



# Techno-economic evaluation of quality improvement of heavy gas oil with different processes



Zoltán Varga\*, Zoltán Eller, Jenő Hancsók

University of Pannonia, MOL Department of Hydrocarbon and Coal Processing, Egyetem u. 10, H-8200 Veszprém, Hungary

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## ABSTRACT

Nowadays, high quality diesel fuels can only be produced by the hydrotreatment of various refinery streams. Preparing catalytic experiments and process engineering calculations are inevitable for selecting the most technically and economically appropriate process. There is limited information in the free literature about the complex approach of hydrotreating processes that deal with both the catalytic and process issues. This paper summarizes the results of an engineering study to evaluate the techno-economic feasibility of different types of hydrotreating processes for producing high quality gas oil blending components from heavy gas oil fraction. Hydrotreatment of feed in one-stage on nickel(Ni) molybdenum(Mo)/alumina and in two-stage on NiMo/alumina followed by on platinum(Pt) palladium(Pd)/zeolite catalysts were investigated. In one stage hydroprocessing sulphur and polyaromatic contents of the products met the requirements of the standard only if strict process parameters were applied (temperature > 370 °C and Liquid Hourly Space Velocity < 1.5 h<sup>-1</sup>) which resulted in operation problems and product loss, moreover the advantageous process parameters of hydrodesulphurization and hydrodearomatization did not coincide. In two-stage hydrotreating results showed that the advantageous process parameters for reduction of sulphur and aromatics coincided. Advantageous process parameters to produce gas oil of quality meeting the requirement of the standard were determined. Based on the experimental results model for both alternatives was prepared, and heat and material balances were determined. By applying two step hydrotreating, the yield of gas oil having low sulphur and aromatic content was 6.2% higher than that obtained in one step. Heat exchanger network analysis and sizing of main equipments were accomplished. Operating and investment costs were estimated for both processes, which showed that all the cost elements were higher for the two-stage process. But it provides the following advantages in comparison to one-stage process: higher yield of gas oil, better product quality allowing the application of low value hydrocarbon streams in higher quantity at the blending, the lower density of products would be the source of extra profit due to diesel fuels are sold on volume basis at the petrol stations. Estimation of emission potential of feed and products was prepared, too. This showed that the hydrotreating of heavy gas oil contributes to significant reduction in emission of particulate matter, hydrocarbons and carbon monoxide, but the effect on the emission of nitrogen oxides was scarce. Product of two-stage process provided considerably greater decrease in carbon monoxide and hydrocarbon emissions comparing to the one-stage alternative (52% vs. 14% and 54% vs. 14%). Consequently, the estimation of emission potential of hydrotreated products can be used as one of the parameters for selecting between various process alternatives.

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## 1. Introduction

The quantity requirements for good quality diesel fuels having low sulphur and aromatic content increased all over the World in

\* Corresponding author.

E-mail address: [vargaz@almos.uni-pannon.hu](mailto:vargaz@almos.uni-pannon.hu) (Z. Varga).

recent years (Stanislaus et al., 2010). The most significant change was the maximum allowable sulphur content of diesel fuels (Varga and Hancsók, 2003, 2014). At the same time, the quality of the available crude oil stocks continuously declined giving tough challenges for refineries to produce high quality diesel fuels in increasing quantity, which can be overcome by change in the refinery configuration (Arora and Mukherjee, 2011) as well as improving the efficiency of the hydrotreating processes (Peng et al.,

2012). The economic and political events occurred recently highlight the attention of the refining industry to diversify the crude oil sources and to increase the flexibility of the whole petroleum processing train (Varga and Danics, 2013).

Hydroprocessing is the turnkey technology of choice for refineries to meet both with the quantity and quality demands (Leliveld and Eijssbouts, 2008). Despite this process more than 75 years old continuous innovation has been taking place to improve both activity of catalysts and efficiency of the entire process. There is no silver bullet; every feedstock requires special approach on processing. Unfortunately, the production of clean fuels increases the environmental burdens on the refinery site. Life-cycle assessment prepared by Burgess and Brennan (Burgess and Brennan, 2001) for producing low sulphur diesel fuel from straight run gas oil showed that the hydrotreater unit is the major contributor to both environmental and economic burdens. While the other units e.g. hydrogen production, hydrogen sulphide removal by absorption/stripping, and sulphur recovery from hydrogen sulphide played only minor role.

The quantity of diesel fuels can be increased with hydrocracking of heavy oils. Kochaphum and co-workers (Kochaphum et al., 2012) investigated the Global Warming Potential (GWP) and Abiotic resource Depletion Potential (ADP) both of straight run diesel and cracked diesel. They determined that the production of diesel fuel by using only gas oil desulphurizing unit (simple hydroskim refinery) required less energy, accordingly lower greenhouse gas (GHG) emission (lower GWP) in comparing to the technology route applying hydrocracking of heavy fractions (complex refinery). In contrast, the hydrocracked diesel has a lower ADP than the straight run one proving that the production of diesel from the heavy distillate indeed helps conserve natural resources. However, the level of GHG emissions of hydrocracking process can be reduced by using energy efficiency techniques e.g. energy integration, hydrogen pinch, waste heat utilization etc., which may also result in further reduction in using of resources.

A good example for this the research work of Umana and co-authors (Umana et al., 2014), who used an integrated approach for refinery process and hydrogen network design. The model is validated with different feedstock properties and shows good agreement with experimental data. Modelling and optimization of the overall network is performed and the effects of process and operational variables on performance indicators and hydrogen production requirements are investigated. Potential hydrogen saving of 2% was realized from varying  $H_2$  inlet conditions and a further reduction of 7% was achieved by optimizing temperature.

Another way to increase the quantity of middle distillates and to decrease the environmental impacts of the hydrocarbon processing industry as well is blending of renewable components into the gas oil pool. Tóth and co-workers (Tóth et al., 2016) investigated the co-processing of vegetable oil and gas oil. They stated that small amount (10%) of gas oil in the mixture can ensure the sulphide state of hydroprocessing catalysts. Quantity of gas oil can be increased by mixing bio-components (e.g. waste animal fat) and low value refinery streams (e.g. LCO) into the straight run products and hydrotreating the obtained stream (Sági et al., 2016). They stated that significant quality improvement can be obtained in all feedstock components at adequate process conditions. Festel and co-workers (Festel et al., 2014) compared various biofuels to their fossil counter parts based on the price of production. According to the model being developed they stated 2nd generation biodiesel from waste oil and from palm oil are most promising with regard to production costs both in short and medium terms. However, a serious doubt for all types of biofuels remains whether sufficient amount of feedstock can be generated to satisfy the growing demand and to achieve the shift from fossil fuel to biofuels. This paper

also supports that production of diesel fuel is rely on fossil sources in long time period.

The heart of hydroprocessing units is the reactor system. Therefore, several books (Ancheyta, 2011) and papers were published to model the reactor of hydrotreating process. The key issue to optimize these units is the availability of reliable kinetic models for this complex, tri-phase reaction. De la Paz-Zavala and co-workers (De la Paz-Zavala et al., 2013) prepared a HDS reactor model using data to be obtained in pilot scale experiments, then extended to industrial scale. de Oliveira and co-workers (de Oliveira et al., 2012) worked out a hydrotreating reactor model applying the detailed feed composition and modelling the processes taking place in a hydrotreating reactor using the kinetic Monte Carlo (kMC) method. Ferreira and co-workers (Ferreira et al., 2013) developed a conventional neural network (NN) training algorithms for inducing NNs to predict kinetic parameters of simplified models for the catalytic hydrodesulfurization (HDS) reaction, using macro properties of the feed as input, the paper proposes and describes an ad hoc methodology for artificially enlarging the initial scarce experimental data. Mederos and co-workers (Mederos et al., 2012) developed a dynamic heterogeneous one-dimensional model of trickle-bed reactor used for catalytic hydrotreating of oil fractions. The required kinetic parameters were determined from experimental data obtained in an isothermal bench-scale reactor during hydrotreating of atmospheric gas oil. The model was used to predict the dynamic behaviour of an industrial hydrotreating reactor within a wide range of reaction conditions. Chandak and co-workers (Chandak et al., 2014) carried out hydrotreating of a gas oil blend on a commercial type catalyst. They investigated the effect of the hydrogen partial pressure on the performance of catalyst. The conclusion was decreasing of the partial pressure below 52 bar shortened the cycle length of the catalyst due to the higher deactivation rate.

As it was shown, all of presented works were highly rely on experimental data obtained in pilot scale reactor system, which clearly shows the importance of catalytic experiments for producing diesel fuels of high quality.

However, few papers were published concerning the model of the whole technology as well as providing different process alternatives. From these Ahmad and co-workers (Ahmad et al., 2011) showed an integrated approach for the design of diesel hydro-treating processes employing a simulated annealing optimization algorithm. The proposed integrated approach takes into account the trade-offs between capital and operating costs as well as interactions between the hydrotreater, distillation column, and the associated heat exchanger network.

The main aim of the presented work was to combine the experimental work and process engineering knowledge to carry out techno-economic evaluation of quality improvement of heavy gas oil fraction applying different technology ways. It covered the identification and quantification of key process parameters for the hydrodesulfurization (HDS) and aromatic content reduction (HDA) of a heavy gas oil carried out in one step and in two steps. Based on the obtained results, process model for both hydro-treating alternatives was prepared. Then the advantageous technology arrangement considering both the HDS and HDA reactions was determined. Emission potential of feed and products of the selected alternatives was estimated.

## 2. Experimental and methods

The aim of the research work was to connect the catalytic experiments with the process engineering in order to produce high quality and clean burning diesel fuel at minimal energy consumption. From this reason, the circumstances of catalytic

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