

# Central Composite and Response Surface Method for Optimizing the Production of Nano Asphalt Rubber from Asbuton

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## Abstract

The number of cars continues to increase due to the worldwide economic growth. This gives an impact not only on the condition of the roads, but also on the accumulation of tire waste. Research on improving the quality of road asphalt by utilizing crumb rubber (CR) from car tires has been carried out by many researchers and has given positive results. However, each researcher suggests a different composition of CR to get the best quality of asphalt rubber. This study provides an overview and prediction regarding the efficiency of Nano Asphalt Rubber (NAR) production by using Central Composite Method (CCM) and Response Surface Method (RSM). This work can give an idea to the researcher or decision maker of how much CR should be added in asphalt mixture to get the optimum result, especially when using Asbuton as the bitumen source. The results of the optimization using the CCM and RSM in Minitab® i.e. regression equations, contour plot and surface plots, can be used to see the Yield range of AR production. The NAR production is carried out by extracting the Asbuton (in-situ) with ultrasonication technique and mixing it with CR according to a 5-factors full factorial design. The result shows that the highest NAR Yield in the optimization process can reach 99.9%.

**Keywords:** Asphalt Rubber • Optimization • Asbuton • Crumb Rubber

## Introduction

The economic growth of any community generally in line with the increase of the road usage, due to the increasing number of cars. This will cause the excessive road loads that lead to the road damage. For this reason, the quality of the asphalt mixture needs to be improved so that the asphalt can handle the heavy load of the vehicles. On the other hand, an increase in the number of cars also results in an increase in tire disposal which in turn creates a pollution problem. This is because tires are not easily degraded and can even last up to 100 years in the landfill [1]. Especially in China and Indonesia, rubber and plastic waste continues to increase and accumulate so that it can have an adverse impact on environmental health as well as on the local and global economy.

Asphalt is a binder that plays an important role in determining whether a road is strong or not. Increasing the quality of the asphalt can be done by adding various kinds of added ingredients to the asphalt mixture. It has been reported that the effect of adding crumb rubber to the asphalt binder can result in increased resistance to the problem of rutting, fatigue cracking, and cracking at low temperatures [2-6]. Crumb rubber (CR) can be produced by grinding the tires into very small particles. Mixing asphalt with crumb rubber from used vehicle tires not only improves the quality of the asphalt, but also helps solve the waste problem so that sustainable development can be achieved.

Mixing asphalt with aggregate on road paving needs to be designed properly because this can significantly affect the stiffness of the pavement. Although the addition of CR to asphalt paving mixes has been investigated during the last decade, however every researcher suggests different %CR to be added to the asphalt mixture in order to improve the quality of asphalt. It can be concluded that the researchers made the variation of CR between 0 – 50% and each one of them claimed different optimum %CR [2, 6-9].

Although many studies have shown an improvement in the quality of road asphalt by utilizing crumb rubber (CR) from car tires, most of these studies use petroleum asphalt in their experiments. In line with the recommendation of the Indonesian government, asphalt from Asbuton rock needs to be utilized. Asbuton (Asphalt from rocks on Buton Island) is one of the natural resources which is abundantly available in Indonesia. The existence of this asphalt deposit is both an opportunity and a challenge for researchers, practitioners and all

parties related to road pavement. Asbuton is the largest natural asphalt deposit in the world, and it can be used as a binder on road pavements to replace petroleum (oil) asphalt. As a challenge, the use of Asbuton as a binder on road pavement is not as simple or as easy as using oil asphalt. However, in principle the researchers have shown that Asbuton can be used on road pavements even though there are still some obstacles in its implementation [10].

As already mentioned, the use of Asbuton or CR as additives in the pavement mixture will give a different effect on the properties of the asphalt mixture. The nature of the mixture containing pure Asbuton bitumen has increased resistance to permanent deformation but reduces the resistance to fatigue cracking, along with the addition of pure Asbuton levels [11]. While the 60/70 penetration oil asphalt containing used tire powder will have better resistance to permanent deformation and fatigue cracking when compared to the 60/70 pen oil asphalt itself at test temperatures of 58, 64 and 70 °C [12]. In that study, it is also shown that Performance Grade (PG), both Asbuton and Asphalt Rubber modified asphalt produces a greater PG value than pure 60/70 pen asphalt. They reported that the use of Asbuton or CR provides additional strength. In their experiment, modified Asbuton asphalt was made by mixing 60/70 pen asphalt and pure Asbuton with a predetermined percentage of pure Asbuton content, namely 2%, 4%, 6%, 8% and 10%. The asphalt used is purely obtained from the extraction of Asbuton Lawele. Analysis of mechanistic rheological parameters and pavement damage criteria for Asbuton modified asphalt was carried out at the optimum Asbuton content which is 10%.

Asphalt Rubber (AR) has been made by mixing 60/70 pen asphalt and used tire powder with different percentage levels of CR from waste tire. The mixing process is carried out when the base asphalt temperature varied in the range of 177 °C – 204 °C. The used tire

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powder is added and stirred in such a way so that a homogeneous mixture occurs. This mixing process is carried out for 1 hour, with the aim of providing sufficient reaction time for used tire powder and asphalt base [4, 13, 14].

One of the obstacles when using Asbuton in asphalt mixture is the hardness of Asbuton which is very different from the 60/70 pen oil asphalt. The level of hardness Asbuton can be classified to hard asphalt with a penetration value of 25 °C. Under these conditions, Asbuton can only be used as an additive in 60/70 pen oil asphalt with a maximum percentage of 10%. From a series of studies and analysis that have been carried out, it was concluded that the addition of CR by 16% is the best composition in increasing the mechanical properties of rheology and reducing the damage criteria [12].

This study is about the in-situ preparation of NAR, where the asphalt directly extracted in the process from Asbuton. The percentage of CR added in the mixing process are varied between 0 – 50 % (or CR fraction of 0 – 0.5), therefore this study can provide a course to predict the Yield of AR production, especially for the in-situ preparation. This can help the decision maker to determine how much CR should be added into the asphalt mixture in order to make an efficient production of NAR. Although the process might not be the same, all have the same principle in preparation. With optimization using the CCM and RSM, regression equations and surface plots can be made to see the range in which the maximum NAR-Yield production that can be generated. The aim of this study is to provide the way to predict the Yield of the experiment when one uses a certain variation of % CR in the NAR preparation.

## Method

### Material

The materials that were used in this research include Asbuton rock (from Lawele) as the source of asphalt, diesel oil as the solvent, lecithin (HLB = 4) and SPAN 85 (HLB = 1.8) as the surfactant, and crumb rubber (150-3000µm) as an additive (Figure 1).



Figure 1. Crumb Rubber (left) and the Asbuton particles (right)

### Formation of Nano Asphalt Rubber (NAR)

The steps in the in-situ preparation of Nano Asphalt Rubber (NAR) from Asbuton and CR with the ultrasonication process is shown in Figure 2. In principle, the method of making NAR comprises of 4 stages, namely the reduction of the size of Asbuton rocks, extraction, separation and washing. At the stage of reduction in size, Asbuton rocks were grinded and mashed by using mortars to an average size of 200 micrometers. The scoured particles are then filtered with a 70 mesh filter so that the coarse particles can be collected and returned to the mortar to be refined. Asbuton fine grains measuring <200 micrometers are collected in a container until the amount of weight as needed. For each experiment, 61.14 g of fine Asbuton granules are taken and put in a 200 ml beaker. Then into another 200 ml beaker, crumb rubber, surfactant and diesel oil (as the solvent) are prepared. The amount of solvent is determined by definition of oil fraction ( $\alpha$ ) which is equal to  $(m_{\text{solvent}} + 30\% m_{\text{Asbuton}}) / (m_{\text{solvent}} + m_{\text{Asbuton}})$ . In this calculation, we assumed

that the content of asphalt in Asbuton is 30%, which is according to the literature [15]. The amount of surfactant added to the asphalt mixture is only 0.05%-wt. Fine Asbuton (<200 micrometers) is slowly inserted into a beaker containing a mixture of surfactant, CR and solvent while stirring with spatula slowly until it appears that the whole mixture of dough has blended well. The mixture is then closed and will be processed later by sonication. After being mixed and heated during the sonication, the mixture is allowed to cool down for then separated between the upper phase which is called NAR, and the lower phase which is called the residue. The experiments were carried out according to the 5 factors of full factorial design ( $2^5$ ) which is shown in (Table 1).

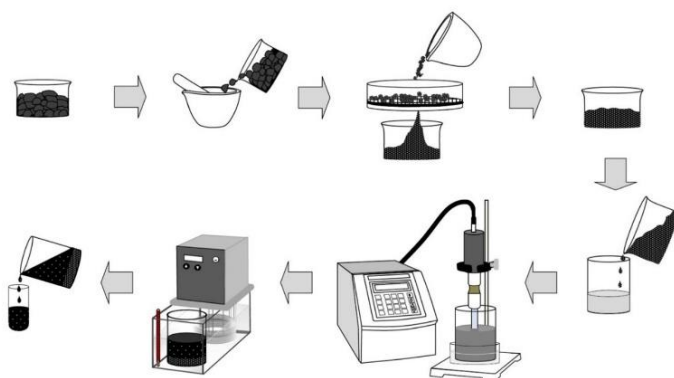


Figure 2. Steps in the in-situ extraction of asphalt for producing Nano Asphalt Rubber (NAR) from Asbuton and mixing with CR by using the ultrasonication process

Table1. Design factors of the experiments

Level	HLB Surfactant (A)	Asbuton particle size, µm (B)	Oil Ratio, $\alpha$ (C)	Crumb Rubber Fraction (D)	Ultrasonication time, min (E)
Low (-)	1.8	300	0.42	0	15
High (+)	4.3	700	0.5	0.5	45

### Optimization Procedure

The most influential factors in the in-situ production of NAR can be analyzed by using the Full Factorial Design ( $2^k$ ) technique. The number of experiments to be conducted according to the factors that are shown in the Table 2 are 32 in total. The results of experiments later will be processed by CCM and RSM method to find the optimum value. The important principle of RSM in the experimental design is the selection of points on which responses should be evaluated. These points are realized by using the CCM. Generally, the mathematical models produced from RSM are polynomial, with an unknown structure. The RSM in Minitab® software will optimize the polynomial equation by finding the peak point from the factors. The output of the optimization will be depicted in the graphs of surface plot and contour plot.

## Result and Discussion

The response of experiments that have been carried out based on the  $2^5$  full factorial design (in Table 1) is presented in Table 2. From this result, regression was completed using the DOE technique and produced a regression equation with uncoded units shown in the following equation:

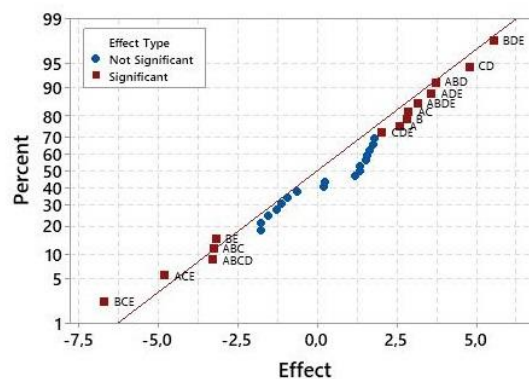
$$\text{Yield} = 520.1 - 92.63 A - 0.5699 B - 1055 C + 217.3 D - 13.22 E + 0.08764 A*B + 210.6 A*C - 10.89 A*D + 3.116 A*E + 1.378 B*C - 0.2497 B*D + 0.01800 B*E - 339.7 C*D + 30.28 C*E + 6.998 D*E - 0.1987 A*B*C - 0.04667 A*B*D - 0.003269 A*B*E + 25.10 A*C*D - 6.680 A*C*E - 6.194 A*D*E + 0.4237 B*C*D - 0.04091 B*C*E - 0.02123 B*D*E - 16.65 C*D*E + 0.05670 A*B*C*D + 0.006655 A*B*C*E + 0.01257 A*B*D*E + 12.47 A*C*D*E + 0.04304 B*C*D*E - 0.02366 A*B*C*D*E$$

**Table2.** Response (Yield of NAR) in the experiments using factorial design 2<sup>5</sup>

Run	A	B	C	D	E	Yield	Yield(*)
						(%)	(%)
1	1.8	300	0.42	0	15	74.93	75.08
2	4.3	300	0.42	0	15	73.98	74.09
3	1.8	700	0.42	0	15	81.19	81.43
4	4.3	700	0.42	0	15	77.31	77.51
5	1.8	300	0.5	0	15	56.82	56.99
6	4.3	300	0.5	0	15	72.01	72.14
7	1.8	700	0.5	0	15	81.82	82.1
8	4.3	700	0.5	0	15	86.19	86.42
9	1.8	300	0.42	0.5	15	85.69	85.84
10	4.3	300	0.42	0.5	15	72.6	72.71
11	1.8	700	0.42	0.5	15	74.13	74.37
12	4.3	700	0.42	0.5	15	66.42	66.64
13	1.8	300	0.5	0.5	15	65.66	65.84
14	4.3	300	0.5	0.5	15	80.98	81.12
15	1.8	700	0.5	0.5	15	81.36	81.65
16	4.3	700	0.5	0.5	15	82.24	82.49
17	1.8	300	0.42	0	45	76.22	76.46
18	4.3	300	0.42	0	45	87.91	88.08
19	1.8	700	0.42	0	45	81.99	82.38
20	4.3	700	0.42	0	45	76.53	76.86
21	1.8	300	0.5	0	45	81.07	81.35
22	4.3	300	0.5	0	45	80.82	81.01
23	1.8	700	0.5	0	45	77.82	78.26
24	4.3	700	0.5	0	45	68.48	68.85
25	1.8	300	0.42	0.5	45	68.32	68.6
26	4.3	300	0.42	0.5	45	61.61	61.83
27	1.8	700	0.42	0.5	45	65.79	66.22
28	4.3	700	0.42	0.5	45	89.7	90.12
29	1.8	300	0.5	0.5	45	78.37	78.69
30	4.3	300	0.5	0.5	45	88.1	88.36
31	1.8	700	0.5	0.5	45	75.54	76.04
32	4.3	700	0.5	0.5	45	83.63	84.12

(\*) Predicted Yield of NAR production without using surfactant in the process

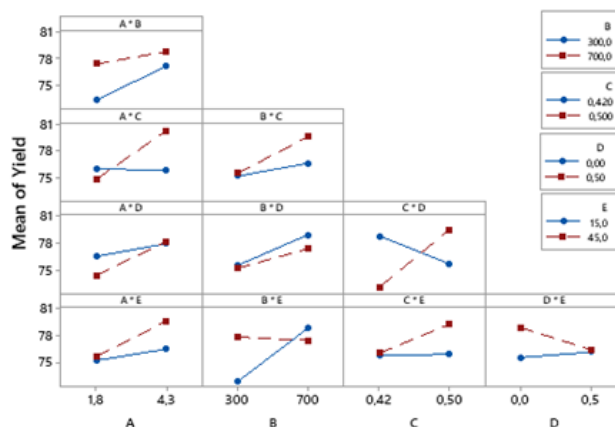
Where: A = HLB, B = Asbuton particle size, C = Oil Ratio, D = CR fraction, and E = sonication time. By this equation, we can also predict how much the Yield of NAR in a certain condition of the experiment. For example when we remove using of the surfactant in the process, we can predict Yield of the NAR by setting the value of factor A to zero in the equation. The result of Yield when putting the factor A to zero can be seen in Table 2. We can notice that there is not so much difference on the Yield (\*) because the amount of surfactant that was used in the experiment is very low, i.e. 0.5% wt. However, it is interesting to see that according to the simulation, the Yield is somewhat increase without using the surfactant. This may indicate that the extraction process is better without helping of the surfactant. Figure 3 shows the normal plot of the effect of all the factors including their interactions.



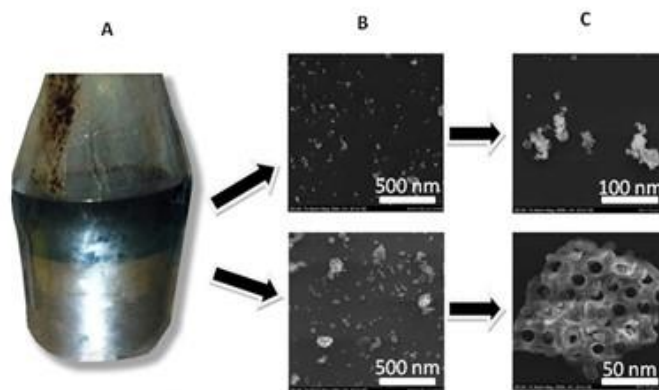
Lenth's PSE = 2,67956

**Figure 3.** Normal plot of the effect of all the factors including their interactions

It can be seen from Figure 3 that the interactions between the factors have more significant influence to the Yield than the individual factors. The interactions between the factors are depicted in Figure 4. It can be seen that the strongest interaction is ensued between the factors C (oil ratio) and D (CR fraction). The high gradients of the intersecting lines in the C: D interaction plot indicate that the effect of solvent amount (C) on the Yield strongly depends on the level of the CR fraction (D) and vice versa. As mentioned before, this effect can only be observed when both factors C and D are varied at the same time.

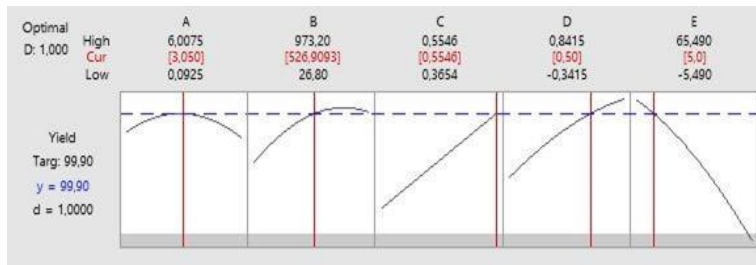


**Figure 4.** Interaction Plot for Yield of NAR production



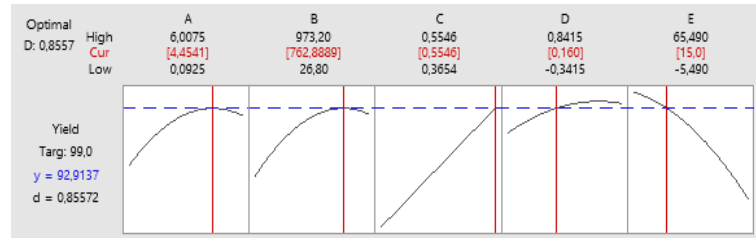
**Figure 5.** NAR in the upper layer (left) and the SEM images of nanoparticles in the NAR (B and C upper) and the residue of Asbuton rock (B and C bottom) in different magnifications

In the optimization process of 2<sup>5</sup> factorial, the CCM produces 54 runs because it makes 32 cube points, 8 center points in cube, 10 axial points and 4 center points in axial. The result of the optimization by using the RSM is presented in Figure 6. In this work, the range of the CR fraction is varied from 0 – 0.5. From Figure 6, it can be seen that the optimum Yield of 99.9% can be found in the NAR production when using HLB (A) = 3.05, particle size (B) = 526.90 μm, oil ratio (C) = 0.55, CR fraction (D) = 0.5, and sonication time (E) = 5 min.



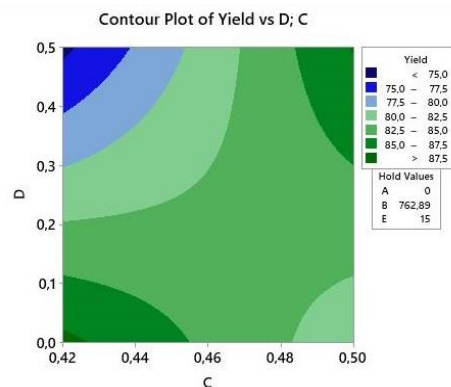
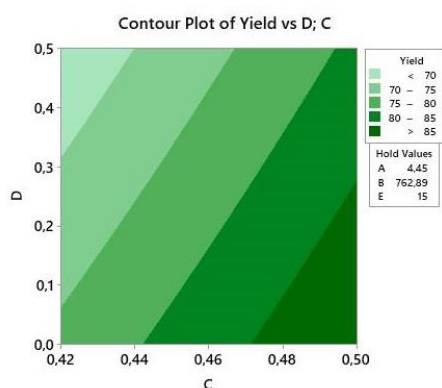
**Figure 6.** RSM of the Yield optimization in NAR production when using CR fraction 0 – 0.5. The optimization can reach Yield of 99.9 % according to the set target.

As an example of application, here we will use the best CR of 16% according to Indriani [12], because we have similar situation, which is using Asbuton as asphalt source and sonication in the process. She claimed that the type of 16% CR-asphalt is strong against the criteria of Fatigue Cracking damage at 22 °C with a value of |G \*|.sin δ of 122.7828 kPa. As can be seen in Figure 7, the maximum Yield that can be reached is 92.91% although the target of optimized Yield has been set to 99%. To give a clear picture of the simulation, the contour plots (Figure 8) and surface plots (Figure 9) can depict the Yield of NAR when we vary the CR fraction.



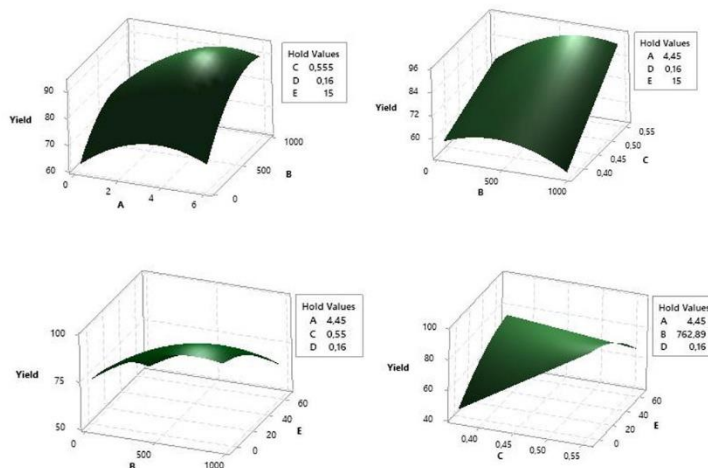
**Figure 7.** RSM of the Yield optimization in AR production when using 16 %CR (D = 0.16). The optimization reaches Yield of 92.9 % although the set target is 99.9%

If the experiment still follow the optimum Yield in Figure 7, that is using 16% CR (D = 0.16), we can see more details that the region of high Yield is in the range of 0 – 25% CR and oil ratio of 0.47 - 0.5 (Figure 5 left). The right-hand side of Figure 5 shows the Yield of NAR when in the experiment, no use surfactant is used in the extraction process. It can be seen that the regions of high Yield are in the range of 0 – 12% CR and oil ratio of 0.42 – 0.45, also in the region of 30 – 50% CR and oil ratio of 0.49 – 0.50.



**Figure 8.** Upper: Contour plot of the Yield in NAR production when using the factors A, B, and E that are hold according to value in Figure 7. Bottom: Contour plot of the Yield in AR production when using no surfactant (A = 0)

Surface plot also can be used as a means to expect the response or the result of an experiment. In the case of the in-situ production of NAR by using Ultrasonication of Asbuton and 16% of CR, the Yield NAR can be predicted according to the surface plot depicted in Figure 9. The fixed parameters (hold values) in each plot in Figure 9 are according to the optimum factors in the Figure 7.



**Figure 9.** Surface Plots of the Yield in the NAR production when using 16% of CR

## Conclusions

The in-situ NAR production from Asbuton and CR with ultrasonication method has been demonstrated. According to the analysis through the response surface of factorial design, it is found that the strongest interaction transpired between the solvent ratio and the CR fraction. This implies that the change of solvent amount will affect the influence of % CR on the Yield of NAR, and vice versa. The optimization process has been simulated by using CCM and RSM. The result of optimization can reach the NAR Yield of 99.9%. The optimum Yield of NAR can be simulated by changing the value of each factor. The expected Yield can be found through the Contour Plot and Surface Plot methods.

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