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Optimization of Asphalt Binder Modified with PP/SBS/Nanoclay Nanocomposite using Taguchi Method

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***Abstract*—**This study has applied the L_{16} orthogonal array of the Taguchi method to determine the optimized polymeric Nanocomposite asphalt binder. Three control factors are defined as polypropylene plastomer (PP), styrene-butadiene-styrene elastomer (SBS) and Nanoclay. Four level of concentration contents are introduced for prepared asphalt binder samples. all samples were prepared with 4.5% of bitumen 60/70 content. Compressive strength tests were carried out for defining the optimized sample via QUALITEK-4 software. SBS with 3%, PP with 5 % and Nanoclay with 1.5% of concentrations are defined as the optimized Nanocomposite asphalt binders. The confirmation compressive strength and also softening point tests showed that modification of asphalt binders with this method, improved the compressive strength and softening points of asphalt binders up to 55%.

***Keywords*—**modified asphalt**,** Polypropylene, SBS, Nanoclay, Taguchi method

I. Introduction

ASPHALT (or bitumen) is used in road pavements as the binder of aggregates in a great extent all around the world. Asphalt pavements must undergo heavy loads and unfavorable environmental conditions for an acceptable period of time. High-temperature rutting and low temperature cracking are the most considerable limitations of unmodified and pure asphalts. Therefore, modification and reinforcement of asphalt binder is necessary.[1]

Improvement in rutting resistance, thermal cracking, fatigue damage, stripping, and temperature susceptibility have led polymer modified binders to be a substitute for asphalt in many paving and maintenance applications, including hot mix, cold mix, chip seals, hot and cold crack filling, patching, recycling, and slurry seal. They are used wherever extra performance and durability are desired. The rheological behavior of asphalt-polymer blends is of great interest because it is closely related to the performance of pavements. Furthermore, the addition of small amounts of polymer dramatically changes the rheological properties of the asphalt.[1]

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Polypropylene (PP) is a widely used commodity polymer known for its good process ability, low cost, integral hinge property, low density, high softening point and good mechanical properties. However, its application as a structural material is somewhat limited because of its relatively moderate fracture performance, especially at sub ambient temperature. The impact toughness of can be improved by addition of elastomers such as EPR[1],[2], EPDM[3],[4], SBS block copolymer[5]-[9].

Many studies were carried out on Nanoclay (NC) modified polymers, but little published information is available about Nanoclay modified bitumen. Many research studies have been performed on bitumen modification by polymer materials such as SBS (Styrene Butadiene Styrene Block Copolymer), SBR (Styrene Butadiene Rubber Latex), and EVA (Ethyl Vinyl Acetate). Chen *et al.* [10] research findings showed that SBS improved the rheological properties of asphalt binder due to the formation of a polymer network in the binder.

This network forms in two stages: at low polymer concentrations, the SBS acts as a dispersed polymer and does not significantly affect properties; at higher concentrations, local SBS networks begin to form and are accompanied by a sharp increase in the complex modulus, softening point temperatures, and toughness [10]. Recently, many researchers have reported effect of Nanoclay in PP/SBS blends on morphology, tensile properties, impact strength and flexural properties [8].

Nanoclays are micro-scale fillers which would make polymers efficient as filler reinforcements. Ghile [11] performed mechanical tests on asphalt mixture modified by cloisite. The result shows that Nanoclay modification can improve mechanical behavior properties of mixture, such as indirect tensile strength, creep, and fatigue resistance [11].

Althoghe there are many researches about Nanoclay or polymer modified asphalt binders, but simultaneous modification with both Nanoclay and polymer was rarely done.

This study utilized the Taguchi L_{16} orthogonal array for optimizing the concentration content of PP/SBS/Nanoclay modified asphalt binders. The optimized sample was obtained from compressive strength test results. The total optimize sample was characterized via softening point and compressive strength test.

TABLE I
BITUMEN'S PROPERTIES

Bitumen type	60/70
Density(gr.cm^{-3})	1.03
Penetration grade(°C)	65
Ductility at 25°C (cm)	100

TABLE II
NANOCLAY PROPERTIES

Treatment/Properties	Cloisite-15A
Organic Modifier	*MT2ETOH* (methyl, tallow, bis-2-hydroxyethyl, quaternary ammonium)
Base	Montmorillonite
Moisture	1.2%
Particle Size Distribution (%)	10μm
Density, gr/ml	1.5

II. EXPERIMENTAL

A. Materials

In this approach, bitumen Bitumen was a 60/70 penetration grade and its properties are shown in Table 1.isotactic polypropylene (PP) (grade: repol H060MG, MFI 6 g/10 min) was obtained from reliance industries limited. SBS block copolymer (carl Prene 501) was purchased from dynasol. A common type Nanoclay used in this research was Cloisite-15A. Properties of Nanoclay is shown in Table II.

B. General description of preparation the modified asphalt samples

The PP/Nanoclay master batches were prepared using Haake Torque Rheometer (Rheomix 600P) fitted with roller rotors (net chamber volume 69 cm^3). The batch weight was optimized to 43g and mixing was carried out at 200^{0}Cand 20 rpm. The Nanocomposites of PP/SBS/Nanoclay were prepared Haake twin screw extruder.

The high tendency for decreasing of viscosity and reaching to a fine mixture in one hand, and the enhancement of polymer destruction in high temperatures in other hand, made the mixing temperature ranged from 180^{0}C to 200^{0}C. Mixing the obtained Nanocomposite with bitumen was done using Industrial high shear mixture,Silverson, model L4R, Italy. Bitumen (4.5%wt) was preheated till 150^{0}C.In a reason of initial mixing, the Nanocomposite was mixed with bitumen with low rate (200rpm) for 10min. according to this, the polymer structure doesn't deteriorate during the second phase of mixing.in the second phase, the total mixture was mixed with high rate (3000rpm) for 30 min.

The aggregates used in this study were crushed limestone aggregates with gradation characterized by 12.5mm nominal size (according to Pavement Guidelines in Iran). Physical properties of the aggregate, both coarse and fine, together with mineral filler are given in Table 3. In a reason of performing the asphalt tests, marshal samples were synthesized according to ASTM D1559 standard.

Compressive strength and softening point of asphalt samples were determined in accordance to the ASTM C39 and ASTM D35 respectively [13].

TABLE III
AGGRIGATE PROPERTIES

Coarse aggregate (ASTM C127)	
Bulk specific gravity, g/cm^3	2.58
specific gravity, g/cm^3	2.68
Absorption, %	0.45
Fine aggregate (ASTM C128)	
Bulk specific gravity, g/cm^3	2.69
specific gravity, g/cm^3	2.71
Absorption, %	0.96
Filler (ASTM D854)	
specific gravity, g/cm^3	2.71
L.A. Abrasion, %, (ASTM C131)	23

III. DESIGN AND CONDUCT THE EXPERIMENTS

A. Describe quality characteristics and design parameters

Quality characteristics selected to be the optimization criteria are divided into three categories, the larger-the-best, the smaller the- best and the nominal-the-best. The quality characteristic of this study is the maximum for compressive strength of synthesized Nanocomposite bituminous asphalt samples., which belongs to a larger-the-best characteristics, and is calculated by using equation below:

$$S/N_L = -10 \log\left(\frac{1}{n}\sum_{i=1}^{n}\frac{1}{y_i^2}\right) \quad (1)$$

Where y_i is the value of quality characteristic measured from the trial and the unit of S/N is dB; n is the number of the tests in a trial.

B. Definition of effecting factors and their levels

Three factors with three levels were chosen and studied in this work:

- Factor (A) Polypropylene (PP) : 2,4,5,7 %wt.
- Factor (B) Styrene-butadiene-Styrene (SBS) : 1,3,4,5 %wt
- Factor (C) Nanoclay : 1,1.5,2,3 %wt

C. Selection of orthogonal array (OA) and assignment of factors

Taguchi's arrays are versatile recipes that are applied to several experimental conditions. The experiments were designed according to the orthogonal array technique. An orthogonal array is a fractional factorial design with pair wise balancing property. Using orthogonal array design can estimate how multiple process variables affect the performance characteristic simultaneously while minimizing the number of test runs. In this paper, we used the L_{16} orthogonal array for three controlling factors. To observe the data reliably on this experiment, compressive strength tests were carried out for two times with same conditions.

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Standard L_{16} OA table with four levels and three factors is shown in Table IV. Each row in the table represents a trial condition with the level of factors.

For analysis of the results and optimization of conditions for setting the control factors, QUALITEK-4 software was used. QUALITEK-4 (QT4) Version 4.75 is the windows version software for Automatic Design and Analysis of Taguchi Experiments.

D.Analysis the results to determine the optimum condition

Analysis of variance (ANOVA) is performed on the experimental results obtained from Taguchi experiments. ANOVA study identifies the relative influence of the factors in discrete terms. According to the three main control factors at four levels, obtaining signal-to-noise ratios (S/N) of the experiment results after selecting the proper orthogonal arrays identifies the relationship between the parameters and the response (the maximum compressive strength), and obtains the primary optimum of the operating parameters. This study uses the ANOVA to identify the important parameters that can affect asphalt binder quality characteristics and determines the most robust set of operating conditions from variations within the results.

TABLE IV
EXPERIMENTAL LAYOUT OF L16 ORTHOGONAL ARRAY (THE NUMBER IN COLUMN S A,B AND C INDICATE THE LEVELS OF THE FACTORS)

Trial	A	B	C
1	1	1	1
2	1	2	2
3	1	3	3
4	1	4	4
5	2	1	2
6	2	2	1
7	2	3	4
8	2	4	3
9	3	1	3
10	3	2	4
11	3	3	1
12	3	4	2
13	4	1	4
14	4	2	3
15	4	3	2
16	4	4	1

IV. RESULTS

A. Taguchi results

The results of the compressive strength for each sample and corresponding S/N ratios are listed in Table V. The quality characteristic chosen in this case is compressive strength because it is so frequently used in asphalt characterization and performing evaluation. In order to evaluate the influence of each factor on the compressive strength, the S/N ratio for each factor should be computed. The S/N ratio averages for each factor at four levels are presented in Fig. 1. Overall mean value was calculated as -46.1 from all Taguchi experiment results taken from compressive strength data (see Fig. 1).

On the other hand, the maximum variation level was determined as -1.5 that is belongs to SBS factor.

The difference between the slopes of factors shows the level of effectiveness of each factor.

Changes in SBS concentration results in the most changes on S/N ratio. It can be seen that the next effective factor is PP polymer.

TABLE V
RESULTS OF COMPRESSIVE STRENGTH TESTS AND CORRESPONDING S/N RATIO

Sample	Trial #1	Trial #2	S/N
1	65	67	-47.948
2	68	71	-47.684
3	75	75	-46.798
4	64	66	-46.202
5	69	71	-46.92
6	66	66	-47.578
7	70	71	-46.236
8	64	65	-46.32
9	68	67	-45.569
10	63	64	-45.145
11	64	63	-46.118
12	65	65	-45.918
13	67	66	-44.994
14	65	67	-44.849
15	63	61	-45.145
16	64	65	-45.986

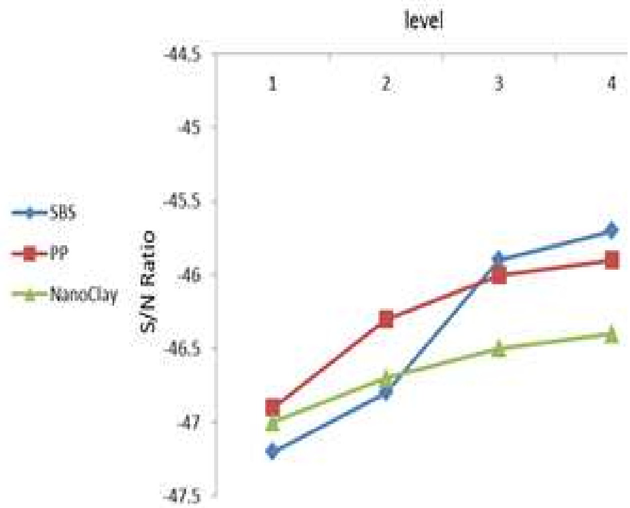

Fig. 1 Main effect plots for S/N ratios obtained from compressive strength data

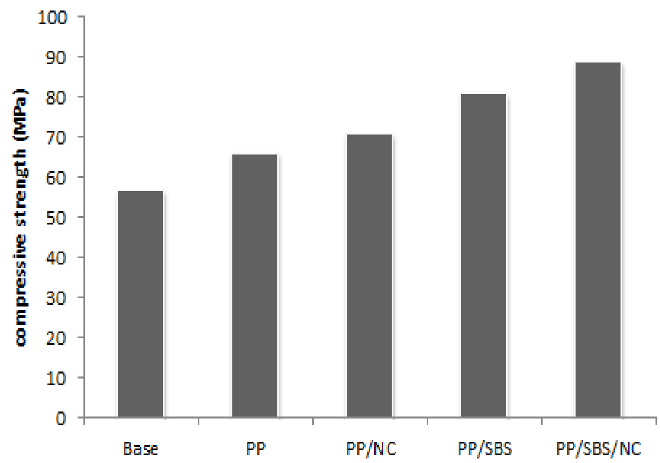

Fig. 2 Effect of different combination of modifiers on compressive strength

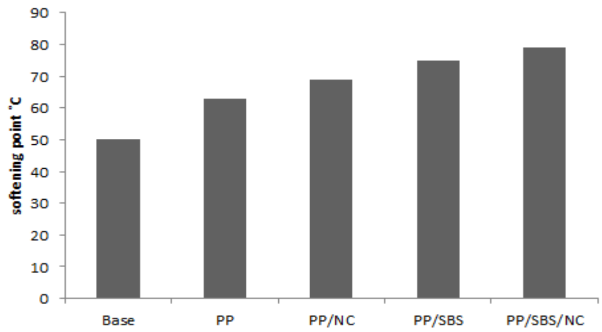

Fig. 3 effect of different combinations of modifiers on softening point

B. ANOVA results

ANOVA was performed in order to see whether the process parameters are statistically significant or not. The results of ANOVA are listed in Table 6. The rows which are marked as error refers to the error caused by uncontrollable factors (noise). In general, the value should be below 50%; otherwise the results would not be reliable. As it can be seen from Table 6, the calculated error is about 7% which is far enough from the limit. It means that the error of the experiment is not significant. Quantitative evaluation can be achieved with using percentage contribution (P%) [12]. It is calculated by dividing the source's net variation by SS_T, which is given as follows:

$$P(\%) = \frac{SS_A - (DOF_A \times MS_e)}{SS_T} \times 100 \quad (2)$$

TABLE VI
PARAMETERSS OF THE STATICAL ANALYSIS FOR COMPRESSIVE STRENGTH DATA

Factor	Degree of freedom	Sum of sqrs.	Variance	Percent (%)
SBS	3	7.977	3.712	46.540
PP	3	5.817	0.186	33.93
NC	3	2.825	1.048	16.48
Other error	6	0.52	0.082	3.04
total	15	17.14	-	100

The order of importance of factors is as follows: SBS> PP> NC. SBS content is the most important factor for compressive strength data.it seems that Higher SBS content results in higher compressive strength. This result, according to the quality characteristics that was chosen for strength data, was to some extent, predictable. The final step in Taguchi robust design, is predicting the optimum conditions and their performances based on entered results. Our total optimum modified bituminous asphalt will be presented according to the obtained results of performed tests. Predicted level description and percent contribution of all factors are presented in Table 7.

C. Confirmation experiment

The confirmation experiment is highly recommended by Taguchi to verify experimental conclusions. After optimum conditions were determined, the confirmation experiment was performed with combination of the optimum levels to compare the results with the predicted performance.

In a reason of consideration the independent quantity of effectiveness of each factor, the compressive strength of optimum Nanocomposite asphalt sample, the sample without SBS , the one without Nanoclay and the one without Nanoclay and SBS were examined and compared with the unmodified bituminous asphalt sample (Fig. 2).

TABLE VII
PREDICTED LEVEL DESCRIPTION AND PERCENT CONTRIBUTION

Factor	Level description	Level	Contribution (%)
PP	5	3	46
SBS	3	2	34
NanoClay	1.5	2	20

V. RESULTS AND DISCUSSION

As it can be seen in Fig. 2, the optimized PP/SBS/NC sample has the highest level of compressive strength. All modified samples showed an improvement in their characteristics but the sample that was introduced via taguchi optimization method had 56% improvement in compressive strength.

Fig. 3 shows that, in presence of PP/SBS/NC content, the softening point of the asphalt was increased more than PP, PP/NC and PP/SBS modified asphalts and the base asphalt. As it could be predicted, the maximum increase was observed with the total optimum sample. The increment of softening point level for PP/SBS/NC modified sample reached to 58%.

According to the results, When SBS was introduced to PP modified asphalt, the PP/ SBS modified asphalt show slight higher softening point comparing to base asphalt sample.Which implies that, the high temperature properties were improved. This may be due to hardening effect of PP and the network structure formed by SBS in the asphalt blend. In the absence of SBS , the PP/NC modified asphalt also showed an increment in softening point but as it could be concluded from Taguchi results, it has a fewer effect than the PP/SBS modified samples. PP/SBS modified asphalt sample has higher effect on asphalt physical properties because it produced less brittleness asphalt binder at low temperature to resist thermal cracking and more stable and flexible asphalt mixture at high service temperature to resist fatigue cracking.[13]. But Nanoclay particles don't chemically react with asphalt mixture. When bitumen is modified with small amounts of Nanoclay, its physical properties are successfully enhanced [14] Nanoclay materials have high aspect ratio and a large surface area and adding even a small amount of these can highly increase compressive strength and softening point(see Fig. 2 and 3). Nanoclay have improved the rigidity of PP which may be attributed to reinforcing effect.

According to results, the simultaneous combination of a plastomer (PP) and elastomer (SBS) and nanoparticle (Nanoclay) can balance the advantage and disadvantage of all three control factors defined in this study.

VI. CONCLUSION

Scientists and engineers are constantly trying to improve the performance of asphalt mixtures by using various methods including asphalt binder modification.

This study aimed at the optimization of asphalt mixture modified with PP/SBS/NC via Taguchi method. Two series of data were obtained thorough compressive strength tests of samples based on Taguchi recipe.

According to the results, this modified asphalt mixture had the best output compared with PP/SBS or PP/NC modified asphalt samples. On the basis of Taguchi results, SBS elastomer with 3% wt. had the highest level of effectiveness due to its ability for increasing the strength of asphalt mixture and softening point. PP plastomer was the second effective control factor with 5% wt. of asphalt content.

Finally Nanoclay with 1.5% wt. had the lowest percentage of contribution. The nature of Nanoclay as filler material results in higher compressive strength and softening point. The most important fact about Nanoclay fillers is that small amount of this material caused huge changes in obtained results.

Although Nanoclay had the least effect on physical characterization of modified asphalt mixture, but compared with the two other factors (PP,SBS) concentrations, it had a significant effect on total results.

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