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Improvement of Deformation Resistancy of Asphalt by Modification with Tire Rubber

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타이어고무를 이용한 개질에 따른 아스팔트 변형저항성 향상 연구

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ABSTRACT : Nowadays, modifying asphalt to improve the resistancy of plastic deformation or rutting on roads has been drawing attention. In this study, asphalts were modified with modifying agents and ground rubber from waste tire (GRT), and the effects of modifier and GRT on the properties of asphalt were analyzed. The Marshall stability of modified asphalt was increased by 98% and the tensile strength increased by 43% compared to straight asphalt. GRT played an important role in enhancing these properties. Viscosity tests, penetration tests and TMA analysis showed the deformation resistancy of modified asphalt. IR and GPC tests indicated that asphalt and modifier have similar chemical structures to each other, and chemical bonding between asphalt and modifier have occurred so that the molecular size lengthened.

요 약 : 포장도로의 소성변형을 줄여보고자 개질아스팔트가 나타나고 있다. 본 연구에서는 타이어 고무 등을 이용하여 개질된 아스팔트의 물성을 알아 보았다. 마샬안정도와 인장강도가 각각 98% 및 43% 증가하여 타이어 고무의 개질효과를 알 수 있었다. 점도, 침입 및 열 분석에서 아스팔트의 변형저항성이 향상되었음을 알 수 있었다. IR 및 GPC 실험에서 개질제의 구조가 아스팔트의 구조와 유사할 경우 두 화합물 사이에 화학결합이 발생하여 분자들의 크기가 증가함으로써 개질효과가 커짐을 알 수 있었다.

Keywords : asphalt, modifier, GRT, plastic deformation, Marshall stability

Ⅰ. Introduction

Today, the number of motor vehicles is increasing as the economy prospers. Consequently, problems caused by waste tires as environmental pollutions, and handling costs of waste tires are current issues of our economy. With no apparent means to reuse these tires economically, financial and physical efforts are occurred just to store them. Some studies have been done about recycling these tires to minimize their effects on the environment and the economy, but these studies showed only limited results. One of the most read studies of this field reports of enhancing performance of road pavement asphalt

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by using waste tires.[1,2]

Asphalt has very strong adhesion and waterproofing properties along with high resistancy against acids, bases, and salt. When heated, its liquid form can be mixed with aggregates, and because of its natural stickiness, it is easily formed into a mixture. Once the mixture is cooled to room temperature, it regains its toughness. These properties allow it to bear frequent traffic on highways or be used a paving for an airport.

The increase in the number of vehicles also leads to heavier traffic, which can cause early breakage or reduced durability of asphalt pavement. As a result, the maintenance cost increases. Reasons for road pavement damage can be listed as heavy traffic, inadequate environment, poor quality of the material, and faulty construction.[3] The quality in asphalt most responsible for such damage is plastic deformation. When asphalt is used as a bonding agent, it is exposed to plastic deformation at high temperatures, and becomes brittle and cracks at low temperatures. For asphalt, as the viscosity decreases, the rigidity also decreases, and the mixture is defenseless against plastic deformation. Plastic deformation is defined, as a change in original form of a material by an outside factor such as temperature or pressure. Plastic deformation in asphalt pavement is caused by repetitive loads on the surface, resulting in permanent deformation on the surface. This can be especially detrimental to a driver's safety as it can damage the vehicle or make the vehicle difficult to maneuver.

In order to prevent plastic deformation, it is recommended that asphalt of high viscosity (low penetration rate) be used in cases of where the asphalt layer is thick or temperatures are high. However, in countries where there are clearly defined four seasons, asphalt with high viscosity can become unnecessarily hard during the winter, and open to the possibility of temperature-caused cracking is a consideration that must be taken into account. Therefore, to effectively prevent plastic deformation at high temperatures and temperature-caused cracks at low temperatures, factors to control the behavior of asphalt within a range of common temperatures that asphalt encounters will need to be specified. Attempts to use modified asphalt, also need to be made. Modified asphalt to prevent plastic deformation is called property-improved asphalt, and is made by adding certain amounts of modifiers.[4] Examples of these are rubbers, thermoplastics and thermosetting resins.[5-11]

Asphalt modifiers need to be similar to asphalt in chemical structure, so as to be compatible with asphalt, and to be able to improve adhesion of asphalt. These similar qualities will add to the performance asphalt helping one of the biggest problem faced by asphalt road pavement, which is plastic deformation.[3]

In this study, common asphalt (AP-5) was modified with two chemical compounds to improve resistance against plastic deformation. The compounds employed here are unsaturated polyester resin and polyoctenamer which is frequently used as plastisizer in the rubber industry. In addition, ground rubber from waste tire (GRT) was also used as a modifier to improve the strength of asphalt by enhancing elasticity. In order to control the size effect of the aggregate added to the asphalt, only aggregates of diameter of 13mm were used. To see the effect of modification, mechanical properties (tensile strength and Marshall stability) of modified asphalt, tests were conducted. Test were also done measuring for the penetration grade, and viscosity; and TMA was used to measure changes in resistance against plastic modification, and IR and GPC were used to test the bonding possibility between asphalt and modifiers.

Ⅱ. Experiment

1. Materials and Reagents

All asphalts used in experiments were AP-5 (Inchon Oil) and had penetration grades between 60 and 70. Polyoctenamer ('Vestenamer', Degussa-Huls), unsaturated polymer made from polymerization of cycleoctene that has a double bond of C4=C5, and unsaturated polyester resin (UPR, Aekyung Chemicals) were used as modifiers. In addition, in order to test the possibility of cross-linking between asphalt and modifiers, dicumyl peroxide (DCP, Aldrich Chemical) for Vestenamer and MEKPO (methylethyl ketone peroxide) for UPR were used. GRT having diameter of 0.6 mm~1.0 mm was provided by the Korea Resource Recycle Association. Tetrahyrofuran was used as a solvent for IR and GPC analysis.

2. Modification Process

The mixture was inserted in a kettle and mixed for 3hours at 180°C and 700rpm.

Materials used as modifiers were added so that the total (sum of asphalt and modifier) is 100wt%, and peroxides used as cross-linking agents for modifiers were added in accordance to wt% of modifiers (100 modifier).

3. Analysis of Mechanical Properties

3-1 Marshall Stability Test

The specimens of modified asphalt were prepared using pre-warmed aggregate. The specimens were molded using a cylindrical mold 50mm in diameter and 100mm in length. The specimens were made and tested in accordance with guidelines of Marshall asphalt mix by ASTM D 1559. First, a 13mm aggregate was heated at 180°C for more than 24hours. Modified asphalt was added at 4wt% to the aggregate. Using a Marshall tamping machine that drops a 4.5kg Marshall hammer from 45.7cm high, the top and bottom surfaces of the specimens were hammered 50times each. The specimens inside the molds were cured at room temperature for 24hours. Seven specimens were made for each type. The specimens were then placed in a 60°C water bath for 30minutes, towel dried, and tested by a compression tester (HJ-1191, Heung-Jin Co.) at a loading speed of 50mm/min. An arithmetic mean was then calculated for each type after excluding maximum and minimum values.

3-2 Indirect Tensile Strength Test

One of the highly anticipated effects of the modification of asphalt is in enhancing tensile strength. Tensile strength is the most important property of asphalt pavement for resistance to loading from vehicles. Increasing tensile strength will increase the life span of asphalt. In cases when direct testing for tensile strength is difficult to perform, indirect testing is performed using a compressive strength tester, instead of tensile strength tester. A 13mm wide metal loading belt with its concave surface similar to that of a Marshall specimen with diameter of 101.6 mm, was used to apply load (speed of 50mm/min).

4. Analysis of Resistance against Plastic Deformation

4-1 Viscosity Test

A Brookfield viscometer was used to test temperature-dependent viscosity of AP-5, modifier, and modified asphalt. 400g of each composition was placed in a 500mL beaker and measured in an oil bath. Starting at 190°C, the temperature drops and its corresponding viscosity changes were measured every 5minutes. Viscometer ran at 12rpm, and the spindle used was LV-4 type.

4-2 Temperature-dependent Behavior Test

The temperature-dependent behavior of asphalt, modifier, and modified asphalt were measured using TMA(TMA 2940, TA Instruments). Special care on measurements was taken at 60°C where plastic deformation of asphalt was anticipated. Temperature was increased from 25°C to 200°C by 10°C/min.

4-3 Penetration Test

Penetration, which represents deformation of modified asphalt, was measured in accordance with KS M 2252. The specimen was placed in a 25°C water bath for 1~2hours before testing. The penetration grade was measured seven times for each specimen, and excluding maximum and minimum value, five data points were used to calculate the arithmetic mean.

5. Analysis of Reaction between Asphalt and Modifier

5-1 FT-IR

FT-IR (NEXUS, Nicolet Instrument) was used to see the reaction between AP-5 and modifier. Each specimen was prepared at 10% concentration in THF, and measured using KBr after THF was volatilized.

5-2 GPC

The molecular weight of asphalt, modifiers, and modified asphalt was measured by GPC (Waters, composed of Waters 1515 isocratic pump and Waters 2414 RI detector). Polystyrene of molecular weight of 1,250g/mol was used as the standard.

Ⅲ. Results and Discussion

1. Preparation of Modified Asphalt

1-1 Effect of Type of Modifier

First, the effects of modifiers were examined. Modifier contents were set at 2wt%, and initiators (peroxides) were added in related modifiers. Thus relative comparison was used to measure the effects. Table 1 describes each composition and its Marshall strengths. UPR did not show good result as a modifier whether used independently or with MEKPO. UPR and AP-5 were not compatible and caused phase separation. When LDPE is used as a modifier, the result was not to satisfaction either because of phase separation.[7,8] Also, when MEKPO was used as an initiator, at 180°C, i.e., the mixing temperature, most of it sublimed. Thus its reactivity with UPR is expected to be small. On the other hand, Vestenamer, a thermoplastic tackifier and plasticizer, is very stable against temperature and decomposition, and because it dissolves well in most of aliphatic and aromatic solvents, which are also good solvents used with asphalt, it can be theorized that the compound may form chemical bonds well with asphalt and thus might improve the property of asphalt. In opposition, DCP, as an initiator, did not improve asphalt's property. One of the reasons for failure is that it also sublimed, slightly though, at the temperature of mixing.

Table 1. Effect of Modifier and Initiator on Marshall Stability

sample	modifier	initiator	modifier content (wt%)	initiator content (wt%)	Marshall stability (kg · f)
AP	·	·	0	0	228
APU-2	UPR	·	2	0	230
APU-M-2-1	UPR	MEKPO	2	1	240
APU-M-2-10	UPR	MEKPO	2	10	231
APV-2	Vestenamer	·	2	0	303
APV-D-2-10	Vestenamer	DCP	2	10	306

1-2 Effect of Amount of Modifier

It has been shown above that Vestenamer is a good modifier for asphalt. Thus, in order to find out the optimal content of Vestenamer as a modifier of ashphalt, a Marshall stability test was conducted on different ratios of AP-5 and Vestenamer. Table 2 depicts the results. 100% pure asphalt showed a Marshall value of 228kg·f, and 100% Vestenamer showed 265kg·f. When asphalt and Vestenamer were mixed, the homogeneity was gained (no phase separation occurred) so that the Marshall value dramatically increased due to their synergistic effect. The higher the Vestenamer content was, the greater leap in the Marshall value there was.

Table 2. Marshall Stability of Asphalts upon Vestenamer Content

sample	asphalt (wt%)	modifier (wt%)	Marshall stability (kg · f)
AP	100	0	228
APV-1	99	1	280
APV-2	98	2	303
APV-5	95	5	276
APV-10	90	10	260
APV-20	80	20	301
APV-30	70	30	365
APV-40	60	40	501
APV-50	50	50	392
V-100	0	100	265

By controlling the ratio between viscous AP-5 and elastic Vestenamer, it is possible to prepare modified asphalt having an ideal viscoelasticity with Vestenamer at under 5% .

When Vestenamer content was at 2%, the Marshall value was measured as 303kg·f, and when at 40%, the value was at its maxium of 501kg·f. Thus, an optimal point of resistance to plastic deformation was achieved when Vestenamer content was at 2%.

1-3 Effect of GRT

The effect of GRT as an asphalt modifier was measured by changing the weight content of GRT to asphalt. Vestenamer was also used during the modification at a fixed value of 2%. According to Figure 1, when GRT content was 1%, the Marshall value showed its highest value. As the content increased, the Marshall value decreased. It is postulated that when excess GRT was used for modification, its increased surface area leads to loss of binding ability of asphalt or/and due to the density difference, it can cause localization of GRT, which then behave as impurities rather than an effective modifier.

Next, another set of measurements were conducted to find out the optimal content of Vestenamer, this time with GRT content set at 1%. According to Table 3, the trend of changes in the Marshall value was similar to that of changes measured without GRT. However, as GRT was added, the width of the Marshall value widened. It is considered that an addition of GRT gives asphalt an elastic character thereby increasing its stability.

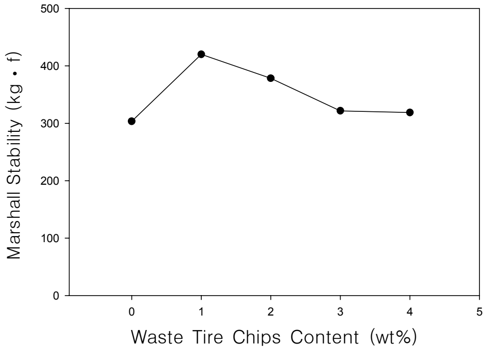

Figure 1. Marshall stability of modified asphalts(Vestenamer 2wt%) upon GRT content.

Table 3. Marshall Stability of Modified Asphalts

Sample	Alphalt (wt%)	Modifier (wt%)	GRT (wt%)	Marshall stability (kg · f)
AP	100	0	1	307
APVR-1-1	99	1	1	412
APVR-2-1	98	2	1	420
APVR-5-1	95	5	1	402
APVR-10-1	90	10	1	378
APVR-20-1	80	20	1	402
APVR-30-1	70	30	1	425
APVR-40-1	60	40	1	625
APVR-50-1	50	50	1	593

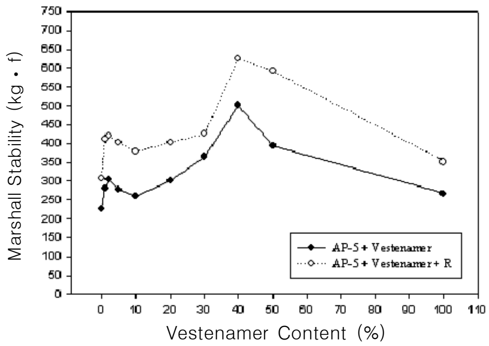

Figure 2. Marshall stability of modified asphalts(R= GRT 1wt%).

In sum, Figure 2 shows the effectiveness of Vestenamer resin and GRT rubber together on the Marshall stability of modified asphalts.

2. Mechanical Properties of Modified Asphalt

AP-5, 2% Vestenamer, and 1% GRT were mixed together for 3 hours to prepare a specimen(APVR-2-1. Table 4 presents the Marshall stability values of the modified asphalt and neat asphalt. The Marshall value of the pure asphalt(AP-5) was measured at 359kg·f, and that of APVR-2-1 was 703 kg·f. The value of APVR-2-1 showed a 96% increase, which is well above the asphalt concrete

Table 4. Mechanical Properties of Straight Asphalt (AP-5) and Modified Asphalt(APVR-2-1)

Test	AP-5	APVR-2-1
Marshall stability (kg・f)	359	703
Tensile strength (kg・f)	385	513

standard for surface pavement of 500kg·f. It is considered that the 96% increment is due to solely the modification, since the size effect of the aggregate was already excluded. The indirect tensile strength read at 359kg·f for AP-5 and 513kg·f for modified asphalt. The strength of asphalt was improved by modification with Vestenamer and GRT as the value increased by 43%.

3. Resistance to Plastic Deformation

3-1 Viscosity of Modified Asphalt

As viscosity of asphalt decreases so does the rigidity of the mixture, thus leading to easier plastic deformation.[11] High viscosity means there are strong interactions between chains. Plastic deformation is caused when an external force greater than the interactions between chains is applied, that, as a result, cause slipping of chains. In other words, molecules with strong interactions will require greater force to start slipping and will have more resistance against plastic deformation.

The viscosity of modified asphalt was measured with a Brookfield viscometer. Since asphalt's resistance to plastic deformation increases as viscosity increases, measuring viscosity of Vestenamer-modified asphalt will give hints on its resistance to plastic deformation. Figure 3 depicts changes in viscosity of different asphalts. When the viscosity of AP-5 was measured by cooling it down to room temperature, the viscosity range went out of the Brookfield range and was not able to be measured. The 2% Vestenamer-modified asphalt(APVR-2-1) showed similar viscosity to that of pure asphalt(AP-5) at high temperatures, although the difference became greater as the temperature dropped below 85°C. Especially the 50% Vestenamer-modified asphalt (APVR-50-1) showed a very high viscosity at low temperatures. It appeared that Vestenamer-modified asphalt had very high resistance to plastic deformation at 60°C (measurement temperature for plastic deformation). However, at the modification temperature of 135°C (measurement temperature for processing), viscosity values were similar to that of pure asphalt. This characteristic seems to be the apparent advantage of using Vestenamer. As a comparison when SBS or SBR is used as a modifier the viscosity tends to increase dramatically when the temperature falls below 135°C during modification, and it lowers workability.[12]

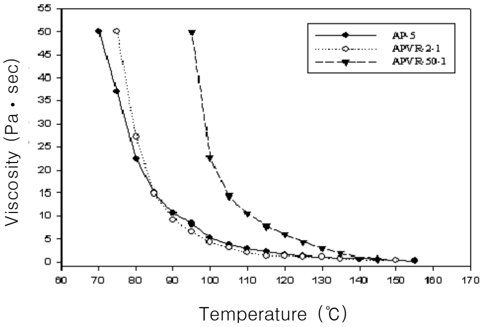

Figure 3. Viscosity against temperature for various asphalts.

3-2 TMA Measurement of Dimension Changes

TMA was used to see the thermal behavior of AP-5, Vestenamer, and modified asphalt at 60°C. In general, TMA is used to estimate the melting temperature of polymer materials. Melting of polymer is evidence of active movement of polymer chains. Plastic deformation of asphalt is also an example of temperature-stimulated molecular chain movements. Thus, TMA can be used to measure the temperature for molecular chains activated in asphalt. AP-5 in Figure 4 showed immediate deformation as the temperature was raised from room temperature. At about 35°C, more drastic deformation was observed, and at 60°C, the temperature which plastic deformation occurs allegedly, the deformation was completed. On the other hand, the modified asphalt (APVR-2-1) showed a prolonged time for maintaining its initial dimension and progressively moving towards more deformation at 60°C. The fact that

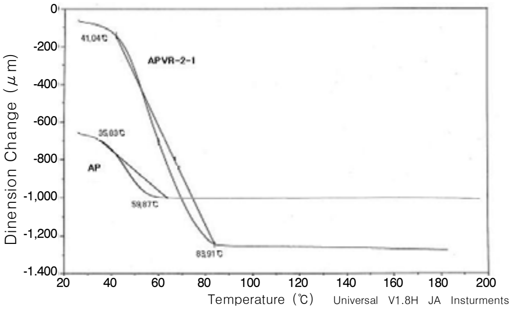

Figure 4. Thermal behavior of asphalts on TGA.
AP: pure asphalt
APVR-2-1: modified asphalt

deformation was completed at about 84°C indicates its resistance against deformation was increased. These results mean that when Vestenamer is contained in asphalt, it increases plastic deformation temperature of AP-5 by lessening its fragility at 60°C, thereby increasing resistance against plastic deformation of modified asphalt.

3-3 Penetration of Modified Asphalt

Standard penetration test methods set by KS M 2201 specifies 5 different penetration levels for road pavement asphalt; 40~50, 60~70, 85~100, 120~150, and 200~300. Higher penetration grades means softer asphalt and higher chances of plastic deformation. However, if the penetration grade is too low, although this will give higher resistance to plastic deformation, there will be higher chances of cracks during cold weather.[12]

Figure 5 depicts penetration grades of asphalts with different Vestenamer content. The unit of penetration is 0.1mm. The penetration grades of modified asphalts were much lower than that of pure asphalt and the trend followed Vestenamer content to AP-5. The penetration grade of pure Vestenamer was measured as 5 approximately. That of 2wt% Vestenamer-modified asphalt was measured as 36. Although the penetration test itself has chances of error, it was obvious that 36 is relatively much lower than the standard penetration degree of general asphalt, 60~70. It has been shown during Marshall stability tests that optimal viscoelasticity was measured when Vestenamer content was under 5%. Similar results were shown on the penetration test, where 5% Vestenamer-modified asphalt showed a penetration degree of 24 and a gradual decrease in the degree, as close as that to pure Vestenamer, when the Vestenamer content was above 20%. Using Vestenamer as a modifier for asphalt, the penetration degree decreased noticeably, which is clear evidence for improved resistance against plastic deformation.

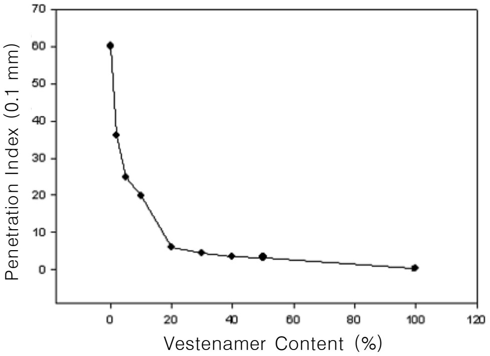

Figure 5. Dependence of penetration index of modified asphalts on Vestenamer content.

4. Reaction between Asphalt and Vestenamer

4-1 IR Analysis

An IR test was conducted to check for reactions between asphalt and Vestenamer. The results are shown on Figure 6. Comparing the two figures, it is evident that the graphs of two compounds are hardly discernible and that the chemical compositions of the two compounds are very similar to each other with most of the chemical composition being aliphatic hydrocarbons(e.g., around 3000 cm^{-1} for C-H, 1460cm^{-1} for CH_2, and 1376cm^{-1} for CH_3 groups). This explains one of the reasons why UPR can't be a candidate for the asphalt modifier. Asphalt modifiers need to be similar to asphalt in chemical nature. Since absorbance at 1650cm^{-1} represents double bonds of vinyl groups, it is suggested that there are small amounts of vinyl groups in Vestenamer. However, such double bonds were not found in AP-5. Instead, as known that asphalt in-

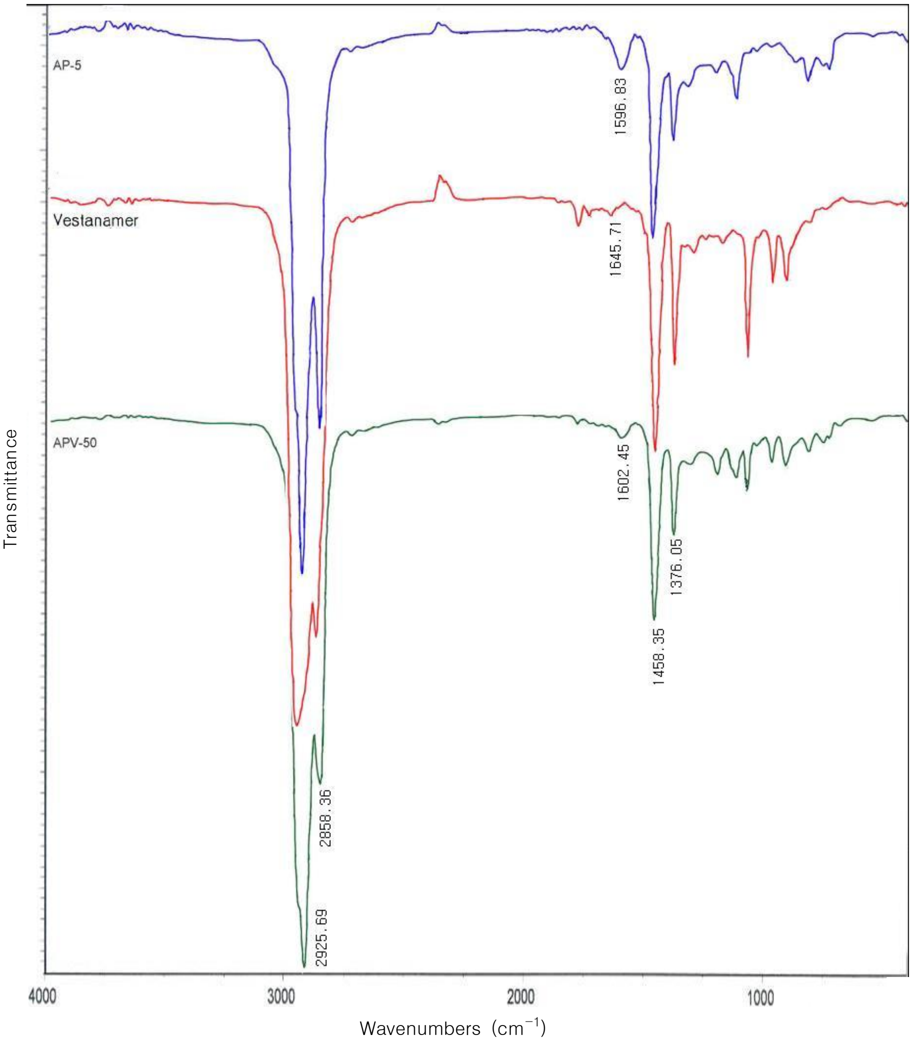

Figure 6. FT-IR graphs of AP-5, Vestanamer, and APV-50.

cludes small amount of aromatic chemicals, the aromatic double bonds were detected at 1600cm^{-1}. Thus, there is a high possibility that the double bonds of Vestenamer had reacted or linked with AP-5 through hydrogen abstraction reaction by peroxide and created a new compound with an improved property. In order to test this possibility, IR tests of 2% Vestenamer-modified asphalt (APV-2) were conducted. However, because of different concentrations between Vestenamer and AP-5, and the small amounts of vinyl groups in Vestenamer, it was difficult to decide whether a reaction had taken place or not. As a solution, IR tests of APV-50 which contained more Vestenamer were conducted. In this case, absorbance that represent asphalt, 1600cm^{-1}, remained intact, but absorbance that represented Vestenamer, 1650cm^{-1}, was lost. It is postulated then that reactions between AP-5 and Vestenamer are feasible due to possible radical formation in molecules in asphalt through hydrogen abstraction by

Tabel 6. Molecular Weights of Asphalts and Vestenamer

sample	Mn (g/mol)
AP	470
APV-2	560
APV-50	2750
V-100	530

peroxide and to double bonds in Vestenamer.

5. Molecular Weight Analysis

GPC was used to measure the molecular weight to decide whether a reaction had taken place or not. Table 6 depicts the average molecular weight of each specimen. The molecular weight of AP-5 and Vestenamer was 470 and 530g/mol, respectively. With a range of errors that GPC possesses, it can be considered that the molecular weights of the two are very similar to each other. 2wt% Vestenamer-modified asphalt(APV-2) had a slightly higher molecular weight of 560g/mol. This small increment is not only because the molecular weights of AP-5 and Vestenamer were as low as that of oligomer, but also because the concentration of Vestenamer was so low, so only a small amount of the vinyl groups in Vestenamer participated in the reaction, if the chain extension reaction had taken place. Thus, in order to see the extent of reactions between AP-5 and Vestenamer, the molecular weight of APV-50, modified asphalt with higher Vestenamer content, was measured. Since the molecular weight increased evidently as Vestenamer content increased, it is considered that there was an extension of molecular chains through the reaction of AP-5 and Vestenamer.

Ⅳ. Conclusion

The increase in the number of vehicles on roads, reduces durability of asphalt pavement. Thus modification of asphalt is needed for the asphalt to resist against deformation. Modified asphalt can ultimately beget safe driving and low road maintenance cost. This study on asphalt modification drew the following conclusions.

1. From Marshall tests, Vestenamer was proven to be a good candidate as an asphalt modifier. It was compatible with asphalt, and has the ability to improve adhesion in asphalt. As Vestenamer content increased, Marshall stability also increased. At a Vestenamer content lower than 5%, the modified asphalt showed viscoelasticity, a good characteristic for a road pavement material, and resistance to plastic deformation. The optimal amount was proven to be 2%.
2. When GRT was used with Vestenamer, asphalt showed dramatically improved resistance against plastic deformation. From Marshall tests, 1% GRT showed the best result against plastic deformation.
3. Higher viscosity and lower penetration degrees of modified asphalt, compared to those of AP-5, are other evidence for improved resistance against plastic deformation.
4. TMA test at 60°C, a temperature for measuring plastic deformation, indicated that modified asphalt was less soft than pure AP-5. This is evidence that Vestenamer-modified asphalt had a greater resistance against plastic deformation than AP-5.
5. GPC tests showed an increment in molecular weight through the modification of asphalt, which also implied the possibility of chain extension reactions.

Acknowledgment

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