acid and nitrogen reactor used as intercalating agents. Author also reported that intra-particle diffusion resistance has been overcome due to stirring and experimental data obtained obeys Langmuir adsorption isotherm model.

Shimizu, et al., [6] used rice husk activated carbon for adsorptive removal of sulphur compounds .The study conducted using rice husks carbonized in N_2 at 400˚C for 1 h and then activated in CO_2 at 850˚C for 1 h. The author have reported that the capacities of rice husk activated carbons (RHACs) to adsorb refractory sulfur compounds of dibenzothiophenes (DBTs) evaluated correlating with their textural and chemical characteristics and the RHACs of 0.5 g were soaked in commercial kerosene of 15.0 g at 10˚C for 100 h.

Isam and Zubaid et al., [7] used Adsorption desulfurization of diesel oil studied using seven sorbents such as Bentonite, acid activated bentonite, date palm kernel powder, acid activated date palm kernel powder, sawdust powder, and commercial powder and granules activated carbon .The study conducted the experiments using amount of sorbents was ranging from 0-5% by mass in a batch process at room temperature with contact time of two hours. The sulfur content was measured using X-ray fluorescence analyzer. Authors reported that the commercial powder activated carbon and acid activated bentonite got the highest sulfur removal efficiency and the sulfur content reduced from 410.9 µg/g to 245.9 µg/g using 5% by mass commercial powder activated carbon and 278 µg/g using acid activated bentonite Authors also reported that data found to follow Langmuir and Frendlich isotherm each sorbent.

Patil, [8] studied ultrasound-assisted desulfurization of commercial kerosene by adsorption. The author investigated ultrasonic irradiation for the sorption of hexyl mercaptan sulfur onto carbon-based adsorbents i.e. activated carbon and carbon nanotubes. Author reported that Carbon nanotubes show higher adsorptive capacity after examining the adsorptive capacity and the experimental data fitted to the Langmuir and Freundlich adsorption isotherm model.

Toida, [9] studied adsorptive desulfurization of commercial Kerosene with Sulfated Alumina Producing Heavier Organic Sulfur Compounds. The studies were conducted for the adsorptive desulfurization of commercial kerosene using acidic adsorbent, especially sulfated alumina at moderate condition on batch system and on continuous flow system. The investigators reported that the acid property seemed to effect on adsorption performance and adsorbents combination of 4.1 l, 0.3 l of activated carbon supporting copper oxide and silver oxide and 3.8 l of Lewis acid type sulfate alumina in series, evaluated for long term adsorptive desulfurization of commercial kerosene without using hydrogen at ambient temperature at a flow rate of 105 cm^3/h.

Liquid phase adsorptive desulphurization of diesel fuel studied by Karagianakis [10].The studies were conducted for the lab-scale liquid desulfurization of commercial diesel fuel via adsorption, under ambient conditions, employing a high-surface area activated carbon (AC) sorbent. During the studies the investigators observed that the breakthrough curves for a processed diesel fuel amount of up to about 20 ml , the total sulfur content maintained below or equal to 2 ppmw and the aforementioned value for the breakthrough content of 2 ppm w of total sulfur was decreased to approximately 11 ml .

Arturo, [11] studied desulfurization of a commercial diesel fuel by different adsorbents in a fixed bed adsorber operated at ambient temperature and pressure. Investigator reported that the best adsorbent, AC/Cu(I)-Y (layered bed of 15 wt % activated carbon followed by Cu(I)Y), is capable of producing 30 cm3 of diesel fuel per gram of adsorbent with a weighted average content of 0.15 ppmw-S, and about 20 cm3 of diesel fuel per gram of adsorbent with a weighted average content of 0.06 ppmw-S authors also concluded that π-complexation sorbents selectively adsorbed highly substituted thiophenes, benzothiophenes, and dibenzothiophenes from diesel, which is not possible with conventional hydrodesulphurization (HDS) reactors.

Isam et al., [12] studied adsorption Process of Sulfur Removal from Diesel Oil Using Sorbent Materials. Investigators treated Local diesel fuel of 410 ppm sulfur content with commercial activated carbon and carbonized date palm kernel powder at room temperature in batch work.

Herna et al., [13] studied hydrodesulphurization process for removal of sulphur from liquid fuel using Co-Mo/Al_2O3 or Ni-Mo/Al_2O3 catalyst. Author also reported that the reactor size needs to be increased by factors of 5-15.4 .Investigators showed that aqueous- phase copper(II)-exchanged Y-zeolites, when auto reduced to Cu(I)-Y, are capable of removing 0.20 mol of organo-sulfur species from a commercial diesel fuel (297.2 ppmw total sulfur) per gram of zeolite5-7 .

Khodadia [14] studied adsorptive desulfurization of diesel fuel using CuO nano particles adsorbent. Author reported that Kinetic study for adsorbent has shown that the best fit is achieved when the pseudo –second order model applied which indicates that the reaction path is second order and experimental data fitted in Freundlich isotherm model.

Desulfurization of model oil via adsorption by copper (II) modified bentonite studied by Dezhi [15] Author investigates desulfurization process by adsorption for removal of sulphur from hydrocarbons. Bentonite adsorbents modified by $CuCl_2$ for the desulfurization of model oil was investigated. Author also reported that the modified bentonite adsorbents are effective for adsorption of DMS and PM and authors studied several factors that influence the desulfurization capability, including loading and calcination temperature.

Huan et al., [16] reactive studied adsorption desulfurization of model gasoline on Ni/ZnO-HY adsorbent. Investigator reported that, under the conditions of this study, which specified a 50% ratio of HY-zeolite in the adsorbent, a Zn/ Ni molar ratio of 10, and a reduction temperature of 400 ℃, the Ni/ZnO-HY adsorbent showed the best desulfurization performance. Authors concluded that, the sulfur capacity of Ni/ZnO-HY adsorbent could be recovered to 92.19% of the fresh one after being subjected to regeneration at 500 ℃, and could be maintained at 82.17% of the fresh one after 5 regeneration cycles.

Zhitao, [17] carried out research on desulfurization of naphtha by modified Y Zeolite. Authors conducted dynamic adsorptive desulfurization of naphtha in the presence of the modified LaY zeolite, and the sulfur content of the treated naphtha samples analyzed by microcoulometry. The investigator reported that under dynamic conditions the LaY zeolite prepared through secondary ion-exchange of NH_4Y zeolite, which was prepared using 1.0 mol/L ammonium salt, with the rare earth salt exhibited better desulfurization efficiency. Authors also reported that the LaY zeolite achieved a best desulfurization effect at an adsorption temperature of 45 ℃ and an adsorbent/oil ratio of 1:2.

Deep desulfurization via adsorption by silver modified bentonite was studied by Xiaolin [18].Author investigated a desulfurization process by adsorption for removing alkyl dibenzothiophenes from liquid hydrocarbons. Authors studied desulfurization of model gasoline by bentonite adsorbents loaded with silver nitrate. Investigators studied several factors influencing the desulfurization capability, including the Ag+ loading, the baking temperature, as well as the reaction temperature. Authors also reported that the adsorption capacity of the alkyl dibenzothiophenes on bentonite loaded with Ag+ ions increased with a decreasing temperature.

Mehdizadeh, et.al. [19] studied deep desulfurization of diesels using alkyl sulfate and Nitrate Containing Imidazolium as Ionic liquids. Investigator investigated the efficiency of sulfur removal from BT and DBT solutions as well as the effect of anion and cation chain length. Author reported the multiple extractions of BT and DBT solutions were also carried out to reduce sulfur content to 81% and 68% for DBT and BT solutions, respectively, after 5 extraction cycles.

Blanc, et.al. [20]studied Removal of refractory organic sulphur compounds in fossil fuels using MOF sorbents. Author reported that adsorption capacity can be improve and sorbent regenerated, by increasing specific desulfurization activity, hydrocarbon phase tolerance, sulfur removal at higher temperature, and development of new porous substrates for desulfurization of a broader range of sulfur compounds. Investigators comprehensively described the adsorption of organo-sulfur compounds present in liquid fuels on metal-organic framework (MOF) compounds.

Velu et al., [21] studied on reganarable adsorbents for the adsorptive desulphurization of transportation fuel cell for fuel cell applications. Authors reported that, adsorptive desulfurization is a promising approach and several new processes, such as IRVAD and Philips S-Zorb processes.

Shiraishi and Naito et al., [22] studied, a desulfurization process for light oil using a polymer-supported imitation agent (PI, sodium N-chloro-polystyrene sulfonamide). Investigators reported that when n-tetrad cane solution (model light oil) containing dibenzothiophene (DBT) stirred with methanol in the presence of PI and acetic acid at a temperature of 323 K, DBT is chemically adsorbed on the PI, via the imitation of sulfur atoms on DBT by the PI, and removed successfully from the oil. And the polymer obtained was insoluble to the solutions and could be recovered by filtration. They also reported that the sulfur concentration of commercial light oil was decreased successfully from 400 ppm to less than 50 ppm (ultra-deep desulfurization level).

Adsorptive desulphurization of kerosene and diesel oil by Zn impregnated monotmorollonite clay studied by Ahmad [23]. Authors used monotmorollonite clay as adsorbent in desulphurization of kerosene and Diesel and adsorption study carried out in1 hr time at room temp 25^0c.Investigators reported that desulphurization of kerosene is 76% and that of diesel is 77% is achieved.

Adsorptive desulfurization and denitrogenation studied by, Haung [24]using a model diesel fuel, which contains sulfur, nitrogen and aromatic compounds, over three typical adsorbents (activated carbon, activated alumina and nickel-based adsorbent) in a fixed-bed adsorption system. The adsorptive capacity and selectivity for the various compounds were examined by investigators and compared on the basis of the breakthrough curves. Author reported that the activated carbon shows higher adsorptive capacity and selectivity for both sulfur and nitrogen compounds, especially for the sulfur compounds with methyl substituent's, such as 4, 6-methyldiben- zothiophene.

M.Muic et al., [25] a design of experiments investigation of adsorptive desulfurization of diesel fuel. Investigators investigated, two designs of experiments (DOE) methods which were carried out in a batch adsorption system using Chemviron Carbon, activated carbon as adsorbent. Authors also reported that, the effects of individual factors and their interactions on sulfur concentration and sorption capacity and statistical models of the process developed. The first-order models predict the behavior of the system rather well but significant curvature detected by investigators. Subsequently developed second-order models were able to give reasonably well descriptions of the system.

An evaluation of desulfurization technologies for sulfur removal from liquid fuel studied by Srivastava [26]. The research efforts for developing conventional hydro desulfurization and alternative desulfurization methods such as selective adsorption, biodesulfurization oxidation/extraction (oxidative desulphurization), etc. for removing these refractory sulfur compounds from petroleum products are on the rise Author investigated current status in detail of various desulphurization techniques being studied worldwide.

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Pyshyev,[27] investigated that the fractions of diesel oil desulphurized by air oxidation without the catalysts in the presence of water and the products of sulphuric compounds oxidation were extracted by adsorption or joint adsorption and rectification. The samples of desulphurized diesel oil have been analyzed in accordance with reference documents after their treatment by alkali. Authors also shows that, lubricity of the raw materials (straight-run diesel oil and hydrogenised fuel), desulphurized straight-run diesel oil (DSDO), hydrogenised fuel with DSDO additives and two samples of the fraction 553–623 K purified in the presence of water and without it has been determined.

Marko,[28] develop an idea to selectively separate less than 1 wt.% of fuel mass by selective adsorption for removing sulfur, and leave the 99 wt.% of non- sulfur-containing fuel mass untouched. Adsorptive desulfurization was carried out by investigators in laboratory apparatus designed for batch adsorption applying activated carbon and aluminum oxide. Kinetic and equilibrium analysis of the adsorption process was done. Authors investigates results of these experiments showed that activated carbon had significantly better performance regarding the lowering of sulfur content and adsorption capacity. Statistical analysis of the data obtained from the experiments, carried out according to 2 3 factorial design, was used to determine the influence of time, initial sulfur concentration, activated carbon mass and their interactional effects on sulfur content and adsorption capacity.

Music, [29] investigates adsorptive desulfurization of diesel fuel by applying two design of experiments (DOE) methods. The experiments were carried out in a batch adsorption system using Chemviron Carbon and activated carbon as adsorbent. The first DOE method employed was a full factorial with three factors on two levels and five center points, and the second was Box-Behneken design with the same three factors but on three levels. The effects of individual factors and their interactions on sulfur concentration and sorption capacity were determined by investigators and statistical models of the process developed. The first-order models predict the behavior of the system rather well but significant curvature was detected. Subsequently developed second-order models were able to give reasonably well descriptions of the system. Authors also studied that the lowest achieved output sulfur concentration was 7.6 mg kg–1 with relatively low sorption capacity of 0.0861 mg g–1.

Wangliang et al., [30] a novel integration study on the feasibility of the use of a combined physical-biological procedure for desulfurization of fuels. Investigators also studied that sulfur compounds are removed through adsorption with the regeneration of the adsorbent. They select suitable π-complexation adsorbents for desulfurization, dibenzothiophene (DBT) was used as a model compound to study the adsorption capacity of these adsorbents. Different metal ions, namely Co^{2+}, Ni^{2+}, Ce^{3+}, and Cu^+ were used to prepare desulfurization adsorbents using the ion exchange method. Authors also studied reduction behavior of Cu (II)-Y by using temperature programmed reduction (TPR) with reducing gas (10% H_2 and 90% N_2, v/v). Authors also

suggested that The acidity of the adsorbents was characterized by pyridine adsorption thermo gravimetric-differential thermal analysis.

Chunshan, [31] discusses the problems of sulfur reduction in highway and non-road fuels and presents an overview of new approaches and emerging technologies for ultra-deep desulfurization of refinery streams for ultra-clean (ultra-low-sulfur) gasoline, diesel fuels and jet fuels. Authors also studied that current gasoline desulfurization problem is dominated by the issues of sulfur removal from FCC naphtha, which contributes about 35% of gasoline pool but over 90% of sulfur in gasoline. Deep reduction of gasoline sulfur (from 330 to 30ppm) must be made without decreasing octane number or losing gasoline yield.

3. Conclusion

This paper review the various papers published in journals on removal of sulphur from liquid fuel. The removal of sulphur by using sorbent is very attractive alternative. Though the research has been carried out by using various sorbents, there is still scope for the research in this field, with new possibilities in terms of improving the removal efficiency and availability of adsorbents.

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