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Research Article

Energy and Environmental Optimization of a Bituminous Waterproofing Plant for BTEX Emission Mitigation using Incinerator and Oxidation Method

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Abstract

Aromatic hydrocarbons such as benzene, toluene, ethylbenzene, and xylene isomers (BTEX) are hazardous air pollutants from industrial processes which have raised many concerns recently. In particular, Bituminous waterproofing (BW) plants are one of the major sources of BTEX compounds. Elimination of these toxic compounds, even in small amounts requires careful attention to modify and improve process performance. In this study, the process of removing BTEX contaminants from a BW production plant has been accomplished by simulation using Aspen Hysys software. In this regard, the process modification based on the Incinerator designing and using the thermal oxidation method has been evaluated. The optimization study involves the analysis of key variables such as temperature and residence time for optimization. According to the results, the incinerator's control efficiency (CF) in removing BTEX pollutants is reported to be 98.5%. Moreover, the energy consumption index has reduced fuel consumption by 23% compared to the system's performance without an incinerator in the same CF.

Keywords: Air Pollution, Aromatic Hydrocarbons, Optimization, Incinerator, BTEX

Introduction

Nowadays, the rapid industrialization of societies, especially developing countries, to meet the needs of citizens is recognized as the main cause of air pollution. Therefore, maintaining environmental standards is necessary to reduce the potential for pollutions and support sustainable development (Su et al., 2022; Syah et al 2021). Recent evolution in environmental regulations regarding the release of aromatic compounds such as benzene, toluene, ethylbenzene and xylene isomers (BTEX) and other volatile organic compounds (VOCs) have made these emissions a major concern in communities. BTEX compounds (formula type C_NH_{2N-6}) are volatile and relatively soluble in water. They pose a serious threat to human health by causing various diseases; hence, identifying contaminants and controlling the emission of these compounds is gaining importance in industrial plants (Zahed et al., 2010; Davidson et al., 2021; Mohajeri et al., 2022).

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Various strategies are used to control the amount of BTEX emissions in different industries. Newly, methods based on chemical degradation, including oxidation, have attracted much attention (Atamaleki et al., 2022; Zhu et al., 2021; Wang et al., 2022; Yu et al., 2021; Atamaleki et al., 2021). Modification of industrial processes through incinerator design and the use of thermal oxidation can result in a significant reduction in BTEX emissions, as shown by Tomatis et al. (2019). However, this technique requires careful design for proper operation. In order to improve the removal efficiency of volatile organic compounds, different parameters on the incinerator design are evaluated (Zheng et al., 2022; Chang et al 2022). The goal of the work reported in Liang et al., illustrated that the rate of removal of volatile organic compounds in the oxidation method is a function of various operational parameters such as the type and concentration of compounds, residence time, and temperature (Liang et al., 2009).

It should also be noted that there have been a number of interesting studies focused on controlling the emission of BTEX compounds using incinerator design on industrial scales. Emphasizing the importance of the need of industries in the United States to control the emission of pollutants, Tessitore et al. (2020) concluded that incinerator design is a relatively simple process with minimal operating costs. Kim et al. (2020) assessed the economic and environmental practices available to reduce BTEX emissions from a petrochemical plant in China. The results indicated that the application of the oxidation method based on the formation of the Incinerator has had a more favorable performance. In order to control the direct and indirect emission of BTEX compounds in the coal power generation process, Peng et al. (2021) reduced the emission of pollutants from the chimneys by about 70% by designing an incinerator and oxidation method.

Petroleum derivatives used in the industry contain large amounts of BTEX compounds that are often widely used due to their very low cost and irreplaceable. Few researches has assessed the BW plants; thus, the amount of BTEX emissions in these industries has been ignored, and its effects on health have been underestimated. The incinerator is expected to reduce the emissions of BTEX compounds significantly but, there is little evidence showing that it is more practical than burning exhaust gases regarding energy efficiency. Questions related to the efficiency and environmental benefits of designing incinerator serve as the primary motivation for the present work. The purpose of this study is to help this evidence by assessing the environmental and technical evaluation of the performance of the incinerator unit in a BW production plant located in Delijan-Iran. In the current study, the optimization of BTEX emissions is based on the design of the incinerator by means of thermal oxidation method. Contrary to the previous studies, where control measures are generally based on the emission modeling (Partha et al., 2022; Ahmadi Givi et al., 2022; Allahabady et al., 2022; Liu et al., 2022), in this plan, system modification and operational strategies have been considered the two main factors to improve oxidation conditions. To the best of our knowledge, based on the previous investigations, this is the first such Iran-based effort of its kind.

Materials and Methods

Process Description

BW production units generally consist of two subsystems primer production unit and cooling unit as shown in Figure 1. In this process, bitumen and other modifiers (usually straw and talc powder) enter the furnace and are heated at very high temperature. Enriched bitumen flow enters the mixer through the transfer tubes to mix with Atactic Polypropylene (APP) and Styrene Butadiene Styrene (SBS). Exhaust gases from this stage are considered the main emission factor of BTEX compounds.

Finally, the materials flow to the cooling unit, and after adding aluminum coating, they are ready for final packaging and storage.

The specifications of the equipment used in the case study unit are listed in Table 1.

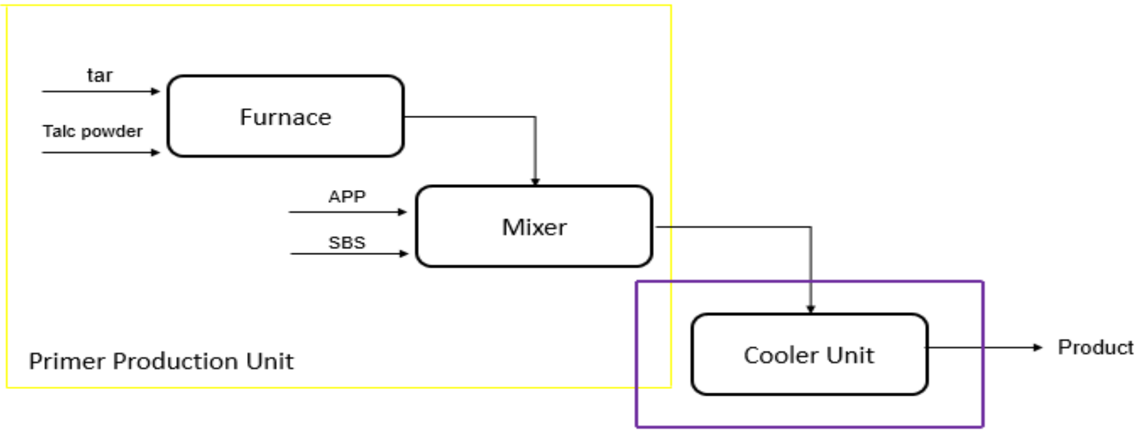

Figure 1. Schematic of BW production process in the case study

Table 1. Type and specifications of the equipment used in the BW unit

Equipment	Capacity (m^3)	Heating Element (Kcal)	Dynamo (hp)	Gearbox (pm)	Flor sheet (mm)	Side sheets (mm)	Top sheets (mm)
Furnace	12	550000	20	400	10 (st 37)[*]	10 (st 37)	15 (st 37)
Mixer	25	-	20	400	10 (st 37)	10 (st 37)	15 (st 37)

[*] *St 37: Steel plate sheet having maximum 0.17 % carbon ratio.*

Process Optimization

Different strategies can be hired to remove volatile organic compounds; however, these methods may not be in line with internationally approved trends. As explained in the introductory sections, Thermal Oxidation was selected for this study to minimize BTEX compounds by using an incinerator in the primer production subsystem. Thermal oxidation has lower operating costs and complexities compared to catalytic oxidation, and it is relatively simple. However, it also has disadvantages because it is less stable at high temperatures.

Modification of process and changes in the operating conditions in order to optimization requires precise simulation. In the present study, simulation of BW production unit has been accomplished using the Aspen Hysys software. Considering the process as steady-state and BTEX compounds as the main pollutants of exhaust gases are the two main assumptions of this simulation. The results are comparable to the actual results and the error is negligible with these assumptions. Moreover, it is feasible to predict the behavior of the system in varied operating conditions with the software. The design parameters in this study include the following:

- Exhaust gas compounds
- Operational conditions
- The amount of oxygen in the exhaust gas

Exhaust gas compounds

The chemical composition of the flow gas entering the incinerator is one of the determining parameters in this design. In this regard, the gas stream derived from the primer production section was sampled, and then the type and concentration of benzene, xylene, toluene, and ethylbenzene (BTEX) were determined.

- Sampling Technique

To identify the exhaust gas compounds, emitted pollutants were sampled using Charcoal absorber tubes. By determining the high concentration of environmental pollutants, the volume of air collected for sampling was such that the adsorbent capacity was not saturated. In this work, sampling pump flow, air sample volume, and measurement time are determined according to environmental conditions (Thom et al., 2009). The pump flow in the measurements is defined $1m^3/min$, and the sampling time is determined between 10-15 minutes. The inlet of the absorber tube was free during the measurement and was away from the surrounding obstacles.

Low flow rates, high temperatures, and humidity can reduce the ability of the substrate to absorb the material, and contamination may also be transmitted from the front of the pipe to other parts; Therefore, keeping the pipe cool reduces this possibility.

Finally, using plastic lids for samples, sealed in a cool place and sent for final analysis due to the possibility of light decomposition away from sunlight.

- Gas Analysis

It is emphasized that the type and concentration of BTEX compounds have been determined using GC-FID model GC-2010 SHIMADZU.

The analysis process in this section includes the steps of sample preparation, calibration and quality control, and measurement. In the preparation stage, the sample is placed in the vicinity of ambient temperature. The moisture test is performed to ensure that the moisture content of the sample is appropriate. Then desorption operations are performed on the sample. In the calibration and quality control stage, the mass spectrometer device has been set up according to the operating conditions defined by the manufacturer.

The results of the analysis of BTEX compounds of exhaust gas by GC-FID before installing the incinerator are presented in Table 2.

By examining the concentration of pollutants before installing the incinerator, it was found that the measured concentration is much higher than the standard limit for benzene, toluene, ethylbenzene and xylene (Salama et al., 2021; Baimatova et al., 2016).

Operational conditions

Complete oxidation of materials is strongly dependent on residence time and temperature. Following an approach similar to those reported in the literature on the designing of incinerators, determining the appropriate range for the operating conditions of the incinerator is defined based on the concentration (Table 2) and the ratio of BTEX compounds (B/T). In our effort, a number of different behavioral incinerator designs with similar condition were reviewed from previous studies. Accordingly, the temperature and residence time are defined in the range between 700 to 750 ° C and 0.75 to 1 second for the incinerator in order to maximize the degradation of the BTEX pollutants. Therefore, in this research, design indicators based on these conditions have been applied in the proposed software (Sorrels et al., 2017; van der Vaart et al., 1996).

Table 2. The specifications of the studied unit before optimization

Parameter	Unit	Level
CO	ppm	22
SO_2	ppm	4
O_2	%	7.2
Benzene	ppm	89
Toluene	ppm	106
NO_2	ppm	1
NO	ppm	5
NOx	ppm	5.1
H2S	Ppm	5
CO_2	%	18
Ethyl Benzene	ppm	11
Xylene	ppm	176
Temperature	℃	103
Velocity	m/s	6.1
Chimney diameter	m	35

The amount of oxygen in the exhaust gas

However, as discussed in Haridass et al. (2017), the amount of oxygen expected to play a significant role in determining the performance of thermal oxidation. If the amount of oxygen in the pollutant gas is more than 20%, the oxidation process is complete; otherwise, the excess air to burn complete volatile organic compounds will be required. According to the following equations, the amount of excess oxygen in the exhaust gases from the primer section is calculated (Haridass et al., 2017). It is emphasized that the concentration of compounds is calculated (ppm), conforming to the information in Table 2.

$$ppm = \frac{\frac{M_P}{GMW} \times 22.414 \times \frac{T_2}{237\,(K)} \times \frac{101.325\,(KPa)}{P_2}}{V_a \times 1000\,(\frac{L}{m^3})} \tag{1}$$

$$\text{Air\% Content} = 100 - \sum((X_n(ppm)/10^6)*100) \tag{2}$$

$$\text{Oxygen\% Content} = \text{Air\% Content} * 0.209 \tag{3}$$

In these equations, GMW the weight (g) of a mole of molecules of a compound, Mp concentration (µg), Va volume (m^3) at ambient temperature and pressure, T_2 ambient temperature 25 ° C, P_2 ambient pressure 101.93 kPa and Xn concentration of pollutant (ppm). Regarding the required air associated with the oxidation to remove BTEX compounds, the percentage of air and oxygen in the exhaust gas have been calculated to be 11.88 and 41.18%, respectively, so the need for a separate inlet for additional air in the simulation should be considered.

Incinerator specifications

Considering the design parameters mentioned in the above sections, finally the incinerator with the following specifications was finalized for operation. Table 3 illustrate in details the final specification of designed incinerator.

Table 3. Final specifications of the designed incinerator

Parameter	Description
Capacity	10 m^3
Diameter	2 m
Length	2 m
Top sheet	15 mm （st37）
Side sheets	15 mm （st37）
Floor sheet	15 mm （st37）
Dynamo	20 hp
Heating Power	650000 KCal
Total Flow rate	3200 Scfm

Results and Discussion

Influence of Process Optimization

Optimization is a preventive method to reduce the release of pollutants into the system. Changes in performance parameters lead to significant changes in volatile organic compounds in the exhaust gases. Increasing the temperature of the incinerator as well as the residence time reduces the emission of BTEX compounds. Due to the significant thermal potential of the incinerator, temperature comprises the largest share in this system, while residence time is the second essential factor of the scheme proposed for the process. Other factors such as feed gas temperature and feed gas content also have a significant effect on the release of BTEX compounds in this system. However, most operators do not have access to the production process variables, so the feed gas temperature and its contents are not considered as control tools in this optimization.

Environmental Assessment

In this paper, the utilization of an incinerator led to the improvement of environmental indicators. Based on the current simulation, the emission rate of BTEX compounds from the exhaust gas in different operating conditions is in accordance with Figures 2, 3, 4 and 5. The results will be discussed in next sections. The oxidation equation in this system is as follows (Nabi et al., 2020):

$$C_xH_yO_z + \left[x + \frac{y}{4} - \frac{z}{2}\right]O_2 \Rightarrow xCO_2 + \frac{y}{2}\mathrm{H}_2\mathrm{O} \tag{4}$$

The figures 2 to 5 show the Benzene, Toluene, Ethylbenzene, and xylene in the various operating conditions derived from simulation. It can be seen that the concentration of pollutants during the constant residence time after reaching the minimum value, continued its path with a constant slope so that the rate of change of pollutants to the final temperature is close to zero. It is also observed that there is an indirect relationship between the residence time and BTEX concentration. With increasing residence time (at similar temperatures), the decrease in the concentration of pollutants has increased to some extent. In general, retention time has less effect on the removal process compared to the oxidation reaction temperature. Moreover, benzene has the lowest concentration among BTEX pollutants. After benzene, toluene, ethyl benzene and xylene have the lowest concentrations in the exhaust gas, respectively.

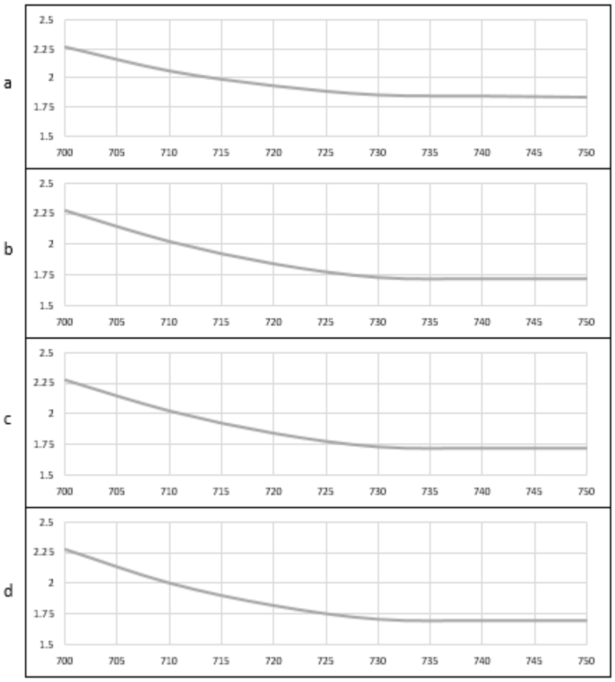

Figure 2. Toluene concentration (ppm) vs temperature at residence times a) 0.75 seconds, b) 0.85 seconds, c) 0.95 seconds and d) 1 second

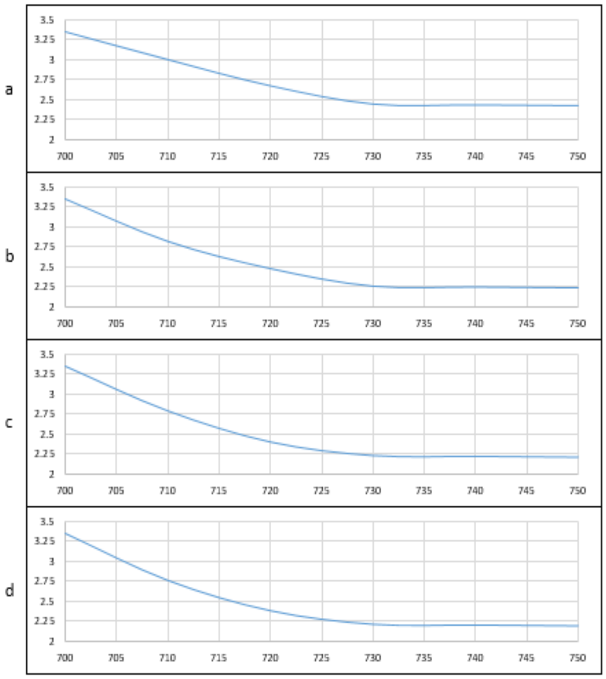

Figure 3. Toluene concentration (ppm) vs temperature at residence times a) 0.75 seconds, b) 0.85 seconds, c) 0.95 seconds and d) 1 second

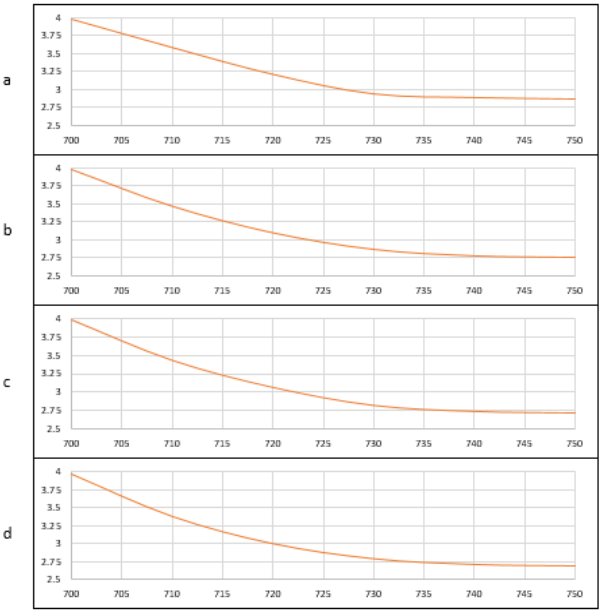

Figure 4. Xylene concentration (ppm) vs temperature at residence times a) 0.75 seconds, b) 0.85 seconds, c) 0.95 seconds and d) 1 second

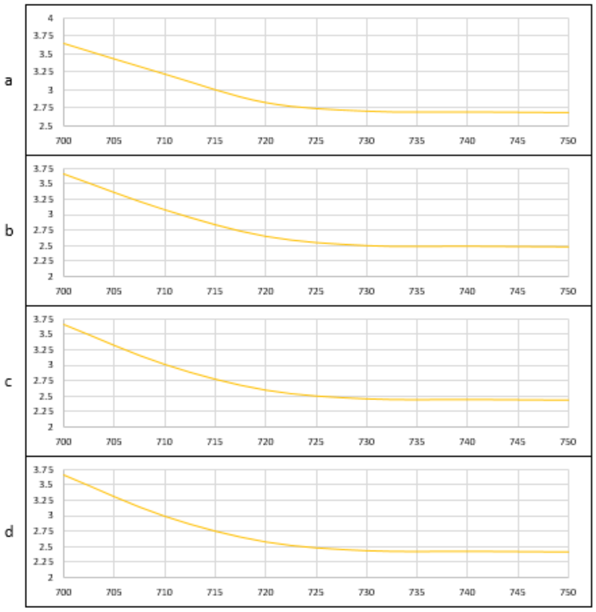

Figure 5. Ethylbenzene concentration (ppm) vs temperature at residence times a) 0.75 seconds, b) 0.85 seconds, c) 0.95 seconds and d) 1 second

The control efficiency (CE) of the incinerator designed to remove BTEX compounds is defined according to Equation 5 (Choi et al., 2018).

$$CF = [C_{inlet} - C_{outlet} / C_{inlet}]*100 \qquad (5)$$

In this regard, C_{inlet} and C_{outlet} are the sum of the concentrations of BTEX compounds when entering and leaving the incinerator, respectively. After optimizing the system, the control efficiency of the incinerator will be calculated.

Energy Assessment

Following an approach similar to those reported in the literature on designing incinerators (Zheng et al., 2022; Michos et al., 2020) energy balance equations are defined based on presented relations.

$$\sum_{i=react}^{n} (m_{in.i}) = \sum_{i=prod}^{n} (m_{out.i}) \qquad (6)$$

$$m = \rho .\, q \qquad (7)$$

In above equation, ρ is the density and q is the rate of volumetric flow. In addition, according to the heat load of the input and output currents to the incinerator, the energy balance of the currents is derived using Equation 8.

$$\sum_{i=prod}^{n} (H_{fi}) = \sum_{i=prod}^{n} ni\,(H_{fi} + \Delta H_{Ti}) \qquad (8)$$

Where $H_{f,\,i}$ is the enthalpy of formation of reactants and products. The enthalpy balance inside the incinerator can be expressed by Equation 4. The total enthalpy content in any chemical is equal to the sum of the chemical, and tangible enthalpies is expressed according to Equation 9:

$$H(f.CxHyZo) + \left(x + \frac{y}{4} - \frac{z}{2}\right)\left(H(f.O2)\right) =$$
$$y/2(H(f.H2O) + \int_{298}^{T} Cp.H2OdT) + x(H(f.CO2) + \int_{298}^{T} Cp.CO2dT) \qquad (9)$$

However, in this case the total amount of enthalpy difference should be calculated. Hence:

$$\Delta h = \sum_{i=1}^{n} Xi\,(\Delta hi) \qquad (10)$$

$$Q_{tot} = \sum_{i=1}^{n} mi\,(\Delta hi) = \sum_{i=1}^{n} \rho i\; qi\,(\Delta hi) \qquad (11)$$

$$Q_{tot} = \int_{0}^{T} m_f C(\frac{dT}{dt})dt \qquad (12)$$

In this regard, X_i is the volume fraction of compound *i*, Δhi is the heat combustion of compound *i* and Δh is the total heat combustion needed for system, m_f the amount of fuel used to perform the process, *dT/dt* temperature changes in terms of reaction time and *C* is the specific heat of the fuel at constant pressure.

Due to airflow in the incinerator, the amount of consumed fuel (EG_2) is reduced compared to the direct combustion mode of the gas flow without the incinerator (EG_1). The rate of reduction of energy consumed in the same result is according to Equation 13.

$$\frac{EG1 - EG2}{EG1} = \frac{(\Delta H\ O2)}{(\Delta H\ total)} \tag{13}$$

Figure 6 provided a graph in which the effect of the incinerator on energy consumption was concluded.

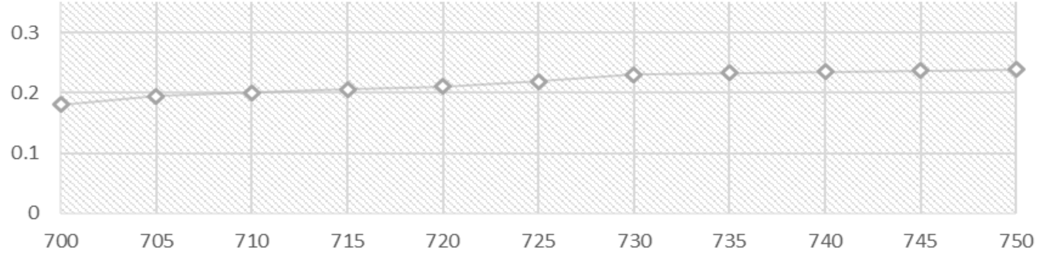

Figure 6. The amount of energy consumption reduction in case study before and after using the incinerator

Statistical analysis

Lambda control and adjustment is one of the most effective solutions to increase fuel efficiency and reduce pollution in thermal equipment such as incinerators. The lambda coefficient is the ratio of fuel to air, and the complete combustion of the burner installed in the incinerator depends on it. In this study, the fuel used by the incinerator is methane, and the total gas flow rate is 3200 scfm in the oxidation reaction. According to previous studies, for one unit of fuel and 17.67 units of air, the lambda coefficient equals one ($\lambda = 1$).

The combustion equation is generally obtained from below equations (Michos et al., 2020):

$$CxHy + A_1\ (O2 + 3.76\ N2) \rightarrow A_2CO2 + A_3\ H2O + 3.76N2 \tag{14}$$

$$O2 + 3.76\ N2 \equiv 4/76\ Air \tag{15}$$

$$A1 = A2 + A3/2 \tag{16}$$

$$\lambda = \frac{m\ (fuel)}{m\ (air)} \tag{17}$$

The proper amount of combustion coefficients related to the fuel consumed by the incinerator provided, as demonstrated in Table 4.

The relationship between fuel consumption by incinerator and oxidation temperature was investigated in the previous sections. The system was validated during operating hours and in different operating conditions using simulation information based on the statistical analysis. Based on the results, in the case where the highest control efficiency of the incinerator (CF) has been calculated, the amount of oxygen measured, air consumption, and λ according to the mentioned relations were calculated to 0.61, 2.9, and 1.14 respectively. As shown in Figure 7, due to the trend of changes in control efficiency (CF) with lambda coefficient, the CF rate first has an upward trend. After reaching a certain limit of lambda coefficient reached its maximum and then without significant change (approximate slope of zero), it remains constant at a certain value. By comparing the measured parameters using the insulator, the energy-saving rate was 23% compared to similar conditions, and the absence of the incinerator was measured.

Table 4. The rate of combustion coefficients related to the fuel consumed by the incinerator

Parameter	A_1	A_2	A_3
Amount	2	1	4

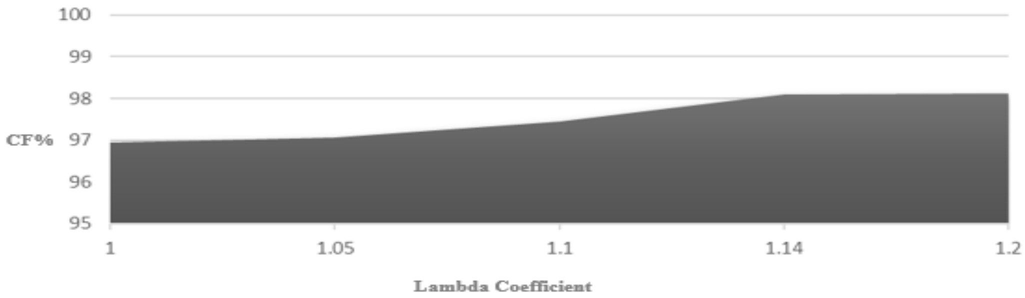

Figure 7. Incinerator control efficiency based on different values of lambda coefficient

As can be observed, Lambda with a value of 1.14, has the minimum amount of emissions, while in higher amounts ($\lambda > 1.14$), more energy consumption is imposed on the system despite the same volume of pollutants. Moreover, it can be seen that utilization of Lambda adjustment in simulation compared to exclusive operational regulation of the system can reduce the volume of emissions of the incinerator and the whole system.

The application of the incinerator not only offers energy saving but also restricts the amount of emissions. This section evaluated the amount of pollutants produced by the proposed system. Considering the parameters mentioned in the previous sectors and operating conditions of the system, the final specifications of the incinerator and its optimal operating conditions are reported according to Table 5. Furthermore, by analyzing the exhaust gases from the incinerator, the amount of removal BTEX compounds are presented in Table 6.

Table 5. Description of optimized parameters in the system

Parameter	Rate
Control Efficiency (CF)	98.09
Residence Time	0.84 s
Temperature	732 ℃
Required Oxygen	0.61
Required Air	2.9
Lambda Index	1.14

Table 6. The amount of BTEX compounds in the optimized system

Compound	Removal (ppm)	Remaining (ppm)
Benzene	87.26	1.74
Toluene	103.77	2.23
Ethylbenzene	108.52	2.48
Xylene	173.26	2.74

The results indicated that this novel design can remove BTEX compounds equally. This is due to the proximity of the boiling point of the studied compounds and the optimal operating temperature of the incinerator. Moreover, the optimization resulted in a more than 90% reduction in BTEX compounds compared to the normal system. Moreover, Table 6 demonstrated the volume of BTEX compounds produced by the system after optimization. Xylenes are the most important indicator of gasoline and additive compounds such as APP in industry. On the other hand, gasoline has been the only fuel used in the studied plant therefore, as expected, Xylene has the maximum reduction among other BTEX compounds (Table 6).

Conclusion

Implementation of the oxidation method with incinerator design is one of the effective methods in reducing BTEX emissions of industrial units. The case study presented in this work is based on the computer simulation of the BW production unit in Delijan-Iran. Based on the parametric study mentioned, the significant parameters chosen for the optimization study were the incinerator temperature and residence time. Optimization of BW production unit parameters resulted in a 98.5% overall reduction of BTEX and a 23% overall reduction in fuel consumption from the incinerator.

Further researches can be conducted using different types of modules in order to meet emission standards. Moreover, the feasibility of coupling renewable energies to incinerators can also be assessed. Overall, Thermal oxidation using incinerator has lower operating costs and complexities that can be harnessed to reduce BTEX emissions in BW industry.

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