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Modern Coke Oven and Its By-Product Gas Treatment Plant Operation Management and Optimization Methods

Naga Sai Prasad[1*], Shakti Prasad Jena[2]

[1*] Associate Professor, Department of Mechanical Engineering, Nalanda Institute of Technology, Bhubaneswar, Odisha, India.
[2]Assistant Professor, Department of Mechanical Engineering, Nalanda Institute of Technology, Bhubaneswar, Odisha, India.

*Corresponding author e-mail: nagasaiprasad@thenalanda.com

Abstract:
Reducing emissions, preventing waste, and conserving energy and the environment were technical concerns throughout the engineering phase of the Gas Treatment Plant (GTP) implementation in the Coke Oven unit of the Bkaro Steel Plant. Three technical solutions are described below as examples of other advancements based on DMT's expertise and experience in the field of by-product plants. A safe operation with stable conditions and a long lifespan for the entire plant are the general goals of the design of the complete GTP, which is based on current features, management, and design upgrades. There were two optimization techniques looked at. When just purity was taken into consideration, concentrations of H2S, NH3, and HCN were determined to be 0.54 g/Nm3, 0.01 g/Nm3, and 0.03 g/Nm3, respectively. The quantities of H2S, NH3, and HCN increased to 1 g/Nm3, 0.5 g/Nm3, and 0.04 g/Nm3, respectively, when the temperature of the washing water was taken into account, however they remained below the environmental standards. The latter scenario, however, will be more energy-efficient because it can be carried out without the use of refrigeration and allows for lower washing water streams.

1. Introduction

One of the most important factors influencing economic growth and development in industrialised nations is the cost of various energy sources [1]. They have an impact on wages, the cost of production, and the costs of raw materials and finished goods [2]. Hence, it is imperative to reduce energy usage, particularly in energy-intensive industries like refineries, steelworks, paper mills, and coke oven facilities. Energy prices reached an all-time high by the end of 2021 and stayed there into the first quarter of 2022. Since the start of 2020, the price of natural gas has climbed by about 500%, while the price of electricity has increased by 390%. Coke oven gas costs are influenced by natural gas. It has consequently increased by approximately the same amount. Over the past two years, the price of an EU carbon allowance has tripled [3]. There won't be any free carbon allocation after 2027, which will increase the cost of installations [4]. Numerous production industry sectors are experiencing serious issues as a result of the sudden increase in energy prices. As a result, a number of businesses have decided to cut back on output or have stopped down. The effects of increased energy prices have been particularly detrimental to continuously operating plants. For instance, coke oven chambers cannot be allowed to fall below a certain temperature since doing so would cause irreparable

damage to the refractory walls and could even cause the unit to collapse. However, by efficiently utilising the available energy, primarily waste products and byproducts from processes, the effect of increased costs can be mitigated [5]. The functioning of process industries is made more challenging by stronger environmental requirements in addition to the ongoing increase in energy prices. Moreover, rules for preventing or at least reducing emissions are outlined in national and international accords. In order to fulfill standards, existing technologies must be updated often and run as efficiently as feasible.
The mentioned issues make it more challenging to operate coke oven plants profitably, although their main product, coke, is the fundamental raw material of the iron and steel industry. Coke is produced by high-temperature pyrolysis of appropriate quality coal blends. During pyrolysis, a large quantity of raw coke oven gas is generated, which can be considered as a by-product or as waste gas. The composition of both the coke and the gas depends on the quality of the coal blends.

Besides the continuous increase in energy prices, stricter environmental regulations make the operation of process industries more difficult. In addition, national and international agreements lay down rules to prevent or at least reduce emissions. Therefore, existing technologies need to be continuously improved and operate as effectively as possible to meet standards. The mentioned issues make it more challenging to operate coke oven plants profitably, although their main product, coke, is the fundamental raw material of the iron and steel industry. Coke is produced by high-temperature pyrolysis of appropriate quality coal blends. During pyrolysis, a large quantity of raw coke oven gas is generated, which can be considered as a by-product or as waste gas. The composition of both the coke and the gas depends on the quality of the coal blends. The most relevant quality parameters of the blends are moisture content, ash content, volatile matter content, sulphur content, and special coal quality parameters (e.g., dilatation, swelling index) [6].Properly cleaned coke oven gas is a valuable energy substitute for natural gas. From the coke oven chambers, so-called raw coke oven gas is collected. The raw coke oven gas contains water, valuable components, and impurities as well. Coal tar and light oil (benzene, toluene, xylene, and other aromatic hydrocarbons) can be merchandised after separation, while impurities such as ammonia, hydrogen sulphide, or hydrogen cyanide need to be removed due to emission requirements. The raw coke oven gas treatment is initiated with a pre cooling step, so coal tar and water condense from the gas. Then, the gas passes through electric precipitators, where fine drops of tar are removed and cooled.

This is followed by the removal of impurities in an absorption section. The final step is the light oil recovering in washing towers by special washing oil. Between 40 and 50% of the cleaned gas is recycled to heat the chambers of the coke oven battery, while the rest of it is used in the integrated steel plant as required, and the surplus is burned in gas engines. The main components of the purified coke oven gas are hydrogen and methane; thus, it has a high heating value (natural gas: 56.6 MJ/kg, coke oven gas: 41.6 MJ/kg, blast furnace gas: 2.7 MJ/kg) [7]. This is why coke oven gas, together with blast furnace gas, covers a significant part of the fuel needs of integrated steel plants, so natural gas is used only as supplementary fuel [8]. Furthermore, using properly cleaned waste gas helps reduce the natural gas consumption, which makes the whole steel production more economically and environmentally sustainable [9]. Using coke oven gas as an energy source, its composition needs to meet regulatory and technological limits. European Directives lay down rules to prevent or reduce emissions in order to reach a high level of environmental protection [10]. The Commission Implementing Decision establishes the best available techniques (BAT) conclusions one missions for industrial plants. In addition, it specifies the standards for cleaned coke oven gas. The residual hydrogen sulphide concentration must be <300–1000 mg/Nm^3(using an absorption system) or <10 mg/Nm^3 (using wet oxidative desulphurization).Furthermore, during the combustion of the coke oven gas, the emission level needs to be<200–500 mg/Nm3 for SOx and <350–650 mg/Nm3 for NOx [11].The most widely applied cleaning technology for coke oven gas is aqueous ammonia absorption. However, there is a trend for new technologies, such as Takahax, Stretford, Potassium Carbonate, or Sulfiban process, etc., which use more efficient absorbents. These processes avoid the adverse effects of trace impurities and provide higher H2S removal efficiency than ammonia scrubbing [12]. However, despite the advantages, the changeover or installing of these processes in existing plants is rather costly and often unfeasible due to continuous operation and the lack of available area in the plant.

It is more and more required to improve the existing coke oven gas purification technologies, which often means not to change the process or modernize the equipment and units, but to optimize and effectively manage the process and train competent operating staff. Implementing these alternative solutions, flow sheet simulators can be valuable tools. With the help of process simulators, the optimal mode of operation can be found adequately, and operational staff can be trained for different situations [13]. Detailed and validated process simulators can assist in aiding understanding of general and specific features of technology behavior. Different effects and experiments can be investigated without disturbing the real operation [14]. On the other hand, creating the proper simulation of this system is a rather complicated task, as coke oven gas purification represents a complex multi component separation process with numerous parallel and competitive chemical reactions.

The studies concerning coke oven gas mainly investigate the development of the technology by new solutions, column pickings, or equipment, but there are only a few studies focusing on modeling, simulation, or optimization. A German research team developed a rigorous dynamic two-phase model for a oretical description of coke oven gas purification. The basis of the model is the two-film theory. It considers several parameters such as diffusion interactions, thermodynamic non-idealities, the effect of chemical reactions on mass transfer, and the impact of structured packings and liquid distributors on hydrodynamics and electrical potential gradients. Steady-state and dynamic experiments were carried out in a pilot-scale gas scrubber, and the results were close to experimental data [15].A German research group described the chemical absorption for the system NH3-CO2-H2S-NaOH-H2O with a non-equilibrium heat and mass transfer model. The validation was performed through experimental studies. This model can extend to other reactive substances, such as mono ethanolamine (MEA) or methyl diethanolamine (MDEA). Moreover, the optimal pH range was determined for selective H2S removal [16]. Then, this group developed a rigorous rate-based model for a coke oven gas cleaning process. The multi component mass transfer of the impurities was investigated in aqueous potassium hydroxide or potash solutions. For validation, a pilot plant and industrial measurements were used. The industrial process was systematically optimized using an evolutionary strategy; thus, a 30% decrease can be reached in annual cost [17].

Another research team developed a model to evaluate solutions for H2S removal from coke oven gas. The model validation was based on industrial data, and further investigations were performed. Three process configurations were compared, and the best configuration could achieve a 5% increase in removal efficiency. Applying this configuration makes possible the use of coals with higher sulphur content [18].A Canadian research group proposed a combined cycle power plant (CCPP) in which coke oven gas can be applied as fuel. However, before the gas can be used in the CCPP, most of the H2S needs to be removed. The investigated plant could not clean the gas as much as required, so a sulphur removal system was designed and simulated with Pro Max. The CCPP was simulated in Aspen Plus and was optimized using GAMS. The optimized CCPP can generate more than twice the electrical efficiency of the existing steel refinery, and hence the purchase of electricity reduces, which contributes to a reduction in CO_2emissions [19].

However, the system is more complex in our case because the three columns are interlinked through the used water streams. The re circulating water streams also increase the complexity of the system. On the other hand, there are physical boundaries which need to be kept in mind, including seasonal changes in the cooling water temperature (that is, with the increasing changes in summer, temperature could reach 35^0C). Creating this model is challenging because it is a complex multi component separation process with several parallel chemical reactions. The task is further complicated by the lack of technical information and the limited number of measured data. However, a proper process simulator can help provide constant gas composition even under changing conditions caused by uncertain market effects (e.g., disruption of raw material supply and rising energy prices).In this study, the steady-state model of an industrial coke oven gas purification process was created. First, the model parameters (construction- and packing-related) were identified. Then, two optimization scenarios were presented, showing different optimal operation and management points with or without considering the washing water temperature.

2. Technology and Methods

2.1 Purification Process

The studied technology is a part of a Bokaro coke oven plant. The whole coke oven gas purification process can be divided into a gas cleaning section and a washing liquid regeneration section. In this work, the process model of the gas cleaning section was created and investigated, as Figure1 shows. Although not the whole technology was implemented, it is necessary to describe each section to understand the input streams.

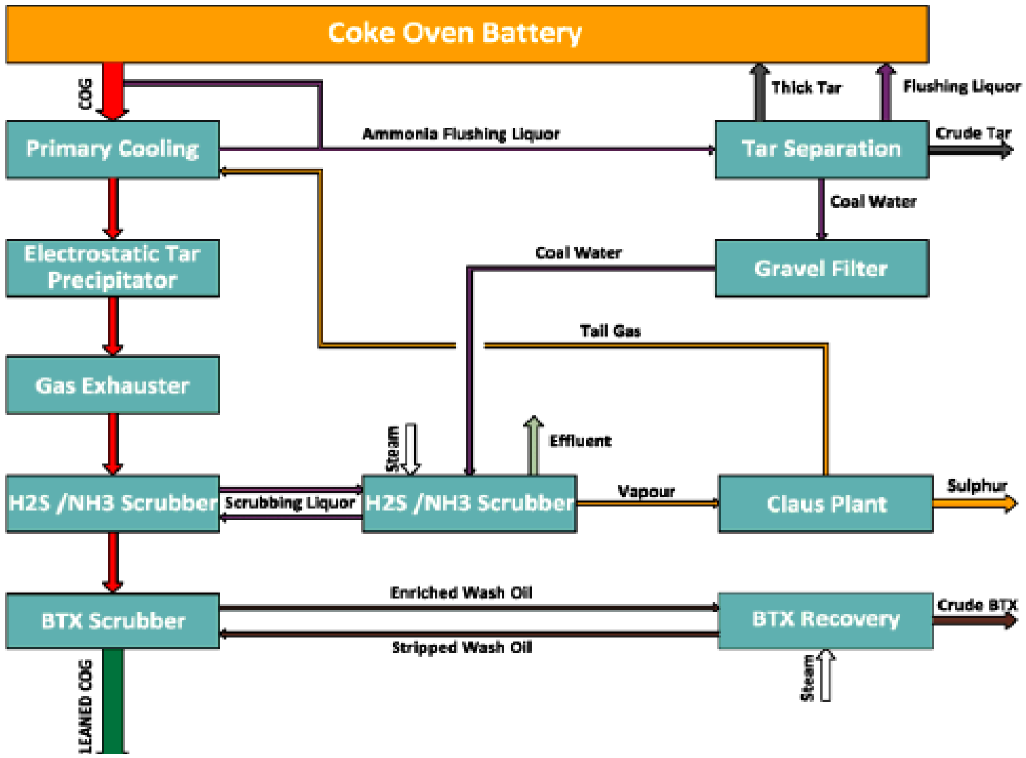

Figure 1: Overview Flow Diagram of the By-product Plant

The exhausted and cooled raw coke oven gas passes through three scrubbers, a H_2Sscrubber, a NH_3 scrubber, and a fine NH_3 scrubber as shown in Figure 2. The H_2S column is higher and narrower than the others. The construction of the towers is essentially similar, with special expanded plate inserts on supports and fluid collectors and distributors above them. The scrubbers are in a series arrangement in which the coke oven gas and the washing liquids are in counter-current flow.

Saturated washing liquid, leaving the towers, is regenerated in two steps. First, the saturated washing liquid enters the de-acidification column, where so-called de-acidified water with high ammonia content is formed as a bottom product. Next, the vapour fraction of the de-acidified tower is fed to the closed Claus technology, where catalytic transformation occurs. A certain amount of de-acidified water is fed to the H_2S scrubber, while the rest of it gets in to a stripping column. All the pollutant components absorbed in the water are removed in the stripping column. The stripped water exits at the bottom of the column and is partly fed to the second NH_3 scrubber, while the surplus is transferred to the waste water treatment plant. Finally, the head product of the stripping tower is fed back to the de-acidification column.

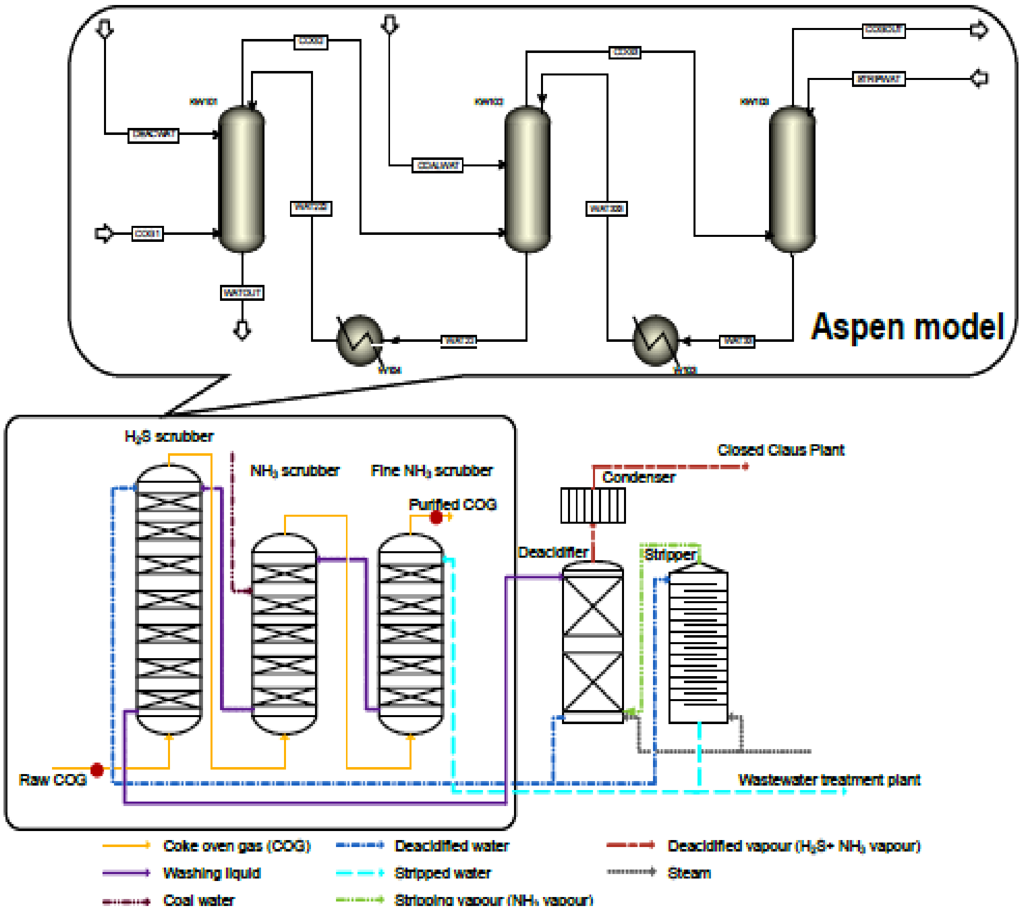

Figure 2: Coke oven gas purification process flow sheet

In the first NH_3 scrubber, an additional water input is fed. The coal water comes from the moisture content of the coal blend.

2.2 Chemical Reactions

The following parallel reversible liquid-phase reactions were considered to describe the processes during the purification.

$H_2O \leftrightarrow H^+ + OH^-$ (1)

$H_2S \leftrightarrow HS^- + H^+$ (2)

$HS^- \leftrightarrow H^+ + S^{2-}$ (3)

$HCN \leftrightarrow CN^- + H^+$ (4)

$NH_3 + H_2O \leftrightarrow NH_4^+ + OH^-$ (5)$CO_2 + OH^- \leftrightarrow HCO_3^-$ (5)

$HCO_3^- \leftrightarrow CO_3^{2-} + H^+$ (6)

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These reactions are based on simple proton transfer, except for Equations (5) and (6), which obey first- and second-order kinetics and can be regarded as instantaneous. AlthoughCO_2 is one of the major impurities in coke oven gas, its removal is not always necessary. To improve the heating value of the coke oven gas, the partial removal of $CO_2$is beneficial. However, complete elimination of CO_2 is only relevant for streams that undergo processes at very low temperatures, such as gas purification to produce pure hydrogen for ammonia synthesis.

2.3 Process modeling

This research work aimed to create the stationary model of the coke oven gas purification scrubbers. The model was built in Aspen Plus, a widely used process simulator program in the commerce and scientific fields. In addition, industrial data from a Hungarian coking plant were used to validate the model.
The investigated purification process contains a large number of electrolytes in the liquid phase, and there are strong intermolecular interactions between them. Therefore, this makes the liquid phase strongly non-ideal. To predict the behaviour of the liquid phase, the Electrolytic Non-Random Two-Liquid (ENRTL) model can be used. The Henry constants and the chemical equilibrium constants for the components were taken from the data bank of the software.

Two different approaches are available for modeling absorption: the equilibrium-stage model and the rate-based model. The first approach is based on the hypothesis that the streams leaving the stages are in equilibrium. However, equilibrium can rarely be reached in actual operations, so the usual method of dealing with departure from equilibrium is by incorporating stage efficiency. The other approach is that the two phases are balanced separately by taking into account the mass and heat fluxes across the interface. Physical properties, reaction rate parameters, and column specific data are needed, and this method avoids the approximation of efficiency entirely. The two approaches for the H_2S scrubber were compared in our previous work. It was found that the rate-based model shows a higher level of agreement with measured data than the equilibrium-stage model. Therefore, a rate-based model was used.

Columns have special expanded packings which allow proper liquid–gas contact but do not cause significant pressure drop along the column. There are four expanded packings in each column. The liquid distribution of the expanded packing is better than the random packing but worse than the structured packing. Therefore, 1m expanded packing was counted as two equilibrium stages, since this number is 1–2 for random packing and 2–4 for structured packing. Thus, the number of stages is 16 in all three scrubbers.
The data shown in Tables 1 and 2 were used to build the steady-state model of the H_2S scrubber and the two NH_3 scrubbers.

Table 1: Specificationofthethreescrubbers

	H_2SScrubber	NH_3Scrubber	FineNH_3Scrubber
Numberof stages	16	16	16
Diameter(m)	2.8	3.5	3.5
Packingtype	SHEET-PACK	SHEET-PACK	SHEET-PACK
Packingmaterial	Metal	Metal	Metal
Packingdimension	350Y	350Y	350Y
Totalheight(m)	32	28	28

Table2: Inputparameters

	CokeOvenGas	De-AcidifiedWater	CoalWater	StrippedWater
Temperature(°C)	24	24	25	27
Pressure(bar)	1.168	1.138	1.138	1.138
Flowrate(m^3/h)	46,190	65	29	22
Composition(kg/h)				
H_2O	-	63,049.43	28,887.15	21,998.6
H_2	2700.84	-	-	-
NH_3	73.47	1389.72	69.58	0.17
H_2S	341.20	73.74	0.51	0.39
CO_2	1920.63	467.97	42.32	0.64
HCN	12.21	19.14	0.43	0.19
CO	2567.05	-	-	-
CH_4	8366.47	-	-	-
C_2H_6	2034.02	-	-	-
N_2	2886.90	-	-	-

The Aspen simulator and MATLAB were connected to perform detailed studies on the model, as Figure 3 shows visually. The calculated results can be more easily handled and evaluated, and more reliable optimization algorithms can be implemented. The connection works like a real technology and a control system: Aspen Plus acts as the technology providing results, while MATLAB can be used as the control system, setting the operational parameters for the technology, as Figure3 describes.

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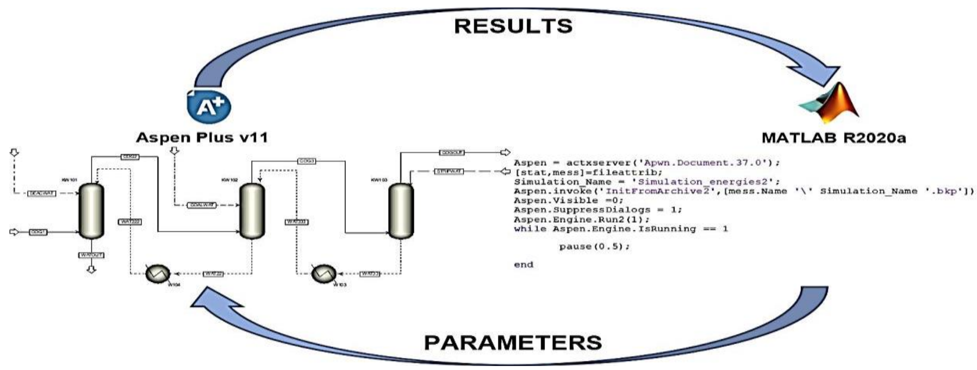

Figure3: Aspen Plus and MATLAB connection

2.4 Model Validation

The model validation was performed by using the results of industrial measurements. Samples of the input and output coke oven gas are taken twice during each shift, and an accredited laboratory analyses the samples to determine the concentrations of the three main impurities. For this purpose, samples are taken in a burette to measure the concentration of H_2S and in washing bottles to determine HCN and NH_3. The H_2S content of the gas is converted quantitatively to cadmium sulphide by cadmium acetate. Then, the cadmium sulphide is decomposed with hydrochloric acid, and H_2S is determined by iodometry. In the case of NH_3, the NH_3 in a given quantity of gas sample is quantitatively absorbed with sulphuric acid, and the excess acid is titrated back using a methyl orange indicator with a sodium hydroxide solution. While HCN content is determined by complexation with iron (II) sulphate, it is then titrated back with ferrous chloride. The error of these analytical measurements is less than 5%. The average measured results and the model results for the three main impurities are summarized in Table3. The relative errors between the measured and calculated results are less than 1% for all three components.

Table3: Measuredandcalculatedresultsofthethreemainimpuritiesintheoutputcokeovengas

	MeasuredResults	**CalculatedResults**	**RelativeError**
	(kg/h)	**(kg/h)**	**(%)**
NH_3	1.39	1.39	0.00
H_2S	37.87	37.98	0.29
HCN	1.75	1.76	0.57

3. Optimization

Based on the results of the sensitivity analyses, optimization tasks were performed. For this purpose, the fmincon function of MATLAB was applied, which is a non linear interior point optimization technique.I n this work, two optimisation tasks were investigated.One was to minimise theH_2S and NH_3 content in the output coke oven gas. To achieve this, the quantity andtemperature of the washing liquids varied within the ranges presented in the sensitivitystudies.Sincethesensitivitystudyfoundthatthetemperatureofthestrippingwaterhadlittle effect on the composition of the output gas, this was not taken into account when setting the variables. The other optimisation task was to ensure that the gas was cleaned with as little energy consumption as possible while complying with the required emission standards. Thus, the temperature of the washing liquids was kept as high as technically feasible to reduce the cooling water demand. The amount of washing liquids was not intended to be minimised, as the pumping energy was not too significant. The maximum amount of H_2S in the output gasis1mg/Nm, while the maximum amount of NH_3 is0.5mg/Nm^3.

4. Results and Discussion

The steady-state model of the coke oven gas purification technology was built as described which shows the mass flow profiles of H_2S, NH_3, and HCN in the gas phase, along with the scrubbers. Optimized operating collects the results calculated to minimize the impurities contentin the output coke oven gas. It can be observed that the temperature of the washing liquids needs to be relatively low, while the flow rate of the water streams needs to be high.

5. Conclusions

This study developed a steady-state model of the coke oven gas purification procedure found in a coke oven factory in Hungary. Given that the three scrubbers are connected by washing liquids, unique expanded packings are used in the columns, and coke oven gas is a multi component gas, the task is very complicated and requires consideration of a number of simultaneous and competitive chemical reactions. The rate-based methodology was used to build the model in Aspen Plus. The fmincon function of MATLAB was used to adjust the liquid mass transfer coefficient factors of the three columns. The relative errors between the measured and estimated impurity levels in the output coke oven gas were therefore less than 1%.

With the adequately identified model, sensitivity analyses were carried out to study the effects of the parameters that can and cannot be influenced. The influence able parameters consist of the flow rate and temperature of de-acidified water, coal water, and stripped water. Based on the results of sensitivity analyses, two optimization tasks were completed with the help of the fmincon function of MATLAB. One part of the optimization was to see whether the current system would still be able to guarantee the required purity of coke oven gas if environmental standards were stricter. The findings indicate that in this situation, far lower temperatures and more cleaning liquid would be required. The facility would have to use more energy as a result, though. With current technology, the output coke oven gas can have an H2S concentration of 0.54 g/Nm3 and an NH3 concentration of 0.01 g/Nm3. This indicates that the technology can also adhere to far higher criteria when used in the appropriate manner.

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