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Title : PHOTOCATALYTIC DEGRADATION OF BENZENE-TOLUENE GASEOUS MIXTURE USING N, Fe-TiO₂ PHOTOCATALYST UNDER VISIBLE LIGHT: RESPONSE SURFACE METHODOLOGY (RSM) AND ARTIFICIAL NEURAL NETWORK (ANN) MODELLING
Supervisor : ASSOC. PROF. DR. JUNAIDAH JAI (MS)
            ASSOC. PROF. DR. JAGANNATHAN KRISHNAN (CS)

Volatile organic compounds, VOCs such as benzene and toluene are hazardous to human health even when exposed at low concentration due to their carcinogenic impact. Recently, photocatalytic degradation has received great attention from researchers as one of the promising method to lower VOC concentration in the air. Common photocatalyst used for this process is titanium dioxide, TiO₂. However, TiO₂ can only be activated under UV light range due to the wide band gap energy. Limited resources are the challenge faced in implementing photocatalytic degradation. This is because only 3% of the sunlight wavelength range is of UV light while the other 45% is of visible light. The conventional method, also known as the ‘one-factor-at-a-time’ is a common approach taken in photocatalytic degradation. This approach is usually difficult and the interaction between process parameters is complicated to interpret as it requires numerous experiments. Besides, the complexity of photocatalytic degradation also lies in predicting the removal efficiency of the pollutants. Hence, this study is attempted to modify the TiO₂ photocatalyst and to activate it under visible light wavelength range. The modified photocatalyst was applied in the photocatalytic degradation of individual benzene or toluene, as well as their gaseous mixture under irradiation of visible light. In this research, TiO₂ was modified by co-doping with nitrogen and iron elements, prepared using sol-gel method in order to activate the photocatalyst in visible light wavelength. The modified photocatalyst was characterized using XRD, BET surface area, UV-Vis DRS, and XPS. The synthesized N, Fe-TiO₂ was applied in the photocatalytic degradation, and tested with individual benzene or toluene, and benzene-toluene gaseous mixture under irradiation of visible light. To reduce the number of experiments, as well as to optimize the degradation process, response surface methodology (RSM) was applied for each photocatalytic degradation cases. Artificial neural network (ANN) models were also developed to predict the complexity of the photocatalytic degradation process under visible light. It was found that N, Fe-TiO₂ photocatalyst is an anatase phase with crystal size of 19 nm and higher active surface sites. In addition, the energy band gap of N, Fe-TiO₂ photocatalyst is 2.33 eV, representing its ability to be activated under visible light wavelength region. RSM models have been developed to predict the removal efficiency of benzene, toluene and benzene-toluene gaseous mixture in order to optimize the responses. It was found that all independent variables that consisted of pollutant concentration, flow rate, and photocatalyst loading played a significant role in the degradation process which was evaluated based on the ANOVA, main effect, and variable interaction analysis. The optimum condition based on the RSM model of benzene-toluene gaseous mixture was 20 ppm (benzene concentration), 10 ppm (toluene concentration), 0.8 L/min (flow rate), and 25.54 (photocatalyst loading) which have successfully degraded 87.66% (desirable: 87.28%) and 90.83% (desirable: 89.95%) of benzene and toluene, respectively. Finally, the ANN models for individual benzene or toluene gas was 3-15-1, and benzene-toluene gaseous mixture was 4-15-2. The models were proven to have a good fit with high determinant coefficient, R² value for all cases, which is near to 1. The developed ANN models were good predictors for the application of the degradation process. The objectives of this research were successfully achieved, thereby marked the applicability of photocatalytic degradation of benzene-toluene gaseous mixture using N, Fe-TiO₂ photocatalyst under visible light irradiation. models (statistical and NARX model) predicted equally good performance ranging from 70% - 90% accuracy. Further analysis is required to test the developed model specifically for different river characteristics. However, the availability of data is a hindrance and to draw these findings, further recommendations are summarized for the validity of the derived equations.

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Title : PYROLYTIC OIL SYNTHESIS VIA CATALYTIC MICROWAVE ASSISTED PYROLYSIS OF AUTOMOTIVE PAINT SLUDGE BY USING ZSM-5 CATALYST
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Pyrolytic oil produced from automotive paint sludge (APS) through microwave assisted pyrolysis process has a great potential to be used as an alternative fuel. Calorific value of APS pyrolytic oil achieved almost 35MJ/kg and enriched with monoaromatic hydrocarbon which is not achievable in previous study by past researcher’s due to the volatile released during drying process which is required by the pyrolysis equipment. However, high oxygenated hydrocarbon compound in the APS pyrolytic oil makes the pyrolytic oil still not achieve the standard requirement of commercial fuel, which the density and viscosity of APS pyrolytic oil is far from the standard due to high oxygenated hydrocarbon. Optimization is done on the process but only permits slightly increased on the pyrolytic oil yield. In addition, it reduced the quality of pyrolytic oil in terms of physical and chemical properties. Oxygenated hydrocarbon was increased to 53.1% of percentage area concentration. This is due to high radiation time need in the optimization method which the formation of more oxygenated and polycyclic aromatic compound. In order to meet the requirement of standard commercial fuel, improvement has been made by introducing catalyst ZSM-5 during the microwave pyrolysis process. As a result, calorific value of APS pyrolytic oil increased to 41 – 46 MJ/kg, which is higher than non-catalytic APS pyrolytic oil. This is due to the increased of hydrocarbon breakdown during the released of volatile compound and form high monoaromatic hydrocarbon and reduction of high oxygenated hydrocarbon compound. Catalytic microwave assisted pyrolysis of automotive paint sludge has been optimize in this research by using RSM-CCD method. It was found that the pyrolytic oil yield was increased to 0.875% by using optimized parameter of 1.24% catalyst mixing ratio, 200g of sample weight loading, 873.5W of microwave power level and 49.9 minutes of radiation time. Moreover, optimized physical properties were determined at 44.7 MJ/kg of calorific value, 4.71 cP of viscosity and 837 kg/m³ of density. Optimized method achieved its target by having low catalyst loading and microwave power level used in the process with the increased yield of pyrolytic oil and its properties which lead to the efficient operating condition at optimum output. Properties of APS pyrolytic oil is improved significantly with the presence of catalyst ZSM-5 and it became more feasible to be used as a commercial fuel.