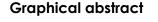
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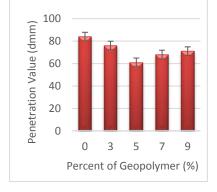
PHYSICAL PROPERTIES AND STORAGE STABILITY OF GEOPOLYMER MODIFIED ASPHALT BINDER

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Abstract

This study was conducted to investigate the physical properties and storage stability of the 80/100 penetration grade asphalt modified with geopolymer. In this research, fly ash and alkali activators, namely sodium silicate (Na₂SiO₃) and sodium hydroxide (NaOH), were used as geopolymer components. The penetration, Ring and Ball softening point, ductility, and viscosity tests were conducted to determine the physical properties of geopolymer modified asphalt (GMA). Five samples of asphalt binders with varying percentages of geopolymer, namely 0, 3, 5, 7 and 9%, by weight of asphalt binder were studied. Results show that geopolymer has good compatibility with asphalt binder. The addition of geopolymer into asphalt binder resulted in improved permanent deformation resistance of the modified binder compared to that of the conventional asphalt. In conclusion, geopolymer could be considered as a potential alternative in the modification of the properties of asphalt binder.

Keywords: Storage stability, physical properties, geopolymer-modified asphalt, permanent deformation, asphalt, fly ash

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1.0 INTRODUCTION

Asphalt binder is a product obtained through the distillation process of crude petroleum; it is a sticky, dark brown or black, semisolid material, and is highly viscous at ambient temperature [1, 2]. This material is widely used in paving, roofing, and road application. Over 96% of the world's road networks is surfaced with hot-mix asphalt (HMA) [3]. However, the increase in economic activity in recent years, which has led to an increase in road traffic volume and heavier vehicle load has resulted in the need to enhance the quality of existing asphaltic materials [4, 5].

Several methods are available to improve the properties of asphalt binders. One of the methods modify asphalt binders by using polymer as a modifier agent to produce polymer modified asphalt binder [6].Many studies have been conducted throughout the world to investigate the effects of polymer, such as Styrene Butadiene Rubber (SBR), Chloroprene Rubber (CR), polyethylene, Ethylene-Vinyl-Acetate (EVA), and Natural Rubber (NR), on modified asphalt binder [2, 7-10]. For example, the use of rubber modified asphalt binder was found to dramatically improve toughness, low temperature properties, and elastic properties in comparison to those of the base binder. Ethylene-Vinyl Acetate (EVA), polyethylene (PE), and or other thermoplastic resin modified asphalt binder dramatically improve asphalt binder's properties at high temperature and produced better temperature sensitivity [11-14].

In view of the large amount of modified asphalt binder is used in road construction, the cost of the modifying agent must be of prime consideration [15, 16]. Fly ash is a by-product of the electric power industry, and this waste material could be recycled and used as a road construction material. The cost of this modifying agent is relatively low and the fact that it is a recycle material lower the cost even further [1]. Currently, fly ash is produced in huge amounts and the

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demand for dumping areas for the disposal of this material is high. The Malaysian Department of Environment (DoE) reported that approximately 2 million metric tons fly ash are generated in Malaysia annually. The use of the alternative materials such as fly ash would not only improve the properties and durability of asphalt binder, but it would also be costeffective and environmentally friendly [17].

In this study, alkaline activator and fly ash were used as the geopolymer components of the geopolymerization process. This research shifted from the chemistry domain to engineering application and commercial production of geopolymer modifier. This paper discusses the physical properties and storage stability of Geopolymer Modified Asphalt (GMA). The following section discusses the details of the tests and the results of the tests.

2.0 EXPERIMENTAL DESIGN

2.1 Materials

2.1.1 Virgin Asphalt

The virgin 80/100 penetration grade asphalt binder was used as a control sample in this study. The physical properties of the virgin asphalt binder is shown in Table 1. The physical properties of the binder meet the minimum requirements set by the standard specification.

Table 1 Physical properties of 80/100 penetration grade asphalt

Property	Standard	Limits	Value
Penetration (25°C, 0.1mm)	ASTM D 5	80- 100	84
Ductility (25°C, cm)	ASTM D 113	100 mini mum	≥100
Softening point (°C)	ASTM D 36	47 mini mum	47.0
Specific gravity (g/cm³)	ASTM D 70	1.00- 1.05	1.03

2.1.2 Fly Ash

The fly ash used in this study was obtained from the Sultan Salahuddin Abdul Aziz Power Station owned by Kapar Energy Ventures Sdn Bhd in Selangor, Malaysia. Based on the ASTM C 618 standard, the fly ash is classified as class F fly ash with a specific gravity of 2.26. The chemical oxide composition of the fly ash is given in Table 2.

Table 2 Chemical composition of class F fly ash (%)

Oxide	Fly ash
SiO ₂	52.50
Al2O ₃	22.82
Fe2O ₃	5.34
CaO	7.16
MgO	2.56
SO₃	0.20
K2O	0.99
Na ₂ O	0.48
LOI	3.35
Source: Chien Yete	et al [18]

Source: Chien Yet et al. [18]

2.1.3 Alkaline Liquid

The alkali liquid used in this study are sodium silicate (Na₂SiO₃) solution with Ms = 2.0 (Na₂O: 14.73%; SiO2: 29.75%; and H2O: 55.52%) and sodium hydroxide (NaOH) pallet diluted in water to produce 8 Molar (8M) NaOH solution. A combination of sodium silicate solution and sodium hydroxide solution was prepared to activate the alumino-silicate precursors in fly ash of dissolution-hydrolysisthrough a series polycondensation [19].

2.2 Preparation of Geopolymer Modified Asphalt (GMA)

In this study, the melt blending technique was used to prepare the Geopolymer Modified Asphalt (GMA). The first step in the process is by preparing the geopolymer gel by combining fly ash and alkaline solution. The virgin asphalt binder was heated at 150°C until it turns into liquid. The geopolymer gel was then added into the melted virgin asphalt binder and the mixture was sheared for 90 minutes at a speed of 1000±10 rpm to produce a homogenous mixture. In this study, the softening point and the viscosity tests were used to evaluate the homogeneity of the mixture [7,20]. A sample for each test was taken every 30 minutes during the 2h mixing time until the value for the softening point and viscosity stabilized. Conventional asphalt binder was used as a control sample and modifying agents were added in the amount of 0, 3, 5, 7, and 9% by weight of the asphalt binder.

2.3 Measurements

2.3.1 Physical Properties Test

The standard tests used to investigate the physical properties of GMA include the penetration (25°C), the Ring and Ball softening point, and the ductility (25°C) tests which were conducted in accordance with ASTM D5, ASTM D36, and ASTM D113 respectively. The penetration test is a common control test for penetration grade asphalt and the values obtained represent the hardness and consistency of asphalt binders. The softening point corresponds to the temperature at which GMA began to turn into fluid. Ductility is identified as the maximum distance to

which a briquette specimen could be elongated without breaking under increasing tensile forces at a specified rate and temperature.

2.3.2 Temperature Susceptibility

Asphalt binder is a thermoplastic material and its consistency changes with temperature. Temperature susceptibility is the change in the rheological properties of asphalt binders with any change in temperature; it is a very important property as the behaviour of the asphalt depends on the rate of loading and temperature. There are two different ways of measuring temperature susceptibility, namely the Penetration Index (PI) and the Pen-Vis Number (PVN). The values for PI and PVN were calculated using Equations 1 and 2 respectively [20,21]:

$$PI = \frac{1952 - 500 \log pen - 20 \text{ softening point}}{50 \log pen - \text{ softening point} - 120}$$
(1)

Pen is the penetration value measured at 25°C

$$PVN = \frac{\log L - \log X}{\log L - \log M} (1.5)$$
⁽²⁾

where, $\log X =$ the logarithm of the viscosity in centistokes measured at 135°C, $\log L =$ the logarithm of the viscosity at 135°C for a PVN of 0.0, and $\log M =$ the logarithm of the viscosity at 135°C for a PVN of -1.5. However, Equations 3 and 4 (based on least-square fits) could be used to calculate more accurate values of *L* and *M*.

The equation for the line representing PVN = 0.0 is:

$$\log V = 4.25800 - 0.79670 \log P \tag{3}$$

The equation for the line representing PVN = -1.5 is:

$$\log V = 3.46289 - 0.61094 \log P \tag{4}$$

where, V = viscosity in centistokes at 135°C and P = penetration at 25°C.

2.3.3 Viscosity

A Brookfield rotational viscometer was used to measure the rotational viscosity of asphalt binder according to the ASTM D4402 standard. The test was done on 10.5 g (\pm 0.5 g) asphalt binder using a spindle 21 with a constant speed of 20 rpm to obtain the viscosity value. In this study, two testing temperatures, 135°C and165°C, were applied.

2.3.4 Storage Stability Test

The storage stability test was conducted in accordance with the ASTM D5976 standard. A portion of the prepared GMA sample was transferred into a 16 cm-high and 3 cm-diameter vertical aluminum-foil

tube. The foil tube was closed and stored vertically in an oven at $163 \pm 5^{\circ}$ C oven for a period of 48 ± 1 hours; the sample was then taken out and immediately placed in a freezer at $-6.7 \pm 5^{\circ}$ C for a minimum of four hours to completely solidify the sample. When the sample has cooled to room temperature, the sample was cut horizontally into three equal sections. Finally, the storage stability of GMA sample was determined by measuring the difference between the softening point of the top and the bottom sections of each samples [1].

3.0 RESULTS AND DISCUSSION

3.1 Physical Properties of GMA

The effect of geopolymer on the properties of GMA was determined from the physical properties indexes of the virgin and modified asphalt binders as shown in Figures 1-3. Figure 1 clearly shows that the addition of geopolymer altered the consistency of the asphalt binder. The penetration values tend to decrease gradually when 3% and 5% geopolymer were added to the virgin asphalt binder. The penetration value for GMA with 3% and 5% geopolymer are approximately 8dmm and 23dmm lower than the penetration value for virgin asphalt (84dmm). However, the addition of 7% and 9% geopolymer into the virgin asphalt binder increased the penetration value since these percentages of geopolymer altered the system of the asphalt binder. The addition of geopolymer into the virgin asphalt increased the consistency of modified binders. The hardening of the asphalt binder could be beneficial as it increased the stiffness of the material, which in turn affects the load spreading capabilities of the structure; however this could also lead to fretting of cracking [23].

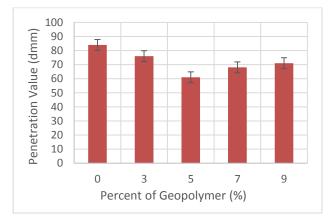


Figure 1 Effect of adding geopolymer on measured penetration values

Figure 2 shows that the softening point fluctuates across all geopolymer concentration. Modification of asphalt binder with 3% and 5% geopolymer increased the softening point from 47°C to 49°C and 56.5°C respectively. The trend for the penetration value shows that the addition of approximately 7% and 9% geopolymer by weight of asphalt binder decreased the softening point value as the penetration value increased. This indicates that the resistance of asphalt binder towards the effect of heat increased and this reduces its tendency to soften in hot weather. Thus, the addition of geopolymer reduces the susceptibility of modified asphalt binder toward temperature change. The addition of geopolymer into the asphalt mixture is expected to decrease the rate of rutting due to the increase in softening point.

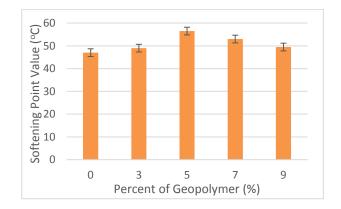


Figure 2 Effect of adding geopolymer on measured softening point values

In this study, the ductility of GMA was observed to gradually decrease with the increase in polymer concentration, as shown in Figure 3. This indicates an increase in the stiffness of GMA.

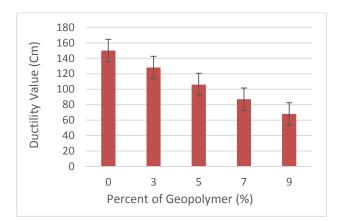


Figure 3 Effect of adding geopolymer on measured ductility values

Table 3	3 PI	and	PVN	of	GMA
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Geopolymer content (%)	PI	PVN
0	-0.70	-0.60
3	-0.42	-0.47
5	0.81	-0.56
7	0.30	-0.54
9	-0.47	-0.50

Table 3 shows the values of PI and PVN for GMA. PI and PVN values are frequently used to estimate the expected temperature susceptibility of asphalt binders [7]. The lower the PI and the PVN values, the higher the temperature susceptibility [7,15,20,21]. The PI and PVN for most paving asphalt binder are between +1 to -1 and +0.5 to -2.0 respectively [21]. In this study, the addition of geopolymer increased PI values and the highest value (0.81) was obtained when the virgin asphalt binder was modified with 5% geopolymer, which in turn increased the PVN values. Therefore, using geopolymer as an asphalt modifier decreases the susceptibility of the asphalt's temperature.

3.2 Viscosity

Figure 4 shows the results obtained from the rotational viscosity test carried out on virgin asphalt and GMA. As expected, the viscosity values for all types of asphalt binders decreased as the temperature was increased from 135 to 165 °C. Figure 4 also shows that asphalt binders modified with geopolymer increased the values of the binder's viscosity. GMA with 5% geopolymer has the highest viscosity value (135°C: 0.460 mPa.s, 165°C: 0.169 mPa.s).

Viscositv determines the flow characteristics of asphalt binders [20]. Asphalt binders must be sufficiently fluid at high temperatures so that they could be easily pumped and handled during production and during the laying of asphalt mixtures for paving constructions. The result shows that higher than normal mixing and compaction temperatures required when a high percentage of were geopolymer (5%) was added to the virgin asphalt binder. Furthermore, the increase in viscosity yielded greater stiffness at high pavement service temperatures and they might also be beneficial in improving resistance towards rutting [22,23].

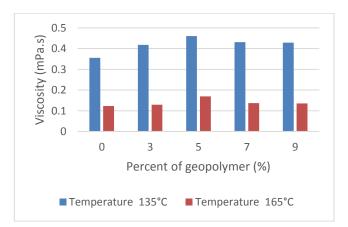


Figure 4 Effect of adding geopolymer on measured viscosity values

3.3 Storage Stability of GMA

Yu *et al.* [1] stated that good storage stability of asphalt binders are obtained when the difference in the values of the softening point between the top and the bottom sections is small (less than 2.5°C). The softening point value for the top and the bottom sections of the GMA are shown in Table 4. The results show that the difference between the top and the bottom sections for all samples did not exceed the permitted value, which imply that these modified asphalt binders could remain stable at high storage temperature.

Table 4 Softening point for the top and the bottom sections of GMA

	Percent of	Softening point		ΔSP
Sample	Geopolymer (%)	Top (∘C)	Bottom (°C)	(°C)
1	3	51	50	1
2	5	60	60	0
3	7	55	54	1
4	9	52	52	0

4.0 CONCLUSIONS

The effect of using geopolymer as an asphalt modifier was investigated in this study. The limited laboratory work conducted showed that the addition of geopolymer into the virgin asphalt binder increased the hardness of the asphalt binder. It may be inferred that GMA provides better resistance against rutting due to their higher softening points when compared to virgin asphalt binder. Additionally, this research could also contribute towards reducing the size of dumping areas for the disposal of fly ash which, in turn, protect the environment. As a conclusion for this investigation, the best result was recorded for GMA with 5% geopolymer. This concentration could be considered as the optimum GMA.

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References

- R. Yu, C. Fang, P. Liu, X. Liu, and Y. Li. 2015. Storage Stability and Rheological Properties of Asphalt Modified With Waste Packaging Polyethylene and Organic Montmorillonite. *Appl. Clay Sci.* 104: 1–7,
- [2] D. Lesueur. 2009. The Colloidal Structure of Bitumen: Consequences on The Rheology and on The Mechanisms of Bitumen Modification, Adv. Colloid Interface Sci. 145(1–2): 42–82.

- [3] M. N. Borhan, A. Ismail, R. A. Rahmat, and F. Suja. 2007. Used Cylinder Oil Modified Cold-Mix Asphalt Concrete. J. Appl. Sci. 7(22): 3845–3491.
- [4] A. Tennøy. 2010. Why We Fail to Reduce Urban Road Traffic Volumes: Does it Matter How Planners Frame The Problem?. *Transp. Policy*. 17(4): 216–223.
- [5] Y. Liu, J. Wu, and J. Chen. 2014. Mechanical Properties of a Waterproofing Adhesive Layer Used on Concrete Bridges Under Heavy Traffic and Temperature Loading. Int. J. Adhes. Adhes. (48): 102–109.
- [6] Y. Yildirim. 2007. Polymer Modified Asphalt Binders. Constr. Build. Mater. 211: 66–72.
- [7] R. a. Al-Mansob, A. Ismail, A. N. Alduri, C. H. Azhari, M. R. Karim, and N. I. M. Yusoff. 2014. Physical and Rheological Properties of Epoxidized Natural Rubber Modified Bitumens. Constr. Build. Mater. 63: 242–248.
- [8] G. Polaccoa, P. Křížb, S. Filippia, J. Stastnab, D. Biondia, and L. Zanzotto. 2008. Rheological Properties of Asphalt/SBS/Clay Blends. *Eur. Polym. J.* 44(11): 3512–3521.
- [9] C. E. Sengul, S. Oruc, E. Iskender, and A. Aksoy. 2013. Evaluation of SBS Modified Stone Mastic Asphalt Pavement Performance. Constr. Build. Mater. 41: 777–783.
- [10] D. Casey, C. McNally, A. Gibney, and M. D. Gilchrist. 2008. Development of a Recycled Polymer Modified Binder for Use in Stone Mastic Asphalt. *Resour. Conserv. Recycl.* 52(10): 1167–1174.
- [11] C. Fang, S. Zhou, M. Zhang, S. Zhao, X. Wang, and C. Zheng. 2009. Optimization of the Modification Technologies of Asphalt by Using Waste EVA From Packaging. J Vinyl Addit. Technol. 15: 199–203.
- [12] M. Garcia-Morales, P. Partal, F. J. Navarro, F. J. Martinez-Boza, and C. GallegoS. 2007. Processing, Rheology, and Storage Stability of Reclyled EVA LDPE Modified Bitumen. *Polym. Eng. Sci.* 47(2): 181.
- [13] P. H. Yeh, Y. H. Nien, J. H. Chen, W. C. Chen, and J. S. Chen. 2005. Thermal and Rheological Properties of Maleated Polypropylene Modified Asphalt. *Polym. Eng. Sci.* 45(8): 1152–1158.
- [14] G. R. Harish and M. N. Shivakumar. 2013. Performance Evaluation of Bituminous Concrete Incorporating Crumb Rubber and Waste Shredded Thermoplastics. Int. J. Res. Eng. Technol. 2321(7308): 233–238.
- [15] J. Guo, J. Guo, S. Wang, and Z. Xu. 2009. Asphalt Modified With Nonmetals Separated from Pulverized Waste Printed Circuit Boards. Environ. Sci. Technol. 43(2): 503–508.
- [16] F. J. Navarro, P. Partal, F. Martínez-Boza, and C. Gallegos. 2014. Thermo-Rheological Behaviour and Storage Stability of Ground Tire Rubber-Modified Bitumens. *Fuel.* 83: 14–15 SPEC. ISS: 2041–2049.
- [17] N. S. Mashaan, A. H. Ali, S. Koting, and M. R. Karim. 2013. Performance Evaluation of Crumb Rubber Modified Stone Mastic Asphalt Pavement in Malaysia, Adv. Mater. Sci. Eng. 2013: 1–8.
- [18] T. Chien Yet, R. Hamid, and M. Kasmuri. 2012. Dynamic Stress-Strain Behaviour of Steel Fiber Reinforced High-Performance Concrete with Fly Ash. Adv. Civ. Eng. 2012: 1– 6.
- [19] A. Kusbiantoro, M. S. Ibrahim, K. Muthusamy, and A. Alias. 2013. Development of Sucrose and Citric Acid as the Natural based Admixture for Fly Ash based Geopolymer. *Procedia Environ. Sci.* 17: 596–602.
- [20] S. I. A. Ali, A. Ismail, N. I. M. Yusoff, M. R. Karim, R. A. Al-Mansob, and D. I. Alhamali. 2015. Physical and Rheological Properties of Acrylate–Styrene–Acrylonitrile Modified Asphalt Cement. Constr. Build. Mater. 93: 326–334.
- [21] E. R. Brown, P. S. Kandhal, F. L. Roberts, Y. R. Kim, D.-Y. Lee, and T. W. Kennedy. 2009. Hot Mix Asphalt Materials, Mixture Design and Construction, Third Edit. Lanham, Maryland: NAPA Research and Education Foundation.
- [22] E. Santagata, O. Baglieri, L. Tsantilis, and D. Dalmazzo. 2012. Rheological Characterization of Bituminous Binders Modified with Carbon Nanotubes. *Procedia - Soc. Behav. Sci.* 53: 546–555.

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[23] A. N. Amirkhanian, F. Xiao, and S. N. Amirkhanian. 2011. Characterization of Unaged Asphalt Binder Modified with

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Carbon Nano Particles. Int. J. Pavement Res. Technol. 4(5): 281–286.