# CHAPTER 4
# OPTIMIZATION OF PROCESS PARAMETERS USING RSM

## 4.1 Introduction

The optimization of process parameters via suitable statistical analysis is essential for finding optimum values of effective variable to achieve highest product yield with rich in BTE content. Optimization refers to improving the performance of a system, or a process in order to obtain the maximum benefit from it (Bezerra et al., 2008). Earlier, the optimization process was carried out by monitoring the influence of one factor at a time on an experimental response/output, keeping the other parameters at their constant value (Bezerra et al., 2008). This optimization technique is called one variable at a time. The major disadvantage of this method is, it does not includes the interaction between effective process parameters and effect of more than one variable simultaneously on output/response (Bezerra et al., 2008). Another disadvantage of one variable optimization is the number of experiments required to optimize the process is more, which leads to increase the overall cost and time (Yolmeh and Jafari, 2017). To overcome these problems, the optimization of analytical procedures have been carried out using multivariate statistical techniques. Among the most relevant multivariate statistical techniques used in analytical optimization is response surface methodology (RSM) (Yolmeh and Jafari, 2017). The response surface methodology was developed by Box and co-workers in 1950 (Gilmour, 2006).

The RSM is based on fitting the mathematical models e.g., linear, square polynomial functions and others to the experimental results from the designed set of experiments and verification of the model obtained by the statistical techniques (Bezerra et al., 2008; Witek-Krowiak et al., 2014). The main objective of RSM is to obtain the optimum process conditions of a process to achieve maximum/highest response or output from the process under consideration (Karimifard et al., 2018). There are various design methods reported in literature which are used for the optimization via RSM technique such as full three level factorial design (Sadi et al., 2018), Box-Behnken design (BBD) (Mo et al., 2014), central composite design (CCD) (Selvaganapathy et al., 2020) and Doehlert design (Caldas et al., 2013). The various advantages and disadvantages related to the above mentioned design methods are discussed below:

(i) Full three-level factorial designs

Full three-level factorial design is applicable, when the effective variable is more than two. The number of experiments required for this design is calculated by the following equation (4.1):

$$N = 3^{k} \tag{4.1}$$

Where, N is the number of experiments and k is the number of selected effective variables.

The main disadvantage of three-level factorial design is that this method requires more experimental run as compared to other design methods for two or more than two effective variables (Bezerra et al., 2008).

(ii) Box-Behnken designs

The Box-Behnken designs (BBD) is more efficient method as compared to the three level factorial design. In this design method, the number of experiments are minimized in quadratic model fitting. It is better to use second order polynomial model to accurately describe linear interactions and quadratic effects (Witek-Krowiak et al., 2014). The number of experiments are calculated using the equation (4.2):

$$N = 2\,(K\text{-}1) + C_p \tag{4.2}$$

Where, k is the number of effective variables and Cp is the number of the central points. All selected variables have to be adjusted at three levels (-1, 0, +1) with equal interval (Bezerra et al., 2008). This design is more economical than other three level designs due to its ability to allow points selection from the three level factorial arrangement (Bezerra et al., 2008).

(iii) Central composite design

The central composite design was presented by Box and Wilson. This design is basically a combination of (i) full factorial design and (ii) an additional design in which experimental points are at a distance α from its center; and (iii) a central point. The number of experiments are calculated using equation (4.3):

$$N = k^2 + 2k + C_p \tag{4.3}$$

Where, k is the number of effecting variables and $C_p$ is the number of runs at central points. The value of α is depends on the number of effective variables and can be calculated by $\alpha = 2^{(k-p)/4}$ and all effective variables are studied in five levels (-α, -1, 0, +1, +α). Thus, the experimental runs required for CCD are more than BBD (Bezerra et al., 2008).

(iv) Doehlert design

The Doehlert design was developed by Doehlert. This design describes a circular domain for two variables, spherical for three variables, and hyperspherical for more than three variables, which accents the uniformity of the studied variables in the experimental domain (Bezerra et al., 2008). Doehlert design has several advantages such as few experimental points, high efficiency, and economically effective. Different from CCD and BBD, these designs are not rotatable due to their number of estimation for varied parameters/variables (Riswanto et al., 2019).

The required number of experiments are calculated using the equation (4.4) as stated below:

$$N = k^2 + k + C_p \qquad (4.4)$$

Where, N is the number of experiments, k is number of variables and $C_p$ is the number of the central points.

It is seen from the thorough literature that most of the researchers viz Mortezaeikia and Tavakoli, 2022; Olalo, 2022; Mo et al., 2014; Salman et al., 2019 used RSM-BBD techniques to optimize the effective process parameters for the pyrolysis process of waste plastic to achieve maximum output for their selected effective variables.

Olalo, 2022 conducted the pyrolysis of waste polystyrene to optimize the three different pyrolysis parameters i.e., temperature (300-500 $^oC$), residence time (60-120 min) and feed stock size (2-4 cm) using RSM-BBD technique in order to obtained the maximum liquid yield. The maximum liquid yield of 91.38 wt.% was obtained for the pyrolysis temperature of 550 $^oC$, residence time of 120 min and feed stock size of 2 cm. Mo et al., 2014 conducted the pyrolysis of polystyrene waste to enhanced the styrene content in the pyrolysis oil. The pyrolysis process parameters were optimized using RSM-BBD tool to achieve the highest liquid yield with maximum styrene content. Three different pyrolysis parameters i.e., temperature, heating rate and carrier gas flow rate were selected as the process variables. The temperature was varied from 375 $^oC$-525 $^oC$, heating rate from 10 $^oC$/min to 40 $^oC$/min and carrier gas flow rate from 50 mL/min to 200 mL/min to achieve the maximum liquid yield rich in styrene content. The optimum conditions for the developed model were found to be at a temperature range of 470 $^oC$- 505 $^oC$, a heating rate of 40 $^oC$/min, and a carrier gas flow rate range of 115-140 mL/min. The liquid yield of 86.08 wt.% was obtained with styrene content of 64.52 wt.% at the optimized process conditions.

Mortezaeikia and Tavakoli, 2022 investigated the catalytic microwave assisted pyrolysis of waste brominated acrylonitrile styrene butadiene (ABS-Br) using silicon carbide (SiC) and iron impregnated on silicon carbide (Fe/SiC) catalyst to obtain the maximum pyrolysis oil yield. The response surface methodology coupled with Box-Behnken design (RSM-BBD) was used for the optimization of the pyrolysis process. The pyrolysis parameters like microwave power, temperature, and carrier gas flow rate and receptor/plastic ratio were considered to maximize the liquid yield. The optimal values were temperature of 471°C, a microwave power of 1.54 amps, carrier gas flow rate of 215 mL/min and catalyst/plastic weight ratio of 0.05. The pyrolysis oil of 56.97 wt.% was recorded at optimized conditions. The GC-MS analysis revealed the high concentration of single ring and polyaromatic hydrocarbons in the pyrolysis oil. The impregnation of Fe on SiC showed higher catalytic activity in conversion of polyaromatic hydrocarbons to single ring aromatics.

The detailed literature review shows that the response surface methodology (RSM) coupled with Box-Behnken design (BBD) is very effective and popular method for optimization of process parameters to achieve maximum liquid yield. In view of this, in the present study RSM-BBD was used to optimize the effective process parameters for the best reactor arrangement i.e., multiphase catalytic pyrolysis of WEPS in order to obtain the maximum liquid yield rich in target molecules BTE. There are many process variables such as reaction temperature, reaction time, pressure (Pinto et al., 2013), heating rate, carrier gas flow rate (Mo et al., 2014), size and weight of feed stock (Selvaganapathy et al., 2020) have been reported in the literature. However, in the present study, three effective variables i.e., temperature (A), heating rate (B) and feed to catalyst ratio (C) were considered as effective operating variables as the information provided in the literature shows that the above mentioned three variables have highest impact on the pyrolysis process for achieving maximum liquid yield. The fresh feed of 50 g was always used for each experimental run and calculated amount of catalyst was used as per the required feed to catalyst ratio. The detailed experimental procedure to perform the multiphase catalytic pyrolysis have already been discussed in Chapter 3 (Page no. 48).

### 4.2 Optimization using RSM-BBD

The RSM was used to find out the optimum value of effective process variables of multiphase catalytic pyrolysis process. As already mentioned, the benefit of RSM is that it takes into account the interaction among the effective variables as well as effect of two variables together on response. It is already mentioned in the introduction section (page no. 62) that, there are various methods of RSM such as Box-Behnken design (BBD), central composite designs (CCD) and Doehlert matrix used to determine the optimum value of effective parameters in order to maximize the response. However, a highly effective BBD method was used in this study for optimization of process variables, as only few combinations of effective operating variables are used to determine the optimum values of various process conditions (Luna et al., 2020; Gupta and Mondal, 2019). Thus, less number of experiments have performed to achieve the highest value of response in less time as comparison to the purely experimental method. Additional advantage of the BBD is that, it is approximately 98 % efficient at predicting a second order model with three factors (Charusiri and Numcharoenpinij, 2017). In this work, design expert software trial version 8.0.7.1 was used to maximize the liquid yield of multiphase pyrolysis of WEPS for three different types of catalyst using BBD in RSM. As it is mentioned earlier, three selected operating process variables i.e., temperature (A), heating rate (B) and feed to catalyst ratio (C) were varied between three levels viz -1 (minimum), 0 (central), and +1(maximum) as shown in Table 4.1.

**Table 4.1** Range of independent process variables.

| Process variables | Experimental level | | |
|---|---|---|---|
| | **-1** | **0** | **+1** |
| Temperature (A) ($^oC$) | 500 | 550 | 600 |
| Heating rate (B) ($^oC/min$) | 10 | 15 | 20 |
| Feed to catalyst ratio (C) | 10:1 | 20:1 | 30:1 |

The number of experiments in BBD method was calculated by the equation (4.5):

$$N = 2n\ (n-1) + N_p \tag{4.5}$$

Where, N is the number of experiments, n is number of process parameters, Np is number of repetition at center point (Panjiara and Pramanik, 2020).

Thus, it is calculated from equation (4.5), that only 17 experimental runs are required to optimize the process variables for achieving highest liquid yield contains highest amount of BTE. The selected 17 experimental runs include 12 unique and 5 center point runs. The responses for each set of input variables were determined by analysis of variance (ANOVA), coefficient of determination ($R^2$) and three-dimension contour plots. The Fischer test value (F-value) and probability value (p-value) were obtained by the ANOVA to see the significance of each independent variable on the response. The p-value should be less than 0.05 (Singh et al., 2019) and F-value should be greater than 1 for a significant model (Panjiara and Pramanik, 2020).

The generalized second-order polynomial model used in the RSM is presented in equation (4.6) below:

$$Y = \delta_o + \sum_{i=1}^{k} \delta_i X_i + \sum_{i=1}^{k} \delta_{ii} X_i^2 + \sum_{i \text{ and } j=1,\, i \neq j}^{k} \delta_{ij} X_i X_j \tag{4.6}$$

Where, Y is the response (liquid yield), $\delta_o$ is the constant regression co-efficient, $\delta_i$, $\delta_{ii}$ and $\delta_{ij}$ are the coefficients for linear, quadratic and interaction parameters. $X_i$, and $X_j$ are the coded independent parameters (Panjiara and Pramanik, 2020**)**. The results obtained from the RSM-BBD analysis are discussed in the next chapter 5 (page no. 70).