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RESEARCH ARTICLE

Optimization and modeling of reactive conditions for free radical solution polymerization of SA-co-BA copolymer based on the yield using response surface methodology

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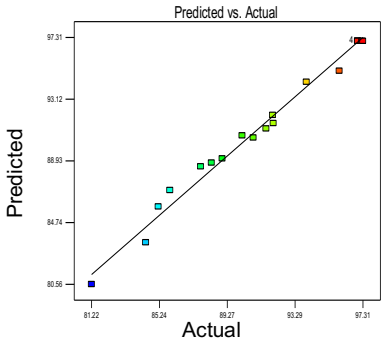
Graphical abstract



Abstract

In the recent years, response surface methodology (RSM) is one of the most common optimization methods employed in the chemical process. The satisfactory model for predicting the maximum yield in solution polymerization has been a challenge due to various conditions during the synthesis process. In this study, interactive impacts of three parameters which are reaction time, concentration of initiator, and reaction temperature on the yield in free radical polymerization of SABA copolymer using toluene as solvent was investigated using experimental design central composite design (CCD) model under response surface methodology (RSM). The result showed the optimization conditions were reaction time of 7 h, initiator concentration of 1 wt %, and reaction temperature of 90 °C with the corresponding yield of 97.31%. The analysis of the regression model (ANOVA) detected an R^2 value of 0.9844, that the model is able to clarify 98.44% of the data variation, and just 1.23% of the whole differences is not clarified by the model. Three experimental validation runs were carried out using the optimal replicate conditions and the highest average yield value obtained is 97.15%. There is an error of about 0.97% as compared to the expected value.Therefore, the results indicate that this model is reliable and is able to predict the yield response accurately. it established that the regression model is extremely significant, indicating a strong agreement between the expected and the experimental values of SABA yield.

Keywords: Free radical polymerization, response surface methodology, SABA copolymer, yield

INTRODUCTION

It is crucial to develop the model's performance and to raise the yield procedures without rising the cost. The technique employed to attain this goal is free radical polymerization. There is a change of factor in the overall preparation of determining the optimum processing conditions while the other factors are kept at fixed levels. This technique is named a one-factor-at-a-time method. The main downside of this method is its lack of comprising interactive impacts between parameters. Furthermore, the complete impact of the variable on the process is also not described by this method. With the aim to eliminate this problem, optimization studies can be conducted by employing response surface methodology (RSM) (Aydar, 2018; Baş and Boyacı, 2007; Teh *et al.*, 2013).

RSM is a set of mathematical as well as statistical method suitable to enhance the experimental model building, developing, and optimizing processes variable. In addition, it can be used to discover the interaction of many affecting variables (Agrawal *et al.*, 2020; Es-said *et al.*, 2019).

RSM is a numerical technology that uses quantitative data from the connected experiment to control the regression model and to improve response (output factor), which is affected by numerous independent factors (input factors) (Jiang *et al.*, 2017; Raza *et al.*, 2017). The greatest feature of this technology RSM is to minimize the number of experimental runs, saving of energy, time, and chemicals. The effects of multiple factors can be considered with the aid of experimental statistical design (Sohrabi *et al.*, 2016; Unal, 2016; Zaroual *et al.*, 2009). The experimental statistical experimental design is an structured method to study factor impacts professionally and in a reliable manner, and it yields additionally suitable and specific data with fewer experiments as compared to OFAT method (Teh *et al.*, 2013). The experimental design and information data analysis detects the factors that are significant and whether there are any interaction influences between the factors. Experimental statistical design has several uses in chemistry, e.g., in synthesis, separation, optimization of logical approaches (Harang *et al.*, 2001; Rautio *et al.*, 2009; Singireddy *et al.*, 2019).

In any experimental design, there are variables like time, amount of solvent, concentration of monomers, temperature, and the initiator concentration that will effect the purity, selectivity, and actual yield. The interrelationships among the factors are difficult, and therefore, the study of this procedure in optimizing the variables is also difficult and consumes a lot of time. Thus, the studies that employ traditional experimental methods are deemed not efficient (Singireddy *et al*., 2019). To optimize chemical reaction, different variables that rule the whole reaction must be considered. For the same reason, this paper is focuses on studying and the optimizing the synthesis of SABA copolymer in order to describe effective conditions for different models. The influence of the experimental factors slike temperature, time and initiator concentration on the actual yield was investigated by the RSM via central composite design (CCD). Optimization during CCD shows rapid and effective single optimum experimental combinations. Furthermore, it needs fewer experimental runs as compared to other techniques (Mohammadi *et al.*, 2020).

In the recent years, RSM was found to have applied in previous contributions (Banerjee *et al.*, 2012; Ghasemi *et al.*, 2010; Heidari *et al.*, 2018; Lee *et al.*, 2011; Nasef *et al.*, 2011; Razali *et al.*, 2015; Zheng *et al.*, 2015). According to Razali et al. (2015) (Razali *et al.*, 2015) used RSM in studying polydiallydimethylammonium chloride grafting and using potassium persulphate as an initiator of free radicals. Four factors were examined by the central composite design (CCD), which were the mole ratio of diallyl dimethylammonium chloride, initiator concentration, temperature, and time in determining their interactive and individual impacts on the percentage of grafting. They found acceptable outcimes since the the value expected by their resulting model was very close to the experimental outcome at the optimized conditions.

According to Aroonsingkarat (Aroonsingkarat and Hansupalak, 2013) have researched the influence of operating states towards transformation in the polystyrene and rubber graft co-polymerization using CCD via RSM. The temperature, amount of chain transfer agent, percentage of deproteinized rubber, and reaction time were the four factors examined. In an associated research, Sresungsuwan and Hansupalak (2013) (Sresungsuwan and Hansupalak, 2013) examined the effect of operating states using RSM towards compatible natural rubber / styrene blend's on mechanical properties. No prior studies investigated for free radical polymerization of styrene to the best of the researchers' knowledge.

In the previous study, we recognized the four parameters effecting the yield in the free radical polymerization of SABA copolymer using the OFAT method (temperature, time, concentration of initiator, and mass ratio of monomer concentration). Nevertheless, this method cannot test the interactions of the parameters considered (Elarbe *et al*., 2019). In the present work, we optimize the effect of each parameter on the yield in the solution polymerization of SABA copolymer. Response surface methodology by CCD was employed in designing the run of experimental, generating a model, and exploring the process factors.

EXPERIMENTAL

Materials

Stearyl Acrylate was used as a monomer (SA, Sigma-Aldrich, 97%), and the other monomer was behenyl acrylate (BA, Aladdin chemicals, 95%). Benzoyl peroxide (Sigma-Aldrich, 99%) were used as the initiator, toluene (Sigma-Aldrich, 99%) as solvent and methanol (Sigma-Aldrich, 99%).

Synthesis of SA-co-BA copolymer

The free-radical solution polymerization method was used to synthesize stearyl acrylate-behenyl acrylate (SA-co-BA) copolymer by utilizing equal proportions of the functional monomers between SA and BA (1:1). The reaction was carried out in a 250 mL three-neck round-bottomed flask that was equipped with a thermometer, reflux condenser, magnetic stirrer, and nitrogen gas inlet. Under constant stirring, a monomers mixture has been dissolved in toluene solution deemed suitable at different reaction temperatures for different reaction times and added a nitrogen atmosphere for a period of 45 min. At this point of time, benzoyl peroxide (BPO) has been dissolved in an amount of toluene deemed suitable, which acted as an initiator at different percentage based on the total monomers. For every 15 min, the solution of benzoyl peroxide has been injected drop by drop for the first one hour after the reaction had started. After the completion of the reaction, it was placed at the room temperature. After that, the copolymer solution was precipitated, filtered, and washed thrice by methanol. The final product that obtained was dried 24 h in a vacuum oven at the temperature of 40 °C for. A white powder was obtained as final state of the copolymer. Each run was carried out in triplicates, and then the average value was taken as the final value. The optimum conditions of independent variables were worked out to get the maximum yield percentage (Y %). The percentage of the yield was determined as per Eq. (1).

$$Yield\ (\%) = (Obtained\ weight\ of\ the\ product\ (g)/Total\ weight\ of\ the\ material\ (g)) \times 100 \qquad (1)$$

Design of experiment

Table 1 shows, the experiment design levels and the range of independent variables used in this work. A three-factor five-level central composite design (CCD) was further studied, needs 20 runs (calculated on the basis of Eq. (2)), including 6 replicate runs at the center, 6 axial runs, and 8 factorial runs.

$$N=2^n+2_n+N_{C=}2^3+2\times3+6=20 \qquad (2)$$

Where N is the number of experimental runs, and n is the number of factors.

The model equation is determined, and the model equation coefficients are expected. The model that has been employed in RSM is commonly a full quadratic equation. The α-value was stable at 1 (face-centered) for this model, and the experiment model's response was obtained. The response (yield) can be employed to improve an experimental model that relate the response to the three parameters using a additional-grade polynomial as follows in Eq. (3).

$$Y = \beta o + \sum_{i=1}^{n} \beta iXi + \sum_{i=1}^{n} \beta iiXii + \sum_{i=1}^{n-1} \sum_{j=i+1}^{n} \beta ijXiXj \qquad (3)$$

Where Y is the expected response, β0, βi, βii, and βij are constant coefficients for interrupt, linear, quadratic and interaction coefficients respectively and Xi and Xj are coded for independent factors of polymerization. Table 2 demonstrates the experimental design model, experimental run order, and the response (yield %) for the three parameters and 20 empirical runs created.

Software of Design Expert 7.1.6 is one of the essential softwares used for regression study and assistances to design the model and explain experiments with multi-variable impacts in addition to minimizing the number of experimental runs. There is a wide range of designs given by this software, consisting of composite models, fractional factorials, and factorials. This can provide the researchers with a assembly of numerical and mathematical methods named response surface methodology (RSM). The statistical analysis of the experimental model, that comprises full quadratic, linear and interaction coefficient has been completed by ANOVA study with F-test to find the empirical correlation among the output and input variables. In order to improve the model, every code of the model is statistically evaluated, specifically the importance of F-values with $P \leq 0.05$. The R^2, adjusted R^2, and expected R^2 values, the adequate precision and lack of fit of models were found to satisfy the feature of the recommended polynomial. The plot of contour and response surface plot have been drawn to visualize the input and output interactions.

The independent variables studied were the temperature (X_1), time (X_2), and concentration of initiator (X_3), which are considered in three levels, as seen in Table 2. In the chosen coordinate system, the codes 1, 2, and 3 describe the low, medium, and high levels, respectively.

All independent factors and their ranges were selected based on our previous study.

Table 1 Five-level three-condition variables with their coded levels

Independent variables	Units	Code Symbol					
			-α	-1	0	+1	+α
Reaction Temperature	°C	A	80	85	90	95	100
Concentration of Initiator	wt %	B	0.5	0.75	1	1.25	1.5
Reaction Time	h	C	6	6.5	7	7.5	8

RESULTS AND DISCUSSION

OFAT experiments were conducted in our previous study (Elarbe *et al.*, 2019). The best condition of the most efficient variables of SABA PPD was obtained using mass ration of (1:1) wt % between two polymers, 7 h of reaction time, 90 °C of reaction temperature and 1 wt % of the concentration of initiator, according to the highest amount of the yield at different conditions. CCD was employed in this paper to minimize the range of conditions with the maximum yield of SABA copolymer. The influence of variables on the yield was investigated. The experiments were run according to Table 2 in order to maximize the yield.

Regression model equation

Table 2. shows the observed percentage yields of SABA copolymer for the 20 experimental runs. These data are employed to estimate the quadratic polynomial equation's coefficient, as previously demonstrated in the design experiment Eq. (3). These determined coefficients for the actual and coded values are illustrated in Table 3**,** together with the coefficients of estimation R^2, adjusted R^2, and predicted R^2. The quadratic models are depicted below in Eq. (4) in terms of actual value of factors. The positive signs in the models signify synergistic impacts of factor. Whereas, the negative sign shows the antagonistic impact.

Table 2 The experimental design matrix for SABA copolymer polymerization synthesization

Std	Factor A Reaction Temperature °C	Factor B Concentration of Initiator wt%	Factor C Reaction Time h	Observed Yield %	Predicated Yield %
1	85.00	0.75	6.50	85.87	86.90
2	95.00	0.75	6.50	88.97	89.05
3	85.00	1.25	6.50	90.15	90.61
4	95.00	1.25	6.50	91.57	91.08
5	85.00	0.75	7.50	85.18	85.79
6	95.00	0.75	7.50	90.81	90.47
7	85.00	1.25	7.50	91.96	92.00
8	95.00	1.25	7.50	95.91	95.00
9	80.00	1.00	7.00	84.44	83.43
10	100.00	1.00	7.00	87.69	88.58
11	90.00	0.5	7.00	81.22	80.59
12	90.00	1.5	7.00	88.33	88.84
13	90.00	1.00	6.00	92.00	91.52
14	90.00	1.00	8.00	93.96	94.32
15	90.00	1.00	7.00	95.80	96.82
16	90.00	1.00	7.00	96.91	96.82
17	90.00	1.00	7.00	96.80	96.82
18	90.00	1.00	7.00	96.95	96.82
19	90.00	1.00	7.00	97.25	96.82
20	90.00	1.00	7.00	97.31	96.82

Final equation in terms of actual factors:

$$Y= -893.44068 +18.28459\,A+100.30318\,B+28.19432\,C-0.10812\,A^2-48.40909\,B^2-3.89727\,C^2-0.33600\,AB+0.2530\,AC+5.000\,BC \quad (4)$$

Table 3 The coefficient of the model.

Variable	Coefficient	
	Coded	Uncoded
Constant	96.82	-893.16114
A	1.29	+18.28459
B	2.06	+100.30318
C	0.70	+28.19432
A^2	-2.70	-0.10812
B^2	-3.03	-48.40909
C^2	-0.97	-3.89727
AB	-0.42	-0.33600
AC	0.63	+0.25300
BC	0.63	+5.00000
R-square	0.9844 %	
Adjusted R-square	0.9704 %	
Expected R-square	0.8918 %	

The positive marks in the models indicate the synergetic impacts of the parameter, whereas the negative mark indicates a hostile impact. The study of the regression model's variance (ANOVA) in Table 3 detected an R^2 value of 0.9844, showing that the model can clarify 98.44% of the data variation whereas only 1.23% of the overall differences are not clarified by the model. For a model to be suitable, the R^2 value must not be fewer than 0.75, especially in chemical field (Le Man *et al.*, 2010). Nevertheless, (Koocheki *et al.*, 2009) posited that a substantial value of R^2 does not always indicate that the regression model is a perfect one, and such suggestion could only be made depending on a similarly great value of adj R^2. The value of the adjusted estimation coefficient is Adj R^2 = 0.9704. Thus, this indicates that the model is greatly significant, confirming a good agreement among the experimental and expected values of SABA yield.

Table 4 ANOVA for response surface quadratic model.

Source	Sum of squares	DF	Mean square	F-value	P-value	
Regression model	451.56	9	50.17	70.12	0.0001	Significant
Linear	102.37	3	34.12	1.53	0.2446	
A	26.52	1	26.52	37.06	0.0001	
B	67.98	1	67.98	95.00	0.0001	
C	7.87	1	7.87	11.00	0.0040	
Square	341.46	3	113.82	159.06	0.0001	
A^2	183.71	1	183.71	256.73	0.0001	
B^2	230.16	1	230.16	321.64	0.0001	
C^2	23.87	1	23.87	33.35	0.0002	
Interaction	7.74	3	2.58	0.092	0.9608	
AB	1.41	1	1.41	1.97	0.1905	
AC	3.20	1	3.20	4.47	0.0605	
BC	3.13	1	3.13	4.37	0.0632	
Residual error	5.71	10	0.57			
Lack of fit	5.67	5	1.13	3.81	0.0844	Not significant
Pure error	1.49	5	0.30			
Total	458.72	19				

According to Rai (Rai *et al.*, 2016) , to be in good agreement, the adjusted R^2 and expected R^2 must be within 20%. This condition is satisfied in this analysis with an expected R^2 value of 0.8918. Thus, beyond the experimental range of process conditions, this model offers 89.18% variability in the prediction yield. The ANOVA of any quadratic model term is shown in Table 4. If the F-value is big and P <0.05, the term is significant. Based on Table 4, the linear terms A, B,

and C are significant, and only one quadratic term AB is not significant interaction. The other terms AC and BC have a positive significant interaction on the yield. In addition, the Lack of Fit is not important in comparison to the pure error which is good. Furthermore, a high F value (70.12) with a low probability (p=0.0001) indicates the high ability of the model in predicting the results.

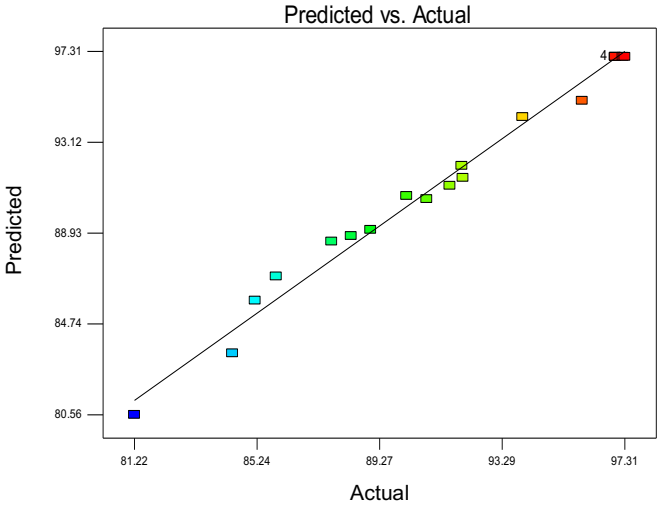


Fig. 1 SABA copolymer yield that was observed by the model in the experiment with expected values.

Fig. 1. appears the plot of expected yield by the improved model beside experimental values. The model was effective in capturing the correlation of process conditions due to the predicted values being very similar to the actual values.

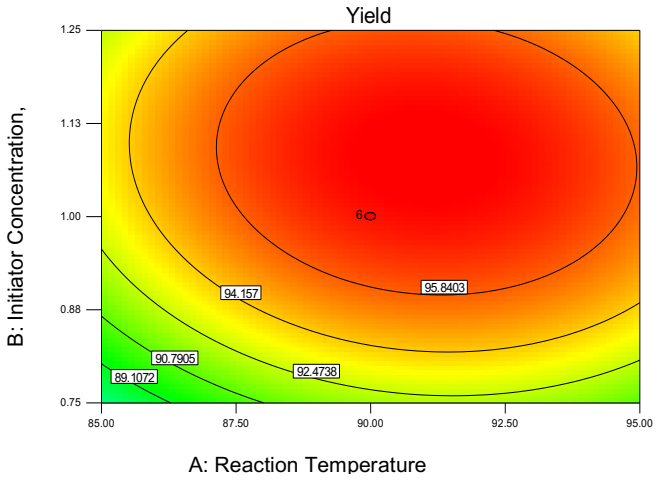


Fig. 2(a) Contour chart of % Yield vs. (A) reaction temperature, (B) initiator concentration.

Fig. 2(a) explains that rise in reaction temperature from 85 to 95 °C raised the initiator concentration from 0.75 to 1.25 wt %. There is a negative interaction between the concentration of initiator and reaction time. This indicates that the yield drops as the concentration of initiator and reaction temperature increase. Probably, there is an indication of a solvent reduction influence under this time at a specific reaction temperature, allowing the radicals generated to escape into the bulk medium and grow. While, Fig. 2(b) explains the combined influence of time and temperature on the yield, considering that the other parameters were constant throughout the reaction. Besides, Fig. 2(c) explains how the rise in the percentage of initiator concentration from 0.75 to 1.25 wt % affected the yield rate. Similarly, the time showed a clear influence on the yield; consequently, a rise in the time from 6.5 to 7.5 h, caused a significant raise in the yield percentage.

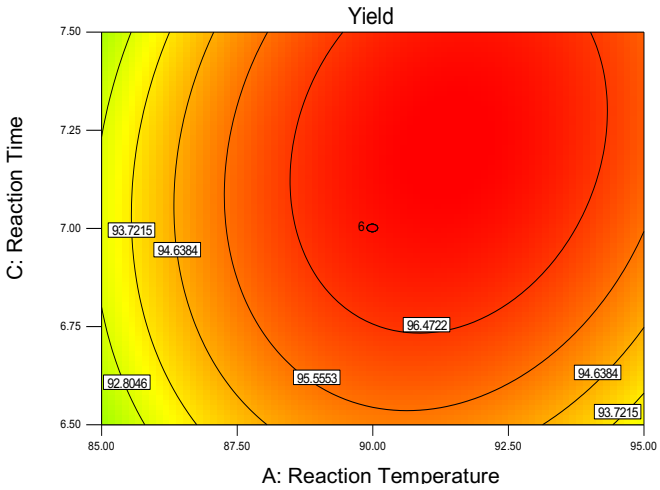


Fig. 2(b) Contour chart of % Yield vs. (A) reaction temperature, (C) reaction time.

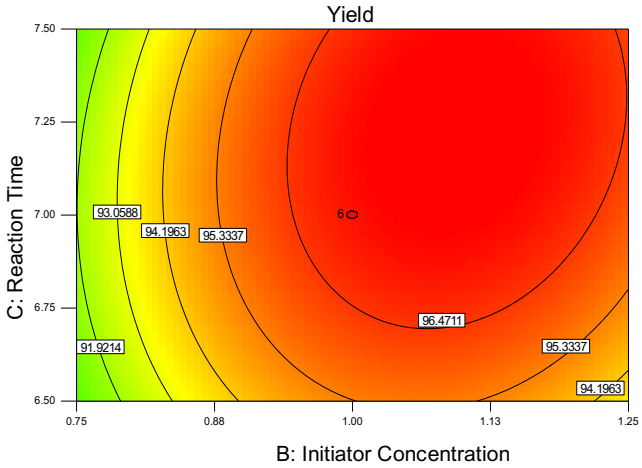


Fig. 2 (c) Contour chart of % Yield vs. (B) concentration of initiator, (C) reaction time.

Analysis of the response surface

Three-dimensional response surfaces have been designed to study the interaction between the factors and to estimate the optimum condition of each variable for maximum SABA copolymer yield. The influence of initiator concentration and temperature on yield at a stable reaction time of 7 h is shown below in Fig. 3(a). As the temperature and initiator concentration rise, the yield decrease. The optimum yield was 96.91% and which was obtained at about 90 °C and at an initiator concentration 1 wt %. In general, there is a negative interactive influence among the two process parameters. This is a weak confirmation of the dependence of yield on both of the reaction temperature and concentration of initiator.

The interactive influence of temperature and time on yield at a stable initiator concentration of 1 wt % is shown below in Fig. 3(b). The yield was appeared to rise with a rise in temperature as compared to that of reaction time. Thus, the highest yield has reached 96.95% at an optimal condition 7 h reaction time and 90 °C reaction temperature. Thus, there is a significant positive interaction among temperature and time. This displays that the yield increases with increasing reaction time as well as reaction temperature beyond the optimum value of 1 wt % concentration of initiator.

The interactive influence of initiator concentration and time on yield at a stable temperature of 90 °C is depicted in Fig. 3 (c). The

yield has been detected to rise with the rise in concentration of initiator as compared to that of the temperature. There is a positive interaction between the reaction time and concentration of initiator. However, the highest yield obtained was 97.31%. This shows that the yield rises with a rise in reaction time and concentration of initiator. In summary, the best result obtained for the yield from Figs. 3(a)(b)(c) was 97.31% and the important factors that have a further impact on polymerization synthesis are reaction time and temperature when compared with other factors..

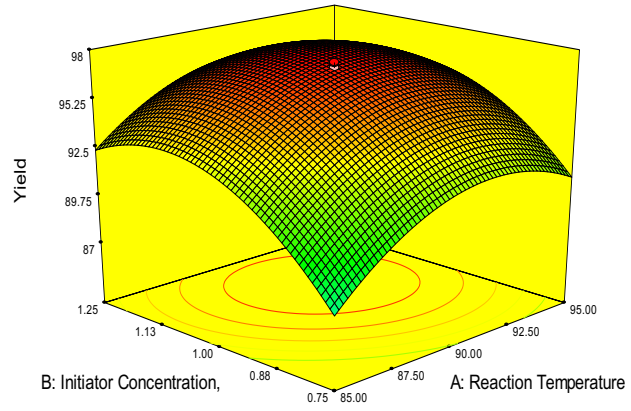


Fig. 3(a) Response surface plot of the interaction between reaction temperature and initiator concentration on percentage yield holding reaction time constant

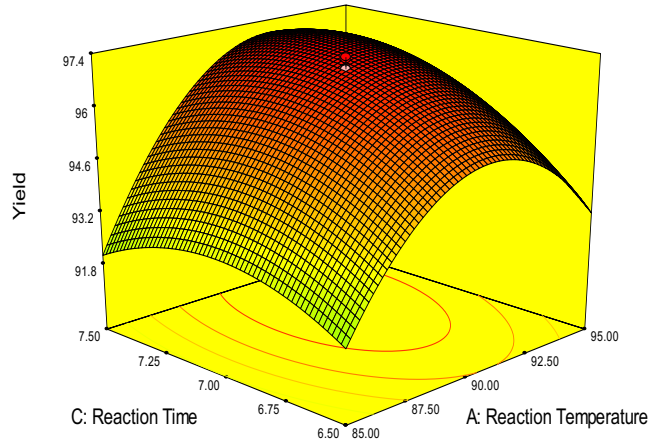


Fig. 3(b) Response surface plot of the interaction between temperature and time on percentage yield holding initiator concentration stable.

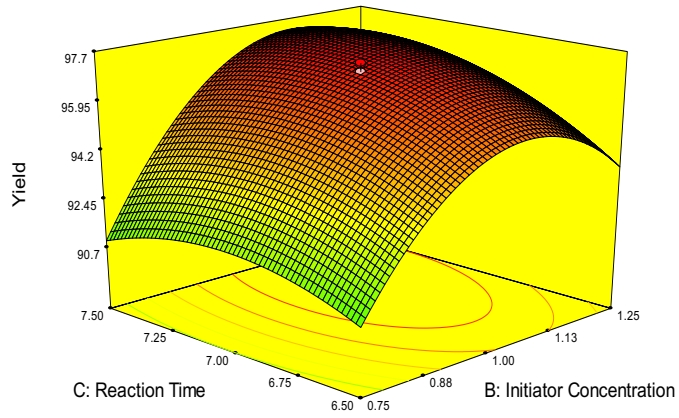


Fig. 3(c) Response surface plot of the interaction between initiator concentration and time on percentage yield holding temperature stable.

Optimization and validation

Optimization was performed using the RSM to achieve the optimal yield that satisfies all process conditions. The optimal operating conditions observed in this analysis were as follows: time of 7 h, temperature of 90 °C and initiator concentration of 1 wt %. The corresponding optimized yield is 97.31%.

Model validation was carried out using optimal replicate conditions by conducting three sets of experiments created from DOE software by comparing the experimental and expected values. The process of optimization was performed by inserting the desirable standards as shown in Table 5. The maximum average yield response obtained was set as the main goal, based on the performed experiments. The results indicate that all the experiments produce a yield response of less than 3% residual standard error (RSE). Hence, the model is validated; this model is expected to be accurate up to 97%. Therefore, the expected value and experimental value based on the model are firmly agreed upon, and the highest validated yield obtained in this analysis was 97.15%.

Table 5. Model validation experiments and expected data

No	Factors			Yield		
	A	B	C	Experimental Value	Expected Value	RSE %
1	95	1	7.5	97.02	95.64	1.42
2	90	1.25	8	96.76	94.46	2.37
3	90	1.25	7.5	97.15	96.20	0.97

CONCLUSION

RSM is a set of statistics and mathematical methods which enhance the empirical model building, developing, and optimizing processes factor. In addition, it can be used to discover the interaction of many affecting variables. The optimal conditions observed as follows from this analysis were: 90 °C of temperature, 7 h of time and 1 wt % concentration of initiator. The corresponding optimized yield is 97.31%. The analysis of variance (ANOVA) of the regression model detected an R^2 value of 0.9844, showing model could clarify 98.44% of the data variation, and just 1.23% of the overall variation were not clarified by the model. Validation was conducted using three replicate conditions and the highest average yield value obtained is 97.15%. There is an error of about 0.97 % as compared to the expected value. Therefore, it indicated that the model is extremely significant, that confirmed a stronge agreement among the experimental and the expected values of SABA yield.

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