

Research Article

Recycling of Waste PET Plastic on Asphalt, by Using Application of Response Surface Methodology: Effect of Production Process Parameters

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Abstract

Plastic, polymers of variable compositions have become object of common use and difficult to digest by micro-organisms, especially single-use plastic waste, such as polyethylene terephthalate (PET) water canisters. These polymers are source of environmental pollution. Therefore, it is important to manage theme in the good way to protect environment. In this study, different conditions of waste plastic PET (Polyethylene Terephthalate) recycling on asphalt were optimized. Response Surface Methodology (RSM) using the Doehlert experimental design has been employed in the optimization. The independent variables considered were bitumen (5-8%), PET (0-12%), Mixing temperature (150-160 °C) and Mixing time (20-30min). Four-second order polynomial models were generated. The responses obtained by the models were well described as: specific density (Y_{SD}), penetrability (Y_P), softening point (Y_{SP}), and flash point (Y_{FP}) of the process with satisfactory fits in terms of absolute average deviation, bias factor and accuracy factor. The optimum responses were 1,04 as specificity gravity (Y_{SG}), 60*(1/10mm) as penetrability at 25 °C, 100g and 5sec (Y_P), 50 °C as softening point (Y_{SP}), and 242 °C as flash point (Y_{FP}). The statistical relation between the four independent variables and the process responses were well described.

Keywords

Optimization, RSM- Doehlert Experimental Design, Blending Condition, Recycling, Waste Plastic, PET, Asphalt Modified, Physicochemical Characteristics

1. Introduction

Plastic, polymers of variable compositions have become object of common use and difficult to digest by micro-organisms. These polymers are a source of environmental pollution [10]. Chad is no exception from this type of pollutant due to the lack of a reliable household waste management

policy, waste plastic made up of polyethylene terephthalate is thrown into the environment with no lesser consequences on the ecosystem [13]. One of the best and most effective methods of managing of waste plastic is the possibility of incorporating it into construction processes [9]. In general

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case, to recycle this waste, two processes emerge: the dry method (coating plastic on the hot aggregate and then mixing with asphalt) and the wet method (mixing powdery plastic waste with asphalt then mixing with aggregate) [21]. Through these processes, PET are easily recyclable because of their high melting point (about 260 °C). The latter, is one of the characteristics which determines the choice of process considered in relation to the types of plastic waste to be recycled [7]. The wet process is better for controlling the properties of the modified asphalt binder. Although this process requires specialized mixing and storage facilities [19]. Also, due to its enhanced thermal behavior, the wet process is currently the most widely used for polymer asphalt modification. Composed of complex molecules, the adhesion of plastic waste into bitumen requires a specific time, generally around 20 to 30min [6]. Likewise, in order to obtain perfect homogenization, the size of plastic plays an important role. It was suggested to use plastic size less than 2,36mm and the optimum asphalt ratio should be up to 10% [5]. Despite the fact that plastic waste improves asphalt properties, this process has some limitations to explain the influences of the raw materials (waste PET and asphalt) on the best optimum condition.

Thus, the present article is devoted to the study of the optimization conditions of recycling waste plastic PET on asphalt by using application of response surface methodology. The aim of process is to identify relation between the in-put parameter (factor) and their effect on the responses (out-put). This can help to choose the good factor for a specify researched characteristic.

2. Materials and methods

2.1. Material

The materials used within the scope of this article are:

1. Waste PET powder size 0 to 0,63mm
2. Asphalt with grade 60/70; 1,03g/cm³ as specific density,
3. 46 °C as softening point and 235 °C as flash point.

To obtain PET on powder form, the plastic bottle was washed, dried and melted about 260 °C which is PET melting point, then pouring at ambient air. After solidifying, the plastic was grinded and sifted. The plastic with granulometry included 0-0,63mm were used to mix with hot bitumen as shown in Figure 1. The suggested ratio of bitumen was 5 to 8% [8]. Wet process of recycling of waste plastic was done here. It is constituted by a mixture of PET plastic powder and the bitumen binder [21].

2.2. Mixing Condition

According to literature review, plastic waste can be used in

hot mix to improve physical properties of bituminous, aggregate and mix by 'Dry Process' or 'Wet Process'. As we need to determine, the influence of some factors which can influence the tests analyses, we opt for wet process. Because of chemical characteristics of asphalt and PET, this method can give more information about optimization condition of recycling waste plastic on asphalt. The process was done by blending PET powder and asphalt. To get mix binder, PET obtained by substitution of asphalt in different ratio (0-12%) was added to hot asphalt. The mix was stirred for 20 to 30min with temperature about 150 °C to 160 °C as shown in figure 1. This process was intended to make some physicals analyses and to find out a good optimum condition of waste PET recycling on asphalt by using Doehlert response surface methodology.

2.3. Response Surface Methodology (RSM)

Optimization studies using response surface methodology (RSM) were deeply investigated [14]. Furthermore, the optimum conditions could assist designers to manufacture simple unit operations that could limit or eliminate the tedious practice of recycling PET on asphalt (rate of waste PET, asphalt, mixing temperature and time). Then absolute average deviation (AAD) and coefficient of determination (R^2) could draw to investigate the adequacy of the proposed models. The present study involves optimization of some parameters that are likely to affect asphalt which can be used for asphalt concrete formulation.

The general practice of determining these optima is by one variable-at-time approach. One of the disadvantages of this approach is that it does not include interaction effects among the variables and is unable to determine the true optimum conditions. In order to overcome this problem, optimization studies were done using response surface methodology. RSM is a collection of mathematical and statistical technique that is useful for modeling and analyzing situations in which a response of interest is influenced by several variables, especially if there is a need to optimize the responses of a process. Doehlert matrix as an experimental design represents a uniform distribution of experimental points in space of coded variables as shown on table 1.

It is used particularly when there is a need to cover an experimental domain of any form of uniformly distributed points in order to explore the total domain (margins and interiors). Moreover, it permits to follow in a sequential manner in studying a response surface of second degree. Polynomial equations with and without interaction could be proposed as models for the mentioned processes. A few studies have been reported on the recycling of waste plastic on asphalt.

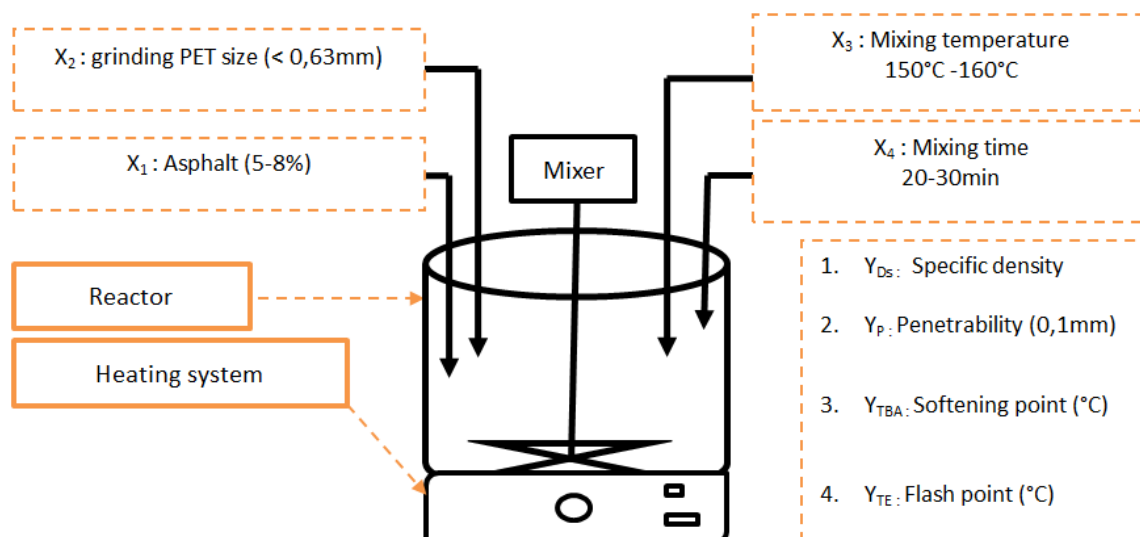


Figure 1. Schematic of recycling process of waste PET.

It seemed to be important to study the rate of optimal PET, obtained by asphalt substitution and the good mixing temperature and time which define the best binding. The objective of this study was to evaluate the influence of asphalt, PET, mixing temperature and time on asphalt modified, which can

be used to make concrete asphalt formulation. The second preoccupation was to assess the good binding condition to obtain the best properties in terms of the response functions: specific gravity (Y_{SD}), penetrability (Y_P), softening point (Y_{SP}), and flash point (Y_{FP}).

Table 1. Doehlert experimental design of four independent variables employed to recycle PET.

Tests number	Asphalt (%) $x_1'(X_1)$	PET (%) $x_2'(X_2)$	Mixing Temperature (°C) $x_3'(X_3)$	Mixing time (Min) $x_4'(X_4)$
1	7 (0.000)	6 (0.000)	155 (0.000)	25 (0.000)
2	8 (1.000)	6 (0.000)	155 (0.000)	25 (0.000)
3	5 (-1.000)	6 (0.000)	155 (0.000)	25 (0.000)
4	7 (0.500)	11 (0.866)	155 (0.000)	25 (0.000)
5	6 (-0.500)	1 (-0.866)	155 (0.000)	25 (0.000)
6	7 (0.500)	1 (-0.866)	155 (0.000)	25 (0.000)
7	6 (-0.500)	11 (0.866)	155 (0.000)	25 (0.000)
8	7 (0.500)	8 (0.289)	159 (0.816)	25 (0.000)
9	6 (-0.500)	4 (-0.289)	151 (-0.816)	25 (0.000)
10	7 (0.500)	4 (-0.289)	151 (-0.816)	25 (0.000)
11	7 (0.000)	9 (0.577)	151 (-0.816)	25 (0.000)
12	6 (-0.500)	8 (0.289)	159 (0.816)	25 (0.000)
13	7 (0.000)	3 (-0.577)	159 (0.816)	25 (0.000)
14	7 (0.500)	8 (0.289)	156 (0.204)	29 (0.791)
15	6 (-0.500)	4 (-0.289)	154 (-0.204)	21 (-0.791)
16	7 (0.500)	4 (-0.289)	154 (-0.204)	21 (-0.791)
17	7 (0.000)	9 (0.577)	154 (-0.204)	21 (-0.791)
18	7 (0.000)	6 (0.000)	158 (0.612)	21 (-0.791)

Tests number	Asphalt (%) $x_1'(X_1)$	PET (%) $x_2'(X_2)$	Mixing Temperature (°C) $x_3'(X_3)$	Mixing time (Min) $x_4'(X_4)$
19	6 (-0.500)	8 (0.289)	156 (0.204)	29 (0.791)
20	7 (0.000)	3 (-0.577)	156 (0.204)	29 (0.791)
21	7 (0.000)	6 (0.000)	152 (-0.612)	29 (0.791)

x : coded value of variables and X : the real value of variables

2.4. Physicochemical Analysis of Asphalt Binder

2.4.1. Determination of Specific Gravity

This test, according to NFT 66-007, is defined as the ratio of the mass of a given volume of a material to equal volume of water. The specific gravity is one of the fundamental properties of bitumen binder. It informs us about the mineral impurity which can be present in bitumen specimen. Specific gravity is going with Archimedes principle: If a solid material is first weighed in air and then weighed after immersing it in water, then the difference in the two weights gives the volume of water displaced by the solid material.

Generally, pycnometer was used to calculate the density as the following formula:

$$SG = \frac{(C-A)}{(B-A)-(D-C)} \quad (1)$$

With;

A : Weight of pycnometer

B : Weight of pycnometer filled with water

C : Weight of pycnometer part filled with bitumen

D : Weight of pycnometer + Bitumen + Water

2.4.2. Determination of Penetrability

This test is used to get information about consistency and the grade of asphalt by penetration test (NFT 66-004). It gets information about the hardness or softness of asphalt by measuring the depth in tenths of a millimeter to which a standard loaded needle will penetrate vertically in 5 seconds under specified temperature, load and duration of loading.

2.4.3. Determination of Softening Point

The softening point of bitumen or tar is the temperature at which the substance attains a particular degree of softening. It can be defined also as the temperature at which a bitumen can no longer support a steel ball with 3,5g as the weight and fall at which a standard ball passes through a sample of bitumen in a mold and falls through a height of 2.5 cm in certain condition. This test helps to know the temperature up to which a bituminous binder should be heated for various road use applications. Softening point is determined by ring and ball apparatus (NFT 66-008).

2.4.4. Determination of Flash Point

Depending of asphalt grades, this hydrocarbon leaves out volatiles, mostly at high temperatures. This fact can be characterized by a flash point. According to NFT 66-118, this test is defined as the lowest temperature at which the vapor of bitumen momentarily catches fire in the form of flash under specified test conditions. This test provides information on the level of light components in a given mixture.

Validation and Optimization of waste PET recycling

2.5. Validation

To express the fit of second-degree equations, the determination coefficient R^2 was used. This coefficient of determination was insufficient for model validation on its own. The absolute average deviation (AAD) was required to validate a model, as was the use of the bias factor and the accuracy factor. As a result, the model validation criterion was calculated using the formulas:

$$AAD = \frac{\sum_{i=1}^n \left(\frac{|Y_{i,exp} - Y_{i,theo}|}{Y_{i,exp}} \right)}{n} \quad (2)$$

$$B_f = 10^{1/n \sum_{i=1}^n \log \left(\frac{Y_{i,theo}}{Y_{i,exp}} \right)} \quad (3)$$

$$A_f = 10^{1/n \sum_{i=1}^n \log \left(\frac{Y_{i,theo}}{Y_{i,exp}} \right)} \quad (4)$$

Where:

AAD , absolute average deviation; B_f , bias factor; A_f , accuracy factor; Y_i , Theo, response obtained using the model; $Y_{i,exp}$, response obtained via experiment and n , number of trials.

The acceptable values of those applications must be within the following ranges: AAD , 0-0.3; B_f , 0.75-1.25, and A_f , 0.75-1.25.

2.6. Optimization Condition

The response surface methodology using Doehlert experimental matrix was used to optimize recycling of PET on asphaltic road. Minitab version 19, Sigma Plot version 14 and Excel, were used for statistical analysis, regression models and graphical optimization. Besides, the fit of models was verified by the coefficient of determination (R^2), the absolute

average deviation (AAD), B_f (Bias Factor) and A_f (Accuracy Factor). Four independent variables namely Asphalt (X_1 : 5-8%), PET (X_2 : 0 – 12%), Mixing temperature (X_3 : 150-160 °C) and the Mixing time (X_4 : 20-30min) were chosen. The range of independent parameters were selected based on literature review and preliminary studies. Twenty-one different experiments were presented in according to the experimental design for the four parameters. The experiments were figured in coded (x) and real (X) values.

The response functions (Y_i) measured were: specific gravity (Y_{SG}), penetrability (Y_p), softening point (Y_{SP}), and flash point (Y_{FP}). These were related to the coded values (x_i) by the second order polynomial that shown in equation (5).

$$Y_i = b_0 + \sum_{i=1}^k b_i x_i + \sum_{i=1}^k b_{ii} x_i^2 + \sum_{i=1}^{k-1} \sum_{j=i+1}^k b_{ij} x_i x_j \quad (5)$$

The coefficients of the polynomial were represented by b_0

(constant term), x_i (linear effects), x_{ii} (quadratic effects) and x_{ij} (interaction effects). X_i and X_j are the independent variables. The analyses of variance were generated and the effect and regression coefficients of individual, quadratic and interaction terms were determined. The significances of all terms in the polynomial were judged statistically at a probability (P) of lower than 0.05 ($P < 0.05$). The regression coefficients were then used to make statistical calculation to generate contour map and response surface graphs from the regression models.

2.7. Recycling Procedure

In each experiment of recycling of used PET plastic in asphalt, 500g of mixed of the cooked sample were used to get information about: Gravity specific at 27 °C, Penetrability at 25 °C, 100g 5sec in 1/10mm, Softening point (°C) and Flash Point (°C).

Table 2. Influences of Asphalt and PET ratio, mixing temperature and mixing time on the responses of modified binder.

N°	Asphalt (%) X_1	PET (%) X_2	Temperature (°C) X_3	Time (min) X_4	Specific Density	Penetrability at 25 °C, 100g 5sec (1/10mm)	Softening point (°C)	Flash point (°C)
1	6,50	6	155	25	1,06	58	48	245
2	8,00	6	155	25	1,06	58	48	245
3	5,00	6	155	25	1,06	58	48	245
4	7,25	11	155	25	1,10	44	55	258
5	5,75	1	155	25	1,03	67	46	235
6	7,25	1	155	25	1,03	67	46	235
7	5,75	11	155	25	1,10	44	55	258
8	7,25	8	159	25	1,08	53	50	252
9	5,75	4	151	25	1,05	60	48	243
10	7,25	4	151	25	1,05	60	48	243
11	6,50	9	151	25	1,10	52	54	255
12	5,75	8	159	25	1,08	53	50	252
13	6,50	3	159	25	1,04	66	46	240
14	7,25	8	156	29	1,08	52	53	254
15	5,75	4	154	21	1,04	62	47	236
16	7,25	4	154	21	1,04	62	47	236
17	6,50	9	154	21	1,07	57	53	248
18	6,50	6	158	21	1,05	60	47	240
19	5,75	8	156	29	1,08	52	53	254
20	6,50	3	156	29	1,05	62	47	242
21	6,50	6	152	29	1,09	55	49	246

3. Results and Discussion

The influence of operating parameters (Bitumen, PET, Mixing Temperature and Mixing Time) on the recycling of PET in the asphalt was determined. The findings were provided in Table 2. The models linked singular factors, quadratic and interactions of the parameters effects to responses variables consisted of: Specific density, penetrability, softening point and flash point.

The results of the analysis of variance, goodness of fit and the adequacy of models were summarized in Table 3. The data showed a good fit with the equation 5, which were statistically acceptable at $P < 0.05$ level. The values of coefficient of de-

termination (R^2) for the SG, P, SP and FP are respectively 0,97; 0,98, 0,98 and 0,99. These values of R^2 showed that the proposed models of all responses are adequate. In fact, it was suggested that, for a good fit of a model, R^2 should be at least 0,8 [1]. On the one hand, it's reported that the closer the value of R^2 to the unity, the better the empirical models [11]. On the other hand, the absolute average deviation (AAD), bias factor (B_f) and accuracy factor (A_f) must be including the range of 0-0.3; 0.75-1.25, and 0.75-1.25 respectively [12]. According to table 3, the values for, SG, P, SP and FP confirm the adequacy of the models. So, the models could be used to generate surface response curves to explain the influence of the independent factors on the responses studied.

Table 3. Regression coefficients, coefficient of determination (R^2), absolute average deviation (AAD), bias factor (B_f) and accuracy factor (A_f) for the four responses of PET recycling.

Coefficient/factors	Specificity Gravity (g/cm ³)	Penetrability at 25 °C, 100g 5sec in 1/10mm	Softening Point (°C)	Flash Point (°C)
CONSTANTE	0,000	0,000	0,000	0,000
FACTORS	Probability			
X1	1,000	1,000	1,000	1,000
X2	0,000	0,000	0,000	0,000
X3	0,450	0,995	0,076	0,023
X4	0,002	0,005	0,006	0,000
X1*X1	1,000	1,000	1,000	1,000
X2*X2	0,456	0,147	0,007	0,064
X3*X3	0,345	0,935	0,201	0,009
X4*X4	0,904	0,879	0,108	0,069
X1*X2	1,000	1,000	1,000	1,000
X1*X3	1,000	1,000	1,000	1,000
X1*X4	1,000	1,000	1,000	1,000
X2*X3	0,612	0,067	0,796	0,476
X2*X4	0,704	0,147	0,213	0,591
X3*X4	0,209	0,576	0,021	0,004
R^2	0,97	0,98	0,98	0,99
AAD	0,00	0,01	0,01	0,00
B_f	1,00	1,00	1,00	1,00
A_f	1,00	1,01	1,01	1,00

$P < 0.05$ /AAD, 0-0.3 / B_f , 0.75-1.25/ A_f , 0.75-1.25.

$$Y_{SD} = 1,06000 + 0,00000 x_1 + 0,04800 x_2 - 0,00302 x_3 + 0,01996 x_4 + 0,00000 x_1*x_1 + 0,0089 x_2*x_2 + 0,0125 x_3*x_3$$

$$+ 0,0016 x_4*x_4 + 0,00000 x_1*x_2 + 0,0000 x_1*x_3 + 0,0000 x_1*x_4 - 0,0067 x_2*x_3 + 0,0053 x_2*x_4 - 0,0200 x_3*x_4$$

$$Y_p = 58,00 + 0,000 x_1 - 14,001 x_2 + 0,006 x_3 - 3,991 x_4 + 0,00 x_1^2 - 4,44 x_2^2 - 0,25 x_3^2 + 0,48 x_4^2 - 0,00 x_1 x_2 - 0,00 x_1 x_3 + 0,00 x_1 x_4 - 6,67 x_2 x_3 - 5,32 x_2 x_4 + 2,00 x_3 x_4$$

$$Y_{sp} = 48,000 - 0,000 x_1 + 6,534 x_2 - 0,804 x_3 + 1,596 x_4 - 0,000 x_1^2 + 4,44 x_2^2 + 1,75 x_3^2 + 2,39 x_4^2 - 0,000 x_1 x_2 + 0,00 x_1 x_3 + 0,00 x_1 x_4 - 0,34 x_2 x_3 + 1,86 x_2 x_4 + 4,40 x_3 x_4$$

$$Y_{FP} = 245,000 - 0,000 x_1 + 15,601 x_2 + 1,195 x_3 + 7,187 x_4 + 0,000 x_1^2 + 2,67 x_2^2 + 4,89 x_3^2 - 2,95 x_4^2 + 0,000 x_1 x_2 - 0,00 x_1 x_3 + 0,00 x_1 x_4 + 1,00 x_2 x_3 + 0,80 x_2 x_4 + 6,60 x_3 x_4$$

With,

Y_{SD} : Gravity; Y_p : Penetration; Y_{sp} : Y_{FP} : Flash Point; X_1 : Asphalt; X_2 : PET; X_3 : Mixing Temperature and X_4 : Mixing Time.

3.1. Specific Density

The specific gravity of bitumen increases significantly ($P=0.000$, Table 3) from 1.02 to 1.1, while the PET rate increases from 0 to 12% (Figure 2). This increase is probably due to the fact that the density of PET (1.34–1.39 g/cm³) is much higher than that of asphalt (1.05 g/cm³), making bitumen consistent. Thus, the density of the mixture is affected, because there is the contact of materials of different characteristics [13]. Although temperature does not have a significant effect on density, it greatly affects the volume of PET and consequently impacts the density of the bitumen. This phenomenon was observed in a recent study [3]. Indeed, at high concentrations, polymers have the ability to absorb maltenes from bitumen, resulting in an increase in the volume of the mixture by forming a high-density polymer-rich phase [18].

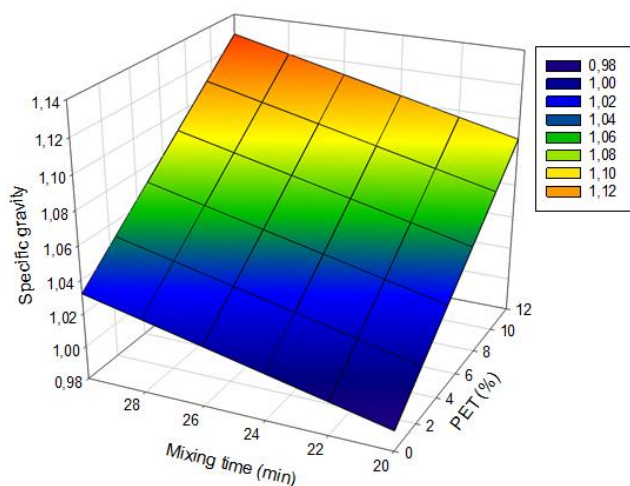


Figure 2. Evolution of specific gravity as a function of mixing time and PET ratio.

In addition, the density of bitumen increased significantly ($P=0.002$, Table 3) from 1.02 to 1.04; when the mixing time is increased from 20 to 30 minutes (Figure 2). This phenomenon can be explained by the fact that time is an essential factor for the kinetics of reactions. In fact, a longer mixing time increases the possibility of the PET adhering properly to the bitumen, which in turn increases the exchange surface between the materials. On the other hand, a reduction in mixing time would limit the intimate contact between PET and bitumen, which can discourage the homogenization of the medium by reducing the density. These changes are consistent with those reported by various research [20].

3.2. Penetrability

The penetrability of modified bitumen decreased significantly when the PET% rate ($P=0.000$, Table 3) increased from 0-12%. This decrease makes sense because the density of bitumen increases with the increase in PET content. Indeed, density and penetrability are two related phenomena. Thus, the increase in the PET content makes bitumen more consistent through the absorption of its lighter components, in this case maltenes (Figure 2), which results in the hardening of the latter. However, a bitumen hardens, resists better penetration or pressure that could be exerted by the penetrability needle, so the grade of the bitumen drops from the highest to the lowest. At 0% PET (Figure 3), the bitumen grade is 70 (1/10 mm), while at 6%, this grade increases to 60 (1/10 mm). With an incorporation rate of 12% PET, the bitumen grade drops to 40 (1/10 mm), resulting in bitumen too hard to be used. This finding corroborates previous research. Indeed, the incorporation of plastic waste at a rate greater than 7% has a negative impact on the penetrability of bitumen, in the sense that the latter can lose its binding properties as well as its plasticity [16]. However, the significant effect observed on the decrease in penetrability of bitumen caused ($P=0.005$, Table 3) by increasing mixing time is that time is a factor that increases the probability of PET interaction with hot bitumen.

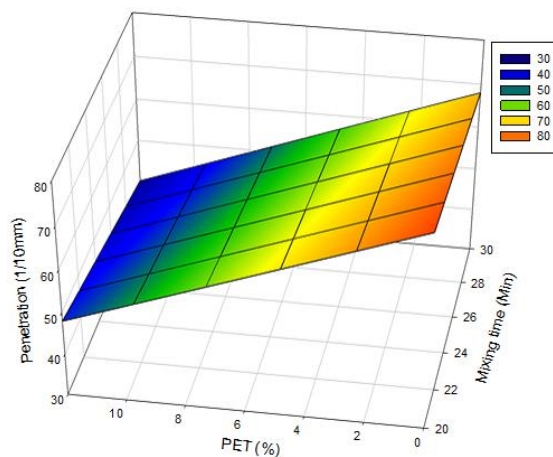


Figure 3. Evolution of penetrability as a function of mixing time and PET levels.

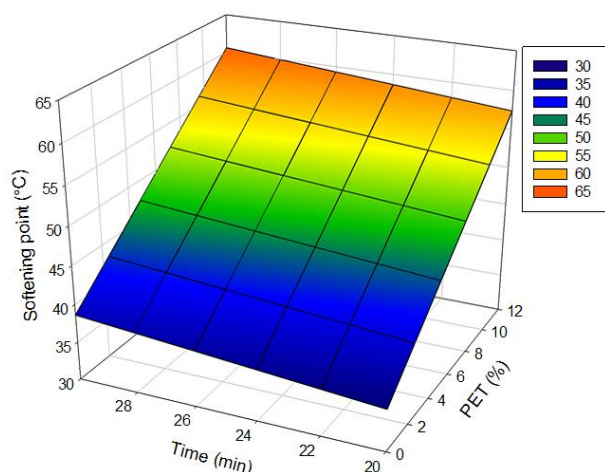


Figure 4. Evolution of Softening point as a function of mixing time and PET levels.

3.3. Softening point

The softening point (SP) of modified bitumen increased significantly when the PET rate ($P=0.000$, Table 3) and the mixing time ($P=0.006$, Table 3) increased by 0-12% and 20-30 min respectively (Figure 4). The increase in the SP as a function of the PET ratio is mainly due to the consistency of the bitumen. Indeed, pure bitumen is a non-Newtonian fluid, which can therefore see its viscosity drop greatly in the presence of heat and applied load stress (balls). However, if on the one hand the incorporation of PET ratio has the effect of increasing its consistency, the longevity of the mixing time allows, on the other hand, a good homogenization of the medium [17]. Thus, the SP increases and allows the bitumen to both withstand the load and thus improve its characteristics at high temperatures, which could increase rutting resistance [2]. These results corroborate the numerous studies carried out in this direction [20].

In addition, the X_3X_4 interaction (temperature and mixing time) significantly increases ($P=0.021$, Table 3) the ball-ring softening temperature of PET-modified bitumen. In fact, this effect was simultaneously accentuated by an increase in temperature and mixing time. When the mixing time and temperature are 25 min and 155 °C respectively (Figure 5), the SP accepts a value of 48 °C, on the other hand, when the mixing time is longer (30 minutes) and the modified bitumen is heated up to 160 °C, the SP increases to 52 °C. Indeed, a longer mixing time will increase the probability of a good dispersion and homogenization of PET in the bitumen while making the medium consistent. However, an increase in temperature would allow the bitumen to have a good viscosity, which would facilitate the adhesion of PET by making the mixture more rigid. Indeed, the rise in temperature undoubtedly leads to the disordered movement of the molecules (maltenes and asphaltenes) contained in the bitumen, which favors the expansion of the bitumen in order to better exchange with the PET and thus increase the SP.

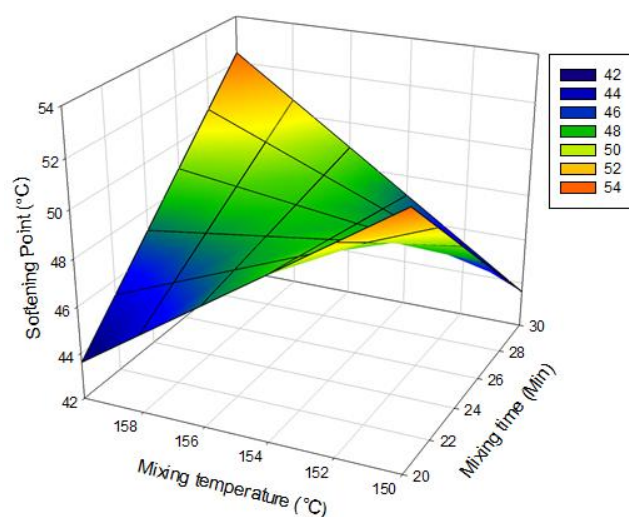


Figure 5. Effect of mixing time and temperature interaction on Softening point

3.4. Flash Point

The flash point of the modified bitumen increased significantly when the PET content ($P=0.000$, Table 3) and the mixing temperature ($P=0.023$, Table 3), increased by 0-12% and by 150 to 160 °C respectively (Figure 6). This change is likely caused by the increase in bitumen consistency with the addition of PET. Indeed, the increase in the PET ratio reduces the quantity of bitumen and consequently its lighter particles like maltenes, which results in an increase in flash point. On the other hand, the increase in the temperature of the mixture can cause an oxidation reaction of the bitumen that can transform the maltenes into heavier asphaltenes, which also has the effect of increasing the lighting temperature. Recent similar studies confirm this trend [4, 15].

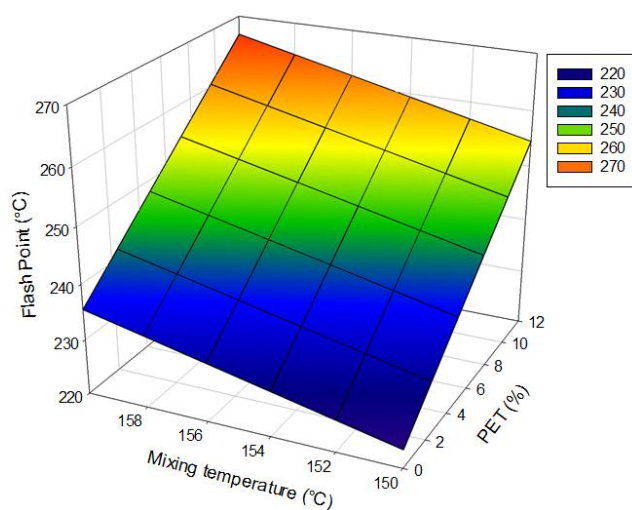


Figure 6. Evolution of flash point as a function of the PET content and the mixing temperature.

Similarly, the flash temperature of the modified bitumen increased significantly when the PET content ($P=0.000$, Table 3) and the mixing time ($P=0.000$, Table 3), increased by 0-12% and 20-30min respectively (Figure 7). As shown in Figure 2 the gravity of modified bitumen increases as a result of the combined effect of mixing time and PET content. This increase in density plays a major role in the composition of the modified binder obtained, in that a change in the structure of the bitumen is observed. Thus, the increase in the PET ration favors, on the one hand, the contribution of new semi-crystalline and less volatile particles in the composition of the mixture. On the other hand, it is obvious that the substitution of PET for bitumen reduces the quantity of volatile matter within the modified binder, which results in an increase in the flash point.

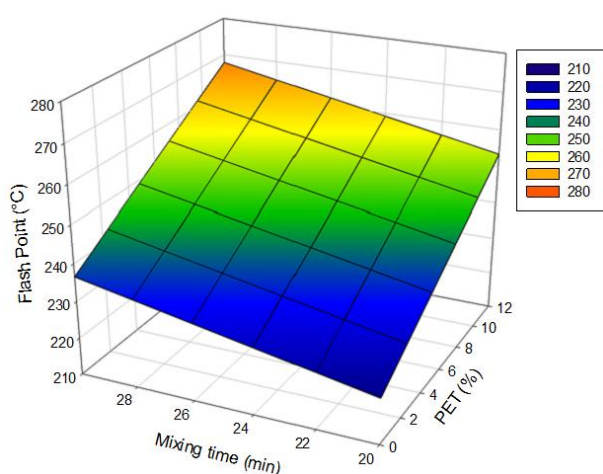


Figure 7. Evolution of the flash point as a function of the PET content and mixing time.

In addition, the X_3X_4 interaction (temperature and mixing time) significantly increases ($P=0.004$, Table 3) the flash point of the modified bitumen by PET.

In fact, this effect was accentuated simultaneously by an increase in temperature and mixing time. When the mixing time and temperature are respectively 25 min and 155 °C

(Figure 8), the flash point (FP) assumes a value of 246 °C, on the other hand, when the mixing time is longer (30 minutes) and the modified bitumen is heated up to 160 °C, the flash point increases up to 252 °C. The increase in FP in proportion to the increase in mixing time and temperature further explains how cured polymer-modified bitumen when the homogenization time is long and becomes heavier by oxidation and transformation of its light components under the effect of heat [4].

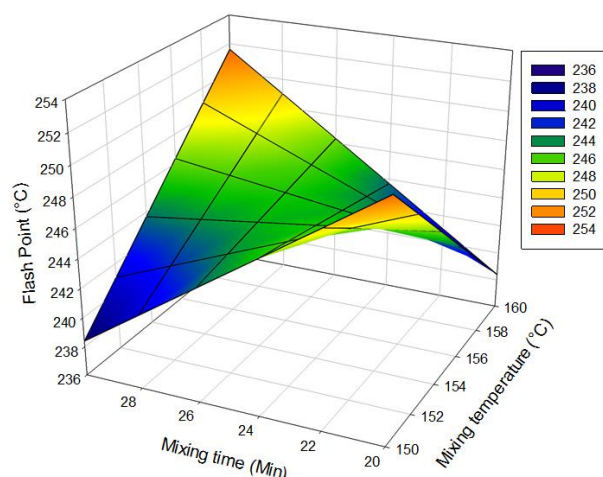


Figure 8. Effect of mixing time and temperature interaction on illumination temperature.

3.5. Optimization

In order to improve the performance of bitumen using powdery PET waste on the one hand and to define the optimal process condition on the other hand, a graphical optimization was carried out using Sigma Plot software and Excel. Such a methodology consists of superimposing the contour lines obtained from Doehlert's experimental design according to the specific criteria imposed. The optimal condition has been defined in order to obtain the best way and a better understanding of the requirements for recycling PET in bitumen, the desired normative qualities of which are mentioned in Table 4.

Table 4. Properties and Specifications of Pure Bitumen.

Characteristics	Specific gravity	Penetrability (25 °C, 100g 5sec in 1/10mm)	Softening point (°C)	Flash point (°C)
Specification	NFT 66-007	NFT 66-004	NFT 66-008	NFT 66-118
Limit values	1,0-1,1	60-70	43-56	>230

To achieve the optimal conditions for PET recycling, multi-response optimization was performed using Minitab. SG, P, SP, and FP are all optimized for this purpose. At the end of this op-

timization, the compromise is as follows: asphalt rate 5%; PET rate 6%; Mixing temperature 154 °C and Mixing time 22min.

This combination yielded responses as follows: 1.04 g/cm³

density (Y_{SD}); $60^{\circ}(1/10\text{mm})$ penetrability at 25°C , 100g and 5sec (Y_P); 50°C softening point (Y_{SP}) and 242°C flash point (Y_{FP}).

The composite desirability of this study indicates the value of 0.91, which is still closer to 1. This value proves that, on the one hand, the parameters seem to produce favorable results for all responses as a whole. On the other hand, individual desirability indicated that responses such as SD, P, SP and FP remain relevant and therefore more effective in terms of target values of 0.99; 0.78; 0.99 and 0.91 respectively.

4. Conclusion

This part of the study examined the physicochemical properties of PET polymer-modified asphalt by using the response surface methodology, particularly Doehlert's experimental designs. It appeared that the incorporation of PET plastic powder as a dope in asphalt made it possible to obtain a bituminous binder with good characteristics in terms of penetrability, softening temperature, specific gravity and flash point. This may justify the relevant choice of such a process in order to solve two equations: plastic pollution and the improvement of the quality of pavements. So, this technique has greatly affected penetrability and specific gravity, indicating that excess PET can make bitumen less cohesive and adhesive. However, the conditions under which these PET were incorporated into the bitumen led to an optimal formulation. The fact that the desirability of the study's composite (0.91) is so close to 1 suggests that the parameters appeared to yield positive results for all responses taken as a whole.

Abbreviations

PET	Polyethylene Terephthalate
AAD	Average Deviation
Bf	Bias Factor
Af	Accuracy Factor
SG	Specific Gravity
P	Penetrability
SP	Softening Point
FP	Flash Point
RMS	Response Methodology Surface

Author Contributions

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Mohagir Ahmed Mohammed: Project administration, Resources, Software, Supervision, Validation, Visualization, Writing – review & editing

Batran Sidick: Resources, Software, Supervision, Validation, Visualization, Writing – original draft, Writing – review

& editing

Conflicts of Interest

The authors declare no conflicts of interest.

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