The Use of Sensitivity Analysis and Genetic Algorithms for the Management of Catalyst Emissions from Oil Refineries

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Abstract: Excessive catalyst emissions from Fluidized Catalytic Cracking Units (FCCU) during start up situations are common, and have been deemed 'normal' with little research conducted on determining their causes. A simplistic model found to predict trends in emission rates under normal conditions has been expanded to better represent the actual processes inside an FCCU. First and second order sensitivity analysis techniques are used to assess the interactions between various operational parameters, with a genetic algorithm used to optimize the operating conditions to minimise air emissions. These 'key' parameters may then be altered to help manage both normal and start up emissions through operational changes. It was also found that significant scale up issues arise with the use of the attrition models found in the literature.

Keywords: *Sensitivity analysis; Genetic Algorithm; Air pollution; Modelling*

1. INTRODUCTION

The petroleum industry currently employs Fluidizing Catalytic Cracking Units (FCCU's) as the major tool in producing gasoline from crude oil. FCCU's typically consist of a rising main where the chemical reactions between catalyst and hydrocarbons occur, a reactor to separate the product and catalyst, and a regenerator to re-charge the used catalyst. The regenerator is a fluidized bed used to combust coke from the used catalyst, with cyclones to remove particles from the flue gas stream before venting to the atmosphere. The recharged catalyst then re-circulates through the rising main and the process is repeated (Kunii and Levenspiel 1991).

In recent years, fine particle emissions from industry have been identified as important contributors to poor environmental and health standards across the United States (Johnson 2001). With increasing demands for cleaner air, catalyst emissions from FCCU's have the potential to impact significantly on the environmental efficiency of the overall refining operation (Rucker and Strieter 1992). Currently, FCCU's are designed and operated in such a way as to maximise output and profitability of the refinery (Lin 1993). With the lack of literature dealing with FCCU emissions, there is a need for the relationships between current operational strategies and air pollution to be better understood.

2. METHODOLOGY

MATLAB was used to extend a simplistic model, already developed and tested on FCCU emissions (Whitcombe et al. 2001 and 2002).
The model comprised the essential processes inside the FCCU including fluidization, elutriation, entrainment and attrition, with model equations sourced from the literature.

The aim of this paper is to further develop the model into a more realistic package, for use in modelling industrial emissions. This will provide a simulation package of the FCCU, where the simulated emission can be studied in terms of system parameters. Emission results are then compared with observations from an operating FCCU, where the FCCU particle emissions during a start up period were identified. Thus allowing the complete range of emission to be seen (Agranovski and Whitcombe 2002).

2.1. Model Background

The original model (refer to (Whitcombe et al. 2001) is of the form y = f(x) where x is a vector of 11 input parameters, and these are listed in Table 1. The output of the model, y is the emission level in mg/m^3 of catalyst particles from the cyclones, and the function f(x) is essentially algebraic. The model accepts the input parameters, and steps through a series of subroutines to calculate specific process outcomes in the regenerator. The non linear equation representing each process, uses a combination of operating parameters and the solution of previous routines, to implement its particular process. This model was tested extensively using published examples as well as sensitivity analysis and was found to be approximately 95% accurate when dealing with small scale worked examples from the literature (Whitcombe et al. 2001 and 2002).

This original model was then expanded to take into account particle attrition and particle feedback from the cyclone into the bed. The attrition model selected was a three part model developed using FCC catalyst particles and which calculates cyclone, bubble and jet attrition separately (Werther and Reppenhagen 1999).

The three part attrition model uses three different attrition co-efficients, one for each of the attrition sources. These three co-efficients are; cyclone attrition, jet attrition and a particle attrition factor which is used to determine bubbling attrition. Co-efficients were determined experimentally by Werther and Reppenhagen (1999), using FCC catalyst in a small scale fluidised bed. As the attrition co-efficients were based on FCC catalyst, all values were deemed reasonable for use in this paper as it is near impossible to obtain accurate industrial attrition coefficients from a large scale operational FCCU. It is important to note that no reference has been found in the literature warning of scale up problems associated with the use of attrition models based on small scale systems (as is the Werther and Reppenhagen model) when used to model industrial systems. After the model was developed it was tested on a small scale system and was found to over predict emissions by 100% to 200%.

The model is designed to have 11 input variables (operating parameters) and one output variable (emission rates). The input factors, consisting of operating parameters are listed in Table 1.

2.2. Sensitivity Analysis

The New Morris Method, as developed by Campolongo and Braddock (1999) and corrected by Cropp and Braddock (2002), was selected and used to test for second and third order interactions in the input parameters for sensitivity. The original Morris Method is a 'one-factor-at-a-time" (OAT) screening method which was selected as it allows first order sensitivity to be determined by alternating each of the parameters individually and then estimating the overall sensitivity measure of that parameter to the output. The New Morris Method is an extension of the original Morris method extended to identify second order interactions between input parameters. In doing so, this New Morris Method provides an estimate of the output sensitivity for any pair of input parameters, while minimising the computational cost of the evaluation (Campolongo and Braddock 1999). The sensitivity analysis was conducted on the entire model

The software allows a mean (μ) and standard deviation (δ) for the first order effects, from the Morris Method, as well as a new mean for the second order effects, lambda (λ), and its standard deviation, to be determined for the input factors of the model. The mean allows the overall influence of the factors to be determined, while the standard deviation identifies factors with possible higher order interactive effects. The new term, lambda, provides a global sensitivity measurement for 2-factor interactions (Campolongo and Braddock 1999). The standard deviation of lambda has been shown to accurately predict possible third order interactions in the system (Cropp and Braddock 2002).

The software requires the identification of the input factor, and the range of values to be set for these factors (Table 1), the number of sample runs (200), and the discretisation of the parameter space (10). The number of samples and the discretisation of the sample space were determined to provide sufficient testing of the sample space whilst minimizing the computational requirements of the model. The upper and lower bounds were determined by the absolute maximum and minimum possible values that could be seen in an operating FCCU. All selections of parameters and bounds were done in conjunction with the practical operations of the refinery. Due to the preliminary nature of this work and the use of an OAT technique, any errors developed by possible correlations in the input values were deemed to be reasonable.

Table 1. List of Factors and their ranges of values used in the Sensitivity Analysis and Genetic Algorithm.

Factor	Parameter	Lower bound	Upper bound
1	Bed Velocity (m/s)	0.1	1.5
2	300μm size fraction (% mass)	0.001	0.20
3	200μm size fraction (% mass)	0.001	0.40
4	100μm size fraction (% mass)	0.05	0.60
5	80μm size fraction (% mass)	0.1	0.90
6	60μm size fraction (% mass)	0.1	0.90
7	40μm size fraction (% mass)	0.05	0.60
8	20μm size fraction (% mass)	0.001	0.40
9	1μm size fraction (% mass)	0.001	0.20
10	Catalyst density (kg/m^3)	1197	1323
11	Shape factor (perfect sphere = 1)	0.70	1

2.3. Genetic Algorithm

Genetic Algorithms (GA) are optimization algorithms based on the mechanics of genetics, that is the idea of the survival of the fittest (Karr et al. 1995). The algorithm allows for the search space of the model output to be optimized through the use of previously determined calculations. Random strings are developed based on the parameter set provided, the output of the model is then determined with each string being assessed on the quality of the solution provided (its fitness). This information is then used to generate the next generation of strings, as the fittest ones will generate the largest copies, allowing the parameter space to be optimized, in this case, to provide a minimum level of emissions from the fluidized bed (Karr et al. 1995). The GA requires the upper and lower bounds of the parameter space (Table 1), the number of generations (100), the size of the populations (30) and the number of bits used in each population (10). The GA calculates the optimal parameters and displays their value along with the minimal emission rate (in mg/m^3) achieved from the fluidized bed. As the GA utilizes random numbers and the generation of successive populations, the optimized values of the parameters can change with each run. To overcome this, the GA was run 10 times with the average and standard deviation for each parameter and the minimum emission rate determined.

3. RESULTS AND DISCUSSION

The following results were obtained from the model.

3.1. Model Results

The model was run using the standard operating conditions for an industrial FCCU, with results presented in Figure 1. The model predicted an emission rate of approximately 11,000mg/m^3, which is considerably greater than the actual emission found for the industrial FCCU, which ranges from a maximum of just over 1,000mg/m^3 to a steady state minimum of approximately 100mg/m^3 (Agranovski and Whitcombe 2002). As the original model was reasonably accurate when dealing with small scaled systems (refer to Whitcombe et al. 2001), it is likely that the attrition component of the model was causing the error. It is recognized in the literature that modeling of emissions from large scale fluidized beds (such as a FCCU) are prone to error due to scale up factors and poor robustness of the developed models (Milioli and Foster 1995; Tasirin and Geldart 1998). However, there is no mention in the literature that scale up problems exist for attrition models, and as the attrition model selected was developed using FCC catalyst all attrition coefficients should be reliable.

To test whether the attrition model does cause the overestimation of emissions from the FCCU system, the attrition term was removed with the results also presented in Figure 1.

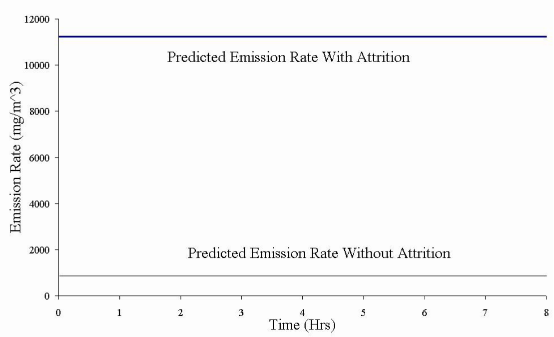

Figure 1. Emission rate from the model with and without the attrition term.

The emission rate without attrition was approximately 800mg/m^3, which is within the range of emissions observed at an industrial FCCU. This indicates that it is the attrition terms which are the likely source for the inaccuracies of the model. To understand which of the three attrition components (cyclone, jet or bubble) causes the excessive emissions, the contribution of each attrition term to the over all attrition rate were plotted in Figure 2.

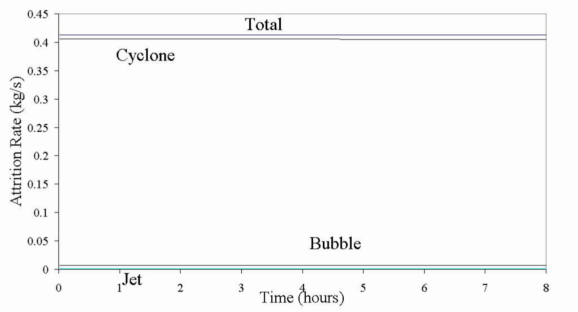

Figure 2. Calculated Attrition Rates

From Figure 2, the main source of attrition within the model was produced from the cyclone attrition term, with the bubble and jet attrition being almost negligible. This indicates that there is indeed some sort of scaling issue which is prevalent in attrition models.

3.2. Sensitivity Results

The first order sensitivity results of the model are presented in Figure 3 with the second order effects shown in Figure 4.

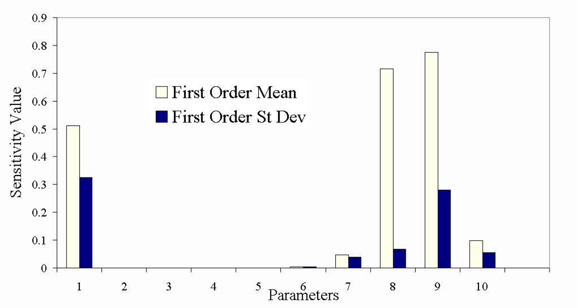

Figure 3. First order affects and standard deviations

The SA results of the updated model clarify the influential parameters in the system, preliminarily identified in (Whitcombe et al. 2002). Overall the sensitivity analysis indicates that the emissions are most sensitive to the bed velocity, the fine particle sizes (factors 9, 8 and 7) and particle density (10) and is insensitive to the mid sized particles (factors 5, and 6) identified in the preliminary assessment. The second order effects support the first order findings with the finer particle sizes, density and gas velocity interactions influencing the rate of emissions. These second order results support the theory that velocity, particle density and the fine particles are the most sensitive in terms of influencing emission rates.

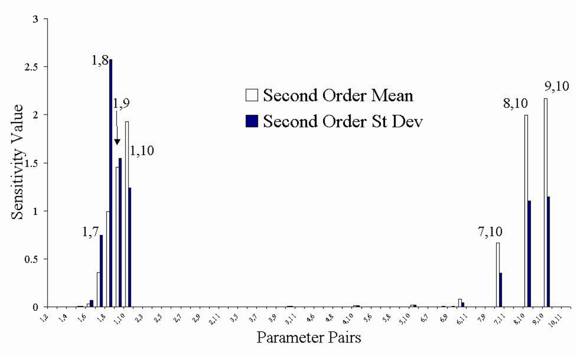

Figure 4. Second order interactions

In a physical sense, the interaction of the particle density, gas velocity and smaller particle sizes relates to the ability of particles to be removed from the bed and the rate at which attrition occurs. The original SA work outlined previously and in (Whitcombe et al. 2002) showed a mid range of parameter sensitivities (mid sized particles), which is not seen in this upgraded model. This suggests that the addition of the attrition tem has led to an increase in sensitivity of the model towards the smaller particle sizes, and the gas velocity whilst reducing the influence of the mid sized particles, as expected in a real FCCU. One reason for this is that at high gas velocities particle attrition is likely to produce a significant amount of fine particle emissions, thus reducing the influence of other parameters.

The sensitivity of air emission towards gas velocity and particle density is understandable. For particles to be emitted from the system they must be first entrained from the fluidized bed and carried up in the gas stream and through the cyclones. An increase or decrease in either the density of particles or the gas velocity will alter both forces acting on a particle and the forces inside the cyclones, allowing fluctuations in emission rates. The low sensitivity seen in the larger sized particles (parameters 2 ,3 and 4) as well as the shape factor (parameter 11) is reasonable, larger sized particles and shape factor are not normally considered important in terms of emissions.

3.3. Genetic Algorithm

Optimised parameter values for the minimisation of particle emissions using the original attrition co-efficients in the model and using the optimised attrition co-efficients, were generated using the genetic algorithm and are presented in Table 2. Results are the average value obtained from 10 separate runs from different initial points of the GA program.

Table 2. Optimal operating conditions to minimise particle emissions determined from the Genetic Algorithm for the original model and optimised attrition constants.

	Original Model		Optimised Attrition Constants	
	Average	CV(%)	Average	CV(%)
Emission rate (mg/m3)	5450.2	6.6966	2899.8	3.0933
Bed Velocity (m/s)	0.66958	16.315	1.2004	12.138
300μm size fraction (% mass)	0.079904	44.682	0.062161	79.668
200μm size fraction (% mass)	0.19751	56.835	0.11585	40.021
100μm size fraction (% mass)	0.090499	87.572	0.098932	97.949
80μm size fraction (% mass)	0.69011	33.35	0.31185	69.548
60μm size fraction (% mass)	0.19509	40.098	0.1262	16.845
40μm size fraction (% mass)	0.051559	3.4567	0.050699	1.8738
20μm size fraction (% mass)	0.0001	0.00	0.0001	0.00
1μm size fraction (% mass)	0.0001	0.00	0.0001	0.00
Catalyst density (kg/m^3)	1652.8	2.6576	1682.6	0.81966
Shape factor (perfect sphere = 1)	0.65049	18.96	0.74892	16.075

The minimised rate of particle emission using the original attrition co-efficients was calculated as 5,450.2mg/m^3. The predicted optimal value is considerably higher than that tested at an industrial FCCU, indicating that the attrition induced error is not removed through optimisation of the model. Also the optimised parameters include a gas velocity of 0.66m/s, which is at the lower end of the operational range.

The low coefficient of variation (CV) indicates parameter sensitivity, as the optimal value is found in a narrow range. The velocity, 20 and 1μm sized particles, and catalyst density are shown in Table 2 to be sensitive parameters affecting the emissions.

When the SA results are compared to the GA results, some interesting trends are seen. Firstly, the large values for the coefficient of variance obtained from the GA runs for the larger sized particles (parameters 2, 3 and 4) indicate that a large range of these parameter values can be used to minimize the emission results, ie they are not sensitive. This supports the SA results that these factors are not influential in controlling particle emissions. Of interest is that the optimal shape factor was 0.65 indicating that a non-spherical particle is the preferred shape to minimize emissions in the industrial system. No reason can be given to explain this.

To test how each of the attrition terms influences the predicted emission rates, the three attrition coefficients (cyclone, bubble and jet) were altered in increments of 10 times the original value, with optimised emission rates then being recalculated. The results are presented in Figure 5.

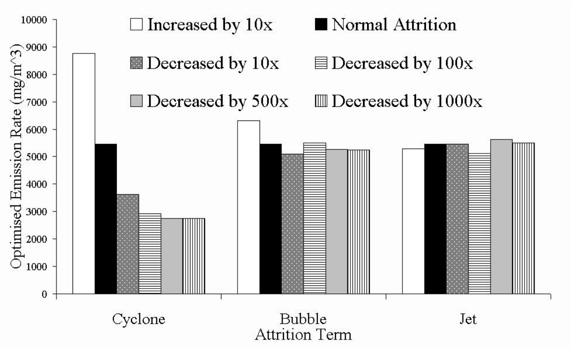

Figure 5. Optimal emission rates from the model with alterations in each of the three attrition coefficients

From Figure 5 it is clear that altering each of the attrition terms influences the optimal emission rate, with the cyclone attrition coefficient being the most influential. Altering the bubble or jet attrition term does not significantly alter the optimal emission rate, whilst the cyclone attrition terms stabilises when decreased by a factor of 100. As all of the attrition terms used were sourced from the literature and based on experimental observations of small scale systems, it appears that the experimental work done on small scale systems are accurate for determining bubbling and jet attrition, but not cyclone attrition in large scale fluidized beds. It appears that large scale industrial cyclones have far less attrition when compared with scaled down laboratory devices. This is a result not directly discussed in the literature and one that is not mentioned when dealing with the modeling of air emissions from fluidized beds.

Knowing that the cyclone attrition coefficient is reduced under larger scaled industrial conditions, the model was optimized using the reduced value (decreased by 100x) of the cyclone attrition rate shown in Figure 5. The new optimum level of the emissions is presented in Table 2. The new level is almost half of the original optimization with an emission rate of 2,899mg/m^3. Although most parameters were found to be similar, the new optimized emission rate was determined at a significantly higher gas velocity. The new velocity of 1.2m/s is at the higher end of the operational range of an FCCU and it is unclear as to why this velocity is optimal. It is known that gas velocity influences emission levels and one would think that a lower velocity would produce lower emissions. One possible reason is that the cyclones are functioning better at this higher velocity, overcompensating for any increase in particle carry over due to higher velocities. This is supported by earlier work which showed that the more simplistic model could produce lower emission levels at higher gas velocities (Whitcombe et al. 2001). The shape factor was also found to be slightly higher (0.75), indicating that a more spherical particle is required to reduce emission under realistic attrition conditions.

4. CONCLUSION

The updated model has shown that fine particles, gas velocity and particle density are the most important operational parameters in terms of minimising particle emissions from a FCCU. Overall the model was found to significantly over estimate the true nature of emissions from an FCCU. It was concluded that this error was due to scale up issues arising from the attrition terms used. Optimization of the model supported the hypothesis that it is the attrition terms, and in particular the cyclone attrition coefficient, which is responsible of the over estimation of particle emissions. It appears that cyclone attrition is substantially less in larger systems than found in smaller laboratory based cyclones. Optimisation of the model with a more realistic attrition term showed that even at high gas velocities emission levels could be minimised. This work has supported earlier studies identifying particle size, density and gas velocity as the main parameters which should be controlled to minimise particle emissions from FCCUs. Further work is needed to develop better attrition models for use in large scale systems, so more accurate emission models can be developed.

5. ACKNOWLEDGEMENT

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