The effects of polymer modified asphalt binder

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The effects of polymer modified asphalt binder incorporating with chemical warm mix additive towards water quality degradation

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ABSTRACT

Naturally, asphalt binder has compacting and mixing temperatures higher than 150 °C. Both temperatures can be increased by modifying the pristine penetration grade asphalt binder with polymer. However, the warm-mix asphalt (WMA) technology contributes to lowering of these temperatures even though the possibility of environmental effect cannot be neglected. Air pollution and water contamination are two crucial environmental concerns when it comes to chemical reactions of any materials. This study was conducted to investigate the physical characteristics, thermal analysis, leaching and toxicity of polymer-modified asphalt binder (PMA, PG76) and its performance as the binding agent. Physical tests were carried out to determine the durability, quality and performance of PG76 binder. Thermal analysis via thermogravimetry analysis was carried out to observe the stability of the blending at higher temperature. The results illustrated that even though the PMA is added in combination with WMA as additives, the mixes were stable beyond their mixing and compaction temperatures. The decomposition started at temperature >360 °C and ended at temperature <500 °C for all samples. Wetting and spreading behaviours of the blending are also perceived by contact angle method. Increasing amount of WMA allowed the spreading of water on the surface. The surface was deemed to be hydrophilic or poorly hydrophobic. Leaching Tank Test was carried out to simulate water suspension during heavy rainfall and its influence in leaching traces of heavy metal from asphaltic binders. The tests were carried out at 0.25, 1, 2.25, 4, 9, 16, 36, and 64 days to observe the leaching behaviour of heavy metals and determine the toxicity of the samples. Inductively Coupled Plasma Mass Spectrometry (ICP-MS) was used to detect the presence of heavy metals (Cu, Zn, Ni, Pb, Cr and Fe) and the readings were analysed to Heavy Metal Evaluation index (HEI), Heavy Metal Pollution Index (HPI), Degree of Contamination (C_d), and Metal Quality Index (MQI). The cumulative values of heavy metals are presented in the form of logarithmic graphs. All indices indicated that all samples did not contribute to heavy metal pollution to the surrounding water resources.

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1. Introduction

Since the introduction of polymer-modified asphalt binder (PMA), it has been widely used to produce asphalt pavements owing to the excellence resistance to moisture, rutting, and fatigue (Zhu et al., 2014). Tariq Ali et al. (2013) has suggested that adding polyethylene into virgin asphalt binder will increase the heat resistance of the binder and the adhesion between the asphalt

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binder-aggregate system. Styrene-butadiene-styrene (SBS), on the other hand, is usually used to modify the asphalt binder by improving the thermal susceptibility, increases the softening point and increases stiffness upon aging. On the other hand, thermosetting PMA binders show high elasticity but lacking of viscous flow behaviour (Porto et al., 2019). However, when PMA is added filled with nano silica, the moisture susceptibility of the mixes improved, signifying the increase in strength. In addition, substantial increase in fatigue life and rutting deformation was observed (Yusoff et al., 2014). Also, due to high-temperature rheological studies and the technical indexes, terminal blend amorphous poly alpha olefin (TB-APAO) PMA and waste tire rubber-amorphous-poly-alpha-olefin (WTR-APAO) PMA performed better than SBS PMA (Yan et al., 2019). However, by means of PMA, with the increase in the production temperature, unnecessary oxidation, polymer phase separation, and asphalt; the degradation of the binders can lead to noteworthy green-house effect, emission of aerosols density and vapours to environment. The effect can be treacherous to human (Boczkaj et al., 2014; Pesch et al., 2011). Consequently, by reducing the compaction and mixing temperatures, the benefit of PMA can be further explored (Widyatmoko et al., 2016).

Warm-mix asphalt (WMA) additive can facilitate the process by reducing viscosity of the asphalt binder resulting in lower production temperature (Rahmad et al., 2020; Yusoff et al., 2013). WMA additive comes in various types namely; chemical based, water zeolite based and wax based; such as Rediset, Advera, and Sasobit (Menapace and Masad, 2014; Xu et al., 2017). Despite the fact that WMA additives are mainly used for the purpose of reducing viscosity of the asphalt binder, other effects such as aging, alteration of rheological properties and mechanical characteristics in binders and mixes are also studied (Akisetty et al., 2009; Xiao et al., 2012). Each WMA additive has a unique function in the binder system. Even so, the WMA additive selection is due to its effects on viscosity of asphalt binders and workability of asphalt mixes. However, the rheological performance of asphalt binders should not be affected by the WMA additive. The extensive use of chemical WMA additives has raised environmental concern since the additive is often associated with various chemical contaminants in order to achieve the preferred results. A recent study has shown that the chemicals present in WMA have a positive impact in suppressing the emission of gaseous pollution during its manufacture (Abdullah et al., 2016). However, there is a dearth of information on the leachability of heavy metals utilised in WMA technology. Recent developed technology has successfully produced WMA from a mixture of chemical and organic additives (Hill et al., 2012). However, very few researches have been carried out to determine the leachability of heavy metal components in WMA. In addition to gas emission, unmonitored problematic road pavements may lead to uncontrolled heavy metals leaching that contribute to environmental pollution.

In tropical countries, pavements are being exposed to high daily ambient temperature throughout the year. For instance, daily temperature for some countries ranges from 30 °C to 40 °C (Zawawi et al., 2011), However, during the monsoon season, some places are prone to flood known as monsoonal flood that could last up to two weeks. The average annual rainfall can be up to 2400 mm with the highest at 358.0 mm as recorded by Aminah Shakirah et al. (2016) occurred for 24 h. Exposed asphalt binder has the tendency to be abraded away and the oxidised material is soluble in rainwater (Hunter et al., 2015). Chen et al. (2018) discovered that asphalt binder can leach metallic components when soaked in water. The pH value of the water and submersion time are two parameters that influence the concentration of metallic elements and determine the degree of water contamination. This study is in agreement with the one conducted earlier by Legret et al. (2005) where they identified the existence of leachate were due to infiltration of rainwater through the pavement surface. There are a handful of leaching tests used to study the effect of asphalt to the environment. Aqilah et al. (2020) conducted the Synthetic Precipitation Leaching Procedure (SPLP) test to study the mobility of organic and inorganic elements present in asphalt binders modified with polyurethane and Cecabase additives for WMA application. The new material has been proven to be environmentally safe. Meanwhile, Hassan and Khalid (2010) studied the environmental characteristics of bituminous mixtures with incinerator bottom ash aggregates by carried out tank leaching test in accordance to NEN7375 testing standard. This study concluded that a model developed from the test can predict cumulative release from binder sample.

The heavy metals components of any construction material have to be analysed in an effort to protect the environment from any form of pollution caused by the leached metals. These heavy metals can be classified as non-critical, very critical and abundant as they are easily found, and toxic or very insoluble or very rare (Wood, 1974). The degree of hazard associated with a material can be established based on this classification. Zinc (Zn), argentum or silver (Ag), cadmium (Cd), cuprum or copper (Cu), plumbum or lead (Pb), cobalt (Co), and nickel (Ni) are classified as heavy metals with high potential to pollute the environment and caused health issues due to their high toxicity. Heavy metals can cause many health complications due to their ability to be absorbed by plants through the soil and underground water, and these plants are then consumed by humans (Singh et al., 2010). Road pavements are structures that are heavily affected by surrounding conditions as they are constantly exposed to hot and cold temperatures. Pavements must be durable and able to withstand the dynamic loads exerted as vehicles pass on them. Because of this, asphaltic pavements are added with chemicals that function to enhance their flexibility. Synthetic zeolites that are added in the foaming process are composed of alumina-silicates of alkali metals and have been hydro-thermally crystallized (Vidal et al., 2013). The application of WMA technology for practical use may cause leaching as a result of the influence of environmental factors, especially during heavy rainfall and flash flood. Tank leaching test is one of the methods used to analyse the heavy metals leached from a particular substance. It is carried out by adding an acidic solution under specific laboratory conditions. Leaching test is often used to simulate the submersion of a solid in water for a certain period of time (Chai et al., 2009) and the result can be used to estimate the amount of leached solids. The leachate is then analysed using the latest equipment and tools, including Atomic Absorption Spectroscopy (AAS) and Inductively Coupled Plasma Mass Spectrometry (ICP-MS) (Fadzil et al., 2011; Gómez and Bosecker, 1999).

The objective of this study was to determine the physical characteristics of PG76 modified with Rediset. The physical properties of the mixture were determined through penetration, softening point, and ductility tests and by conducting tank leaching test to simulate heavy rainfall and flash flooding on the road by submerging the sample in acidic solution (pH of 4) for different periods of time (0.25, 1, 2.25, 4, 9, 16, 32, and 64 days) as suggested by Fadzil et al. (2011) and in accordance to NEN 7347, 2006 testing standard. The leachate was analysed using ICP-MS instrument and the cumulative release of Cd, Cu, Zn, Ni, Pb, Cr, Al, and Co were tabulated and compared with the values set by the Malaysia Department of Environment, DOE to determine its toxicity. Thermal analysis of the new material was also carried out to study their characteristics upon exposure to high temperatures.

2. Experimental design

2.1. Materials and sample preparation

A performance grade asphalt binder namely PG76 and warm mix asphalt (WMA) additive are used in this study. The WMA additive namely Rediset LQ 1106 (Rediset) was supplied by AkzoNobel. This additive is known to be capable of increasing the stability (Vahora and Mishra, 2017), tensile strength (Leng et al., 2014), anti-rutting properties and life-span (AkzoNobel, 2014). PG76 is modified by adding Rediset at varying percentage of 1, 2, 3, 4, and 5 wt% of total binder weight. The blending is labelled as R0 for the control sample, R1, R2, R3, R4, and R5 with reference to Rediset weight added. The binder and Rediset were mixed at 160 °C for 30 min in a rotary shear mill at a speed of 1250 rpm. The physical characteristics of PG76 binder is presented in Table 1. The physical properties of Rediset LQ1106 are presented in Table 2.

2.2. Physical tests

Physical tests, including penetration (ASTM D5) to determine the hardness and consistency of the bituminous material when it is reheated and softening point tests (ASTM D36), to establish the point at which the bituminous substance starts to soften are carried out for this study. The results of the tests are compared to establish the fluidity of the binders. The ductility test (ASTM D113) is used to measure the tensile properties of both asphaltic materials. For each test, three replicates sample were tested and the values were taken as the average readings.

2.3. Wettability via contact angle

Wettability is defined as the attraction of a liquid phase to solid surface, and it is typically quantified using a contact angle with the solid phase. A sessile drop device called goniometer was used to measure contact angles of water on the surface of the blending to characterise its spreading and wetting characteristics.

2.4. Thermogravimetry analysis

Thermogravimetric Analysis (TGA) was carried out for this study to assess the weight change of the sample with the function of temperature under controlled conditions. The changes in weight was observed up to 600 °C. The analysis was carried out using Shimadzu TGA-50 thermogravimetric analyser in a protective atmosphere of nitrogen gas, with a flux of 50 ml/min and a heating rate of 10 °C/min. The analysis was carried out in accordance to ASTM E 1311 (ASTM, 2012).

2.5. Tank leaching test

The tank leaching test was carried out to determine the leaching of the new materials. The test was conducted in accordance to the Netherlands tank leaching test NEN 7345. This test was meant to simulate the condition of which the new material submerged in rainwater during the monsoon season. However, the test was modified to suit the study purpose. The materials were left submerged until extraction time. This way, the worst flood scenario is imitated. As for the sample preparation, the binder samples were poured to fill the bottom part of the bottle followed by initial weighing. Fig. 1 shows a sample ready to be submerged in acidic solution for further analysis while Fig. 2 shows the samples during the test. The contaminants were then analysed by inductive coupled plasma mass spectrometry (ICP-MS). A total of 29 elements were analysed by ICP-MS.

Table 1
Physical properties of PG76.

Parameter	Unit	Value	Requirement	Test Standard
Softening point	°C	93	Min. 70 °C	ASTM D36
Penetration	0.1 mm	46	Min. 45	ASTM D5
Viscosity at 135 °C	Pa s	2.45	Max. 3 Pa s	ASTM D4402

Source: Yusoff et al. (2014).

Table 2
Physical properties of Rediset LQ 1106 (AkzoNobel, 2014).

Item	Index
Appearance @25 °C	Liquid
Colour	Dark brown
Odour	Slight
pH at 0.1% solution	10
Pour point, °C	3
Flash point, °C (Pensky Martens)	165
Boiling point, °C	215
Viscosity @40 °C, mPa s	216
Density @40 °C, g/cc	0.99
Solubility in water	Partly soluble

2.5.1. Heavy metal evaluation index (HEI)

Heavy Metal Evaluation Index (HEI) gives the water overall quality with respect to heavy metals. The values are categorised into 3 classes to specify the level of contamination. They are identified as low (HEI<400), medium (HEI = 400–800), and high (HEI>800) (Edet and Offiong, 2002). Equation (1) is used to calculate HEI.

$$HEI = \sum_{i=1}^{n} H_c \Big/ H_{mac} \tag{1}$$

In this equation, H_c is the monitored value of the ith parameter while H_{mac} is the ith parameter maximum admissible concentration.

2.5.2. Heavy Metal Pollution Index (HPI)

The Heavy Metal Pollution Index (HPI) is developed in two stages; a weight (Wi) that is allocated for selected parameter as rating scale and base to compute the index by selecting the pollution parameter. HPI is calculated by equation (2).

$$HEI = \frac{\sum_{i=1}^{n} WiQi}{\sum_{i=1}^{n} Wi} \tag{2}$$

where W_i is the ith parameter unit weightage and n denotes the

Fig. 1. A binder sample prepared for tank leaching test.

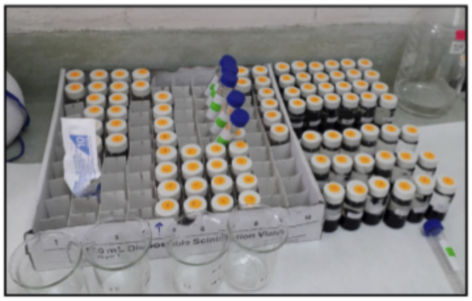

Fig. 2. The samples during tank leaching test.

amount of parameters considered in the calculation. Q_i represents the sub-index of parameter *i*th, calculated by the equation:

$$Q_i = \sum_{n=1}^{n} \frac{\{Mi(-)Ii\}}{(Si - Ii)} \times 100$$

In this case, M_i is the *i*th heavy metal parameter monitored while I_i and S_i are the ideal and standard values of the heavy metal respectively. The upper part of the calculation is in absolute number, meaning to say that the negative sign is ignored. The critical pollution index value is 100 for the water to be able for consumption (Prasad and Bose, 2001). Cr, Cu, Fe, Ni, Pb, and Zn were used to compute the indexes for this study.

2.5.3. Degree of contamination (C_d)

The degree of contamination (C_d) for each sample is calculated individually as the total of water contamination factor of each element that exceeds the upper permissible values. C_d values are classified into three groups, that are low ($C_d < 1$), medium ($C_d = 1-3$) and high ($C_d > 3$). C_d is calculated as in Equation (3):

$$C_d = \sum_{i=1}^{n} Cf_i \quad (3)$$

where Cf_i is the single contamination coefficient derived as:

$$C_{fi} = \frac{C_{Ai}}{C_{Ni}} - 1$$

From the equation, C_{Ai} denotes the measured value of ith element and C_{Ni} represents the maximum permissible limit of the element.

2.5.4. Metal quality index (MQI)

Equation (4) is used to calculate the metal quality index (MQI) (Tamasi and Cini, 2004). For this index, M_i is the heavy metal value obtained from the test while S_i is the maximum permissible value of the particular heavy metal.

$$MQI = \sum_{i=1}^{n} \frac{M_i}{S_i} \quad (4)$$

MQI represents the index of heavy metal concentration compared to its maximum permissible value. The higher the concentration will result a value > 1. It is the threshold value of which, the higher the value, the worse the quality of water.

3. Results and discussion

3.1. Physical tests

The results of penetration test are shown in Table 3. The penetration value increased with the incorporation of higher percentage of Rediset. The penetration values of the PG76 binder with the inclusion of 1, 2, 3, 4 and 5 wt% Rediset are 42.1, 47.5, 49.9, 55.0 and 60.8 dmm, respectively, while the value for the control sample is 46 dmm. Modifying PG76 binder with the addition of higher percentage of Rediset produced softer asphalt binder.

The softening point test was carried out according to ASTM D36. The softening point temperature for the control sample was 90 °C while the temperatures for the sample modified with 1, 2, 3, 4 and 5 wt% Rediset were 64.2, 62.3, 66.3, 61.7 and 68.7 °C, respectively. The softening point temperature decreased at higher Rediset content, with the exception of samples modified with 3 and 5 wt% Rediset. There is a relationship between softening point temperature and penetration value where binder with lower softening point temperature has higher penetration value. Higher percentage of Rediset content subsequently produces softer modified asphalt binder sample. The ductility test was performed according to ASTM D113 to determine the elasticity of modified asphalt binder. In this study the ductility value was taken as the length in centimeter at which the asphalt binder sample started to break. The minimum ductility value should be at least 100 cm. The obtained ductility values of the samples were not consistent. The control asphalt binder has a ductility value of 100.50 cm, while the values for the samples modified with 1, 2, 3, 4 and 5 wt% were 110.40, 97.77, 150, 108.50 and 120.67 cm, respectively. The result of ductility test shows that the values exceeded the minimum length required for elastic behavior of asphalt binder. Thus, the modified asphalt binder is suitable for use as pavement layer in terms of ductility measurement. The inconsistency in the ductility values could be due to non-uniformity in dispersion of Rediset in the binder system. This seems tally to the data obtained for the softening point temperature.

The physical tests indicated that R1 has a penetration value of 42.1 dmm and thus, is the best binder for use in regions with warmer climate. PG76 is preferred for use in regions with warmer climate due to its lower penetration grade. Binders with high penetration value are recommended for regions with colder climate. On the ther hand, softening point test indicated that PG76 with 5 wt% Rediset binder has the highest temperature of 68.7 °C while PG76 with 4 wt% Rediset binder has the lowest temperature of 61.7 °C. The softening point test is to determine the temperature at which the new material softens beyond some random softness. The results indicated that the incorporation of Rediset has an impact to the temperature of which physical property of the binder showed changes. Even though it reduced the softening point temperature in general, the reduction exhibited inconsistency. Meanwhile, for ductility test, the results show that R3, which PG76

Table 3
Physical tests data for modified PG76 with the inclusion of 1–5 wt% Rediset.

Sample	Penetration (dmm)			Softening Point (°C)			Ductility @ 25 °C		
	Ave	SD	CV	Ave	SD	CV	Ave	SD	CV
R0	46.00	1.58	3.44	90.00	1.00	1.11	100.50	5.00	4.98
R1	42.10	1.47	3.50	64.20	0.44	0.69	110.40	12.04	10.91
R2	47.50	2.89	6.09	62.30	1.60	2.57	97.77	13.57	13.88
R3	49.90	2.07	4.16	66.30	1.07	1.61	150.00	2.00	1.33
R4	55.00	1.17	2.13	61.70	1.34	2.17	108.50	16.31	15.03
R5	60.80	0.57	0.94	68.70	0.61	0.89	120.67	3.35	2.78

*Note: Ave: Average; SD: Standard Deviation; CV: Coefficient of Variation.

with 3% Rediset combination has the highest elasticity of 150 cm compared to other modified binders. On the other hand, R2 has the lowest elasticity of 97.77 cm.

3.2. Penetration index

The penetration index (PI) represents a quantitative measure of asphalt binder on how it reacts to various temperatures. The changes in penetration and softening point have been notably shown when calculating the Penetration index as a parameter of temperature susceptibility (Fig. 3). In this study, the addition of Rediset into PG76 affected the PI and changed the characteristic of the binder from low temperature susceptible with PI values above 2 to higher temperature susceptible binder. PI value increased to 2.2 again with 3 wt% and 5 wt% Rediset while with 4 wt%, the value decrease to below 2.0. It was clearly shown that since the softening point temperatures were not consistent; the penetration index gave the same pattern, even though the penetration values increased constantly.

3.3. Contact angles, wetting, and spreading

Wetting implies the tendency of a liquid to either blob or sheet on a surface. However, the behaviour is determined by the properties of the liquid and the surface. From the behaviour, a surface is classified as hydrophilic or hydrophobic. Water has remarkably large value of surface tension which can be attributed to the extensive hydrogen bonding interactions. Water molecule is attracted to surface with electrostatic charges (Bagampadde et al., 2004). By coating the aggregates with hydrophobic substances means a reduction in the water-aggregate adhesion free energy. Greater potential for hydrophilicity nature for aggregates will take place (Hamedi et al., 2020). In other words, for being hydrophobic, the asphalt binder will retard the water diffusion into the binder-aggregate system. Contact angle is one of the methods to measure wettability of a system (Good, 1992). For this study, the contact angles of the six samples are as shown in Fig. 4. It was obvious that with the increasing of WMA additive percentage, the contact angle between the water and material decreased. The control binder labelled as R0 has contact angle of 98.5° that clearly showed hydrophobic characteristic. Asphalt binder acted as a protective barrier to the aggregates from water damage. However, with time, water may diffuse into the binder and change the properties. This can lead to premature distress (Hung et al., 2017). When WMA additive being incorporated into the PMA, such as R1 with WMA additive of 1 wt% has a contact angle of 90.9°. This occurrence showed that the binder still has bad wetting or hydrophobic. As the

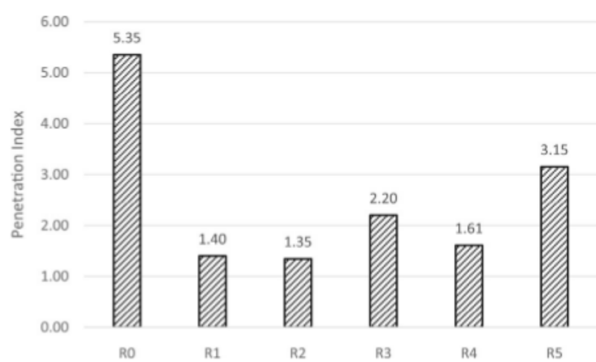

Fig. 3. Penetration Index of all tested binders.

content of WMA additive increased to 2 wt%, the contact angle decreased to 71.9°. The contact angles keep decreasing to 28.4° and dropped to 5.0° and finally to almost flat with about 2.0° when the quantity of WMA additive been added to 3, 4 and 5% respectively. The surface of the binders had turned to hydrophilic as the maximum percentage weight of WMA additive added into PG76 PMA. Omar et al. (2018) used this method to study the water damage resistance of nano-clay modified asphalt binder because the material is known to reduce water uptake. Their findings are satisfying and concluded that 4% of nano-clay particle is the optimum content to improve resistance to water damage.

The Spreading Parameter, S is a concept that enables the visualisation and understanding the effects of incorporation of Rediset into the PMA. In addition to the contact angle, the dispersion capability is also evaluated by the spreading parameter (Kalin and Polajnar, 2013), which, in general, represents the difference between adhesion work (W_a) and cohesion work (W_c), as in the following equation:

$$S = W_a - W_c = \gamma_s - (\gamma_{sl} + \gamma_l) \tag{5}$$

where W_a and W_c is the adhesion and cohesion work of the binder respectively and γ_s is the surface energy of the binder, γ_l is the surface energy of water, and γ_{sl} is the interfacial tension between binder and water. Table 4 lists the spreading parameter of each blending in this study. It is observed that for R0, when the contact angle was $\theta > 90°$, $S < 0$. This means that the surface was poorly wetted. As the content of Rediset increased, value of $S > 0$ and $\theta < 90°$. This showed that the surface is almost wetted. This result indicated that increasing content of Rediset allowed the spreading of water on the surface. The surface is deemed to be hydrophilic or poorly hydrophobic.

3.4. Thermogravimetry analysis

The thermogravimetric analysis curves (TGA and DTG) of R0, R1, R2, R3, R4, and R5 are shown in the thermograms (Fig. 5a–f). TGA curve represents the mass loss in the function of temperature. All samples show single step degradation. It clearly falls into the criteria of thermal decomposition with the formation of gaseous reaction products. Thus, it is noticeably seen that the plot has three zones. The first zone represents the elimination of moisture and volatiles compounds which sometimes refers to the drying phase while the second zone is the crucial part that signifies the decomposition of the materials. The third zone represents the sublimation process. On the other hand, a differential thermogravimetry curve (DTG) is generated together in the same plot as the first derivative of the weight with respect to temperature or time. The DTG curve is used to provide the peak height and temperature at maximum weight loss measurements for this study.

Fig. 5a represents the TGA and DTG curves for R0 – the control sample. Without any addition of Rediset, the binder starts to decompose at 406.5 °C while the peak temperature of maximum weight loss is at 463.6 °C. The decomposition process of R1 starts at temperature of 393.8 °C and the peak temperature is 459.5 °C. For other blending combinations, the decomposition starts at temperature more than 360 °C and ends at temperature less than 500 °C. It is noticeable that all the specimens show maximum weight loss at temperature ≥450 °C. As for the percent of weight loss during the process, R1 shows the highest with 91.66% while R3 gives the lowest value of 66.91%. Fig. 5d seems to show weight loss slightly exceeds 100%. Since asphalt binder volatilizes (gives off vapour) when heated, it will foam and make contact with the furnace wall. When this happen, the increasing of weight might occur seemingly. There were also some fluctuations in DTG curves. Fig. 5f shows

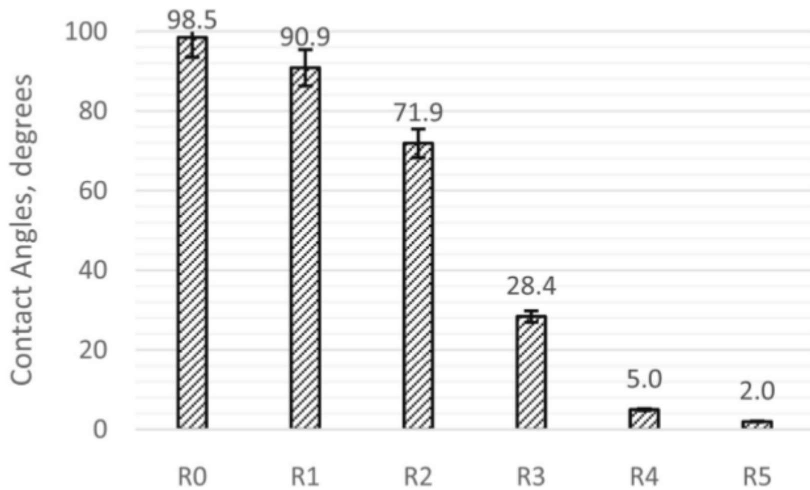

Fig. 4. Contact angles determined by goniometer for all the samples.

Table 4
Spreading parameter for all samples.

Sample ID	R0	R1	R2	R3	R4	R5
Spreading parameter, S	−109.26	−92.04	−49.28	35.64	67.52	70.76

several obvious fluctuations in the DTG curve. The fluctuations in DTG curve exist due to the disturbance during the mass loss during the thermal decomposition process because DTG curve represent the rate of change of mass. This noise might occur from physical effects of heat and mass transfer, minor reactions or chemical effect, or unforeseen experimental errors that affect the mass loss data such as sudden loss of mass resulting from the expulsion of part of the sample when it decays with the formation of gas. The results illustrated that even though PMA has been added together with WMA additive, the new binders are stable beyond their mixing and compaction temperatures.

3.5. Leaching test

3.5.1. Tank leaching

Charts presented in Fig. 6 are the cumulative concentration of each element obtained from ICP-MS at day 64 for every sample. The charts are in $\log_{10}$ axis to clearly show the overall the pattern of the analysis. Most elements detected at concentration of below 0.1 mg/m^2. There were also some elements detected to be in the range of 1.0–10.0 mg/m^2 identified as aluminium (Al), calcium (Ca), Iron (Fe), magnesium (Mg), strontium (Sr), and zinc (Zn). At the same time, potassium (K) and sodium (Na) (also known as Natrium) was found to be contributing the most weightage with concentration of more than 100 mg/m^2. However, K and Na are no threat to the public health. The recommended daily requirement for K is greater than 3000 mg and is seldom found in drinking water (World Health Organization, 2017) thus the trace of K found in the tested material can be neglected because there is no evidence of this element to pose any risk for the health of consumers. On the other hand, Na is a common component found in almost all food and drinking water. Even though more than 200 mg/L of sodium gives unacceptable taste, there is no proposed concentration as health-based guideline.

3.5.2. Heavy metal

Six (6) common heavy metals have been given extra attention in this study because of the threat they bring to soil and groundwater. Such study had been conducted by a number of researchers. Poshtegal and Mirbagheri (2019) studied the presence of As, Mn, Ni, Se, Pb, Al, and Fe while Zarcinas et al. (2004) (Zarcinas et al., 2004) focused on As, Cd, Cr, Cu, Hg, Ni, Pb and Zn for their study. The elements selected for this study are Cu, Zn, Ni, Pb, Cr and Fe. Fig. 5 shows six charts representing each element in every sample.

The purpose of running the tank leaching test was to observe the leaching trend of the blending combination. The observation was plotted in a cumulative graph for each element to monitor how much leachate produced at designated time as well as the total leaching after 64 days being immersed in acidic water. All six heavy metals showed an increasing trend. The running totals increased at every designated time. As the number of day increase, the total concentration of the elements was also increasing.

Fig. 7 showed the presence of Cu, Zn, Ni, Pb, Cr and Fe in cumulative value for each percentage combination of PMA and WMA additive. Fig. 6a shows that the reading for Cu for all samples which have similar pattern except R0 and R1 which have higher reading on day 9 and day 16 respectively. The reading for Fe is shown in Fig. 6b where R2 shows the highest concentration all along compared to other samples. However, the values are consistent with small differences. On the other hand, concentration for Ni as shown in Fig. 6c shows only R4 has higher increment at day 36 while other sample showed a consistent pattern. Meanwhile, readings for Pb concentrations showed uneven pattern for all samples. The plots can be seen clearly in Fig. 6d. The concentration for Cr as shown in Fig. 6e seems to be divided into two patterns. One is where the increment is consistent and another one shows a slightly higher reading at day 36 for R3, R4, and R5. Finally, Fig. 6f shows a uniform curve pattern for all samples.

Reliability test measures the consistency of the element concentration for each sample compared to days of test as well as the repeatability of the test. The correlation between the concentration of element for each sample and the number of days was determined. Table 5 shows the Pearson's correlation coefficient for each element obtained from each sample. All the coefficients are positive in values. The value carries the meaning that for every additional day of being submerged, the concentration of element increased. This relationship is true for all samples. However, the values varied

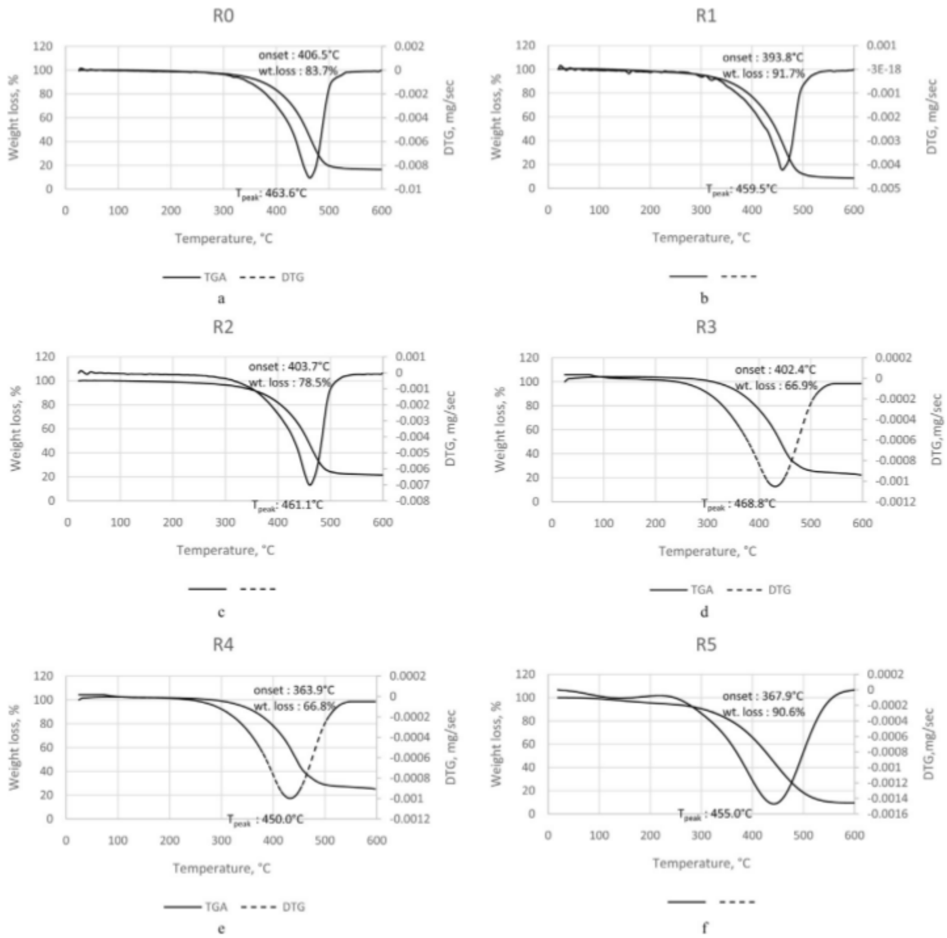

Fig. 5. TGA and DTG thermograms for each sample
*note: wt. loss: weight loss; T_{peak}: peak temperature.

from 0.51 to 0.97. Nonetheless, 89% of values are more than 0.7 indicating that the two variables have strong relationship. To further analyse the reliability of the test, Spearman correlation coefficient was used to examine the strength and direction of between the two variables. It is found that all relationships give $r_s = 1$; meaning by ordinary standards, the relationship between the two variables considered to be statistically significant. By comparing Pearson's correlation coefficient to Spearman correlation coefficient, the two are in such relationship that while one variable increases when the other increases, the amount is not consistent.

3.5.3. Reading at day 64

Fig. 8 comprises all six heavy metal reading at day 64 for all samples. The charts are meant to give overall view of the cumulative concentration of heavy metals of every blending combination. As per see, 4 elements showing a reduction of concentration as the WMA additive quantity increased. Cu and Fe show a consistent pattern while for Ni, R5 gives the same concentration with R4. From Fig. 8, it is found that Zn to be significantly higher than other elements. One of the reasons why is that, Zn is an ordinary element that come about naturally in water, soil, and air. Zn is known to be a soluble metal and it is very pH reliant. The deterioration of the asphalt binder is also accountable for the increasing discharge of zinc (Aalbers et al., 1996). Thus, the source of Zn could be contributed by both substances, the binder and the acidic water. Zn shows a pattern of concentration increased as the content of WMA additive increased. Nonetheless, the relationship is only true up to the loading of 2 wt%. The concentration was then reduced when 3 wt% of WMA additive was added. The concentration continued to drop at 4 wt% and it became constant when the addition of Rediset is 5%. Since Rediset LQ is also soluble, the concentration of Zn might reduce due to the reaction between the elements in the Rediset-asphalt binder and water. However, the reaction only goes beyond consistent when more than 3% of Rediset added into PG76. As for Cu which also a soluble metal, the reduction of concentration as the Rediset content increasing might happened due to the same reason.

3.5.4. Drinking water

The leaching elements was then analysed for drinking water standard. Drinking water is defined as water that consumed by human which does not contain any compounds, alone or combined with other compounds as well as any parasites and microorganisms in such concentration that can be harmful to health (World Health Organization, 2017). Table 6 lists the concentrations of heavy metals detected via ICPMS from the studied samples. Heavy metal

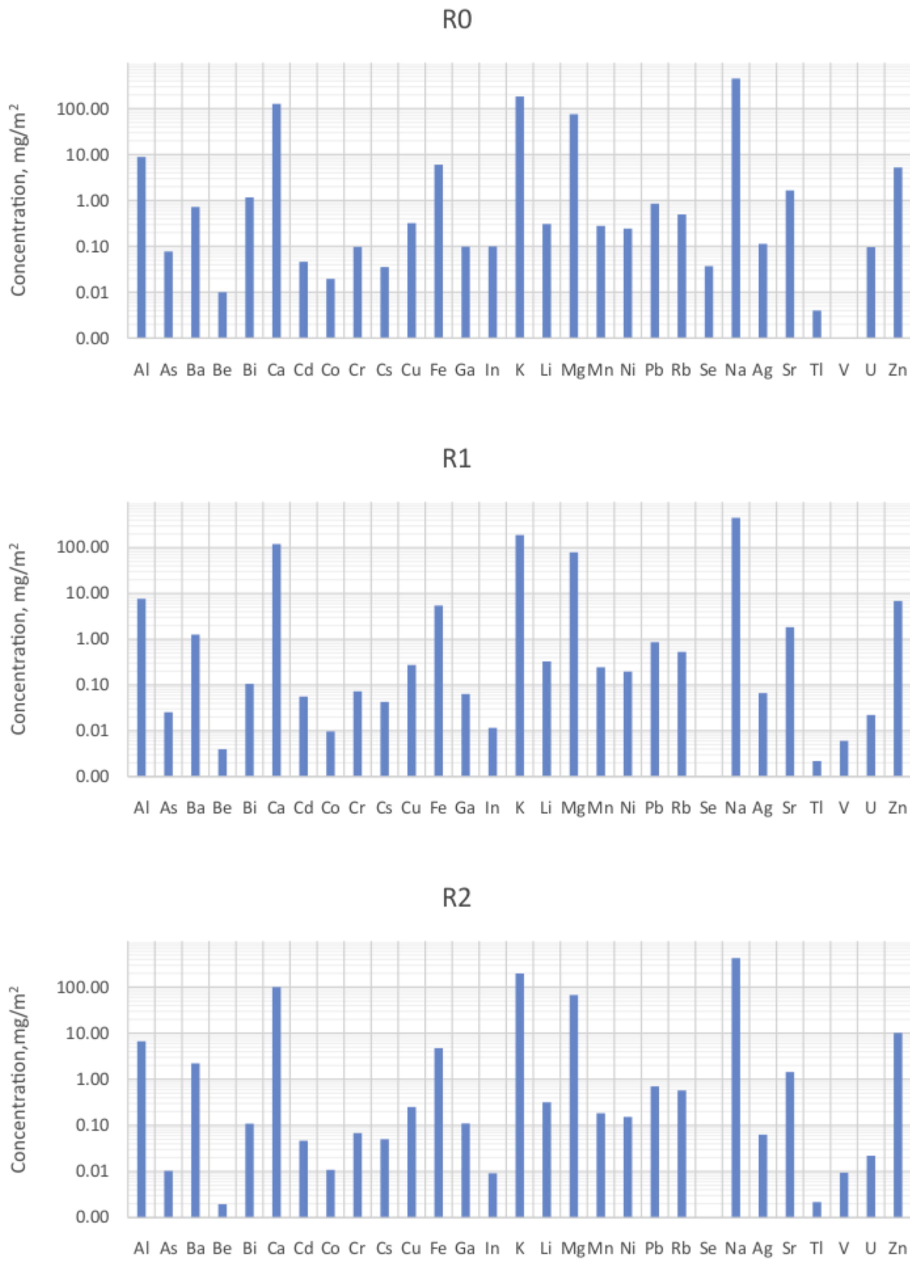

Fig. 6. Cumulative concentrations of each element measured after day 64.

toxicity can be a threat to human. Pb gives various symptoms related to the nervous system and lead poisoning. Headache, irritability, abdominal pain are among the common symptoms caused by Pb (Järup, 2003). As for Fe, even though it is useful to human body, it can be a threat because it can generate hydroxyl radical (OH•) while Ni has extensive range of carcinogenic mechanism. Engwa et al. (2016) also listed that Cu and Cr have been recognised to partake in oxidation and reduction reactions and generate free radicals from H_2O_2 respectively. Generally, all the results are below the minimum standard of WHO and the local authority, Ministry of Health, Malaysia. Amongst all the samples, R0 has the highest reading of Cu concentration; 0.001425 mg/L while R1 found to be the highest with Fe, Ni, and Zn concentration after 64 days with readings of 0.012328 mg/L, 0.000907 mg/L, and 0.02743 mg/L respectively. As per element of Pb, R3 showed the highest reading of 0.004733 mg/L and lastly, Cr found to be the highest in R4;

0.00078 mg/L. Thus, even though the blends are to be submerged under rainwater for 64 days, the water from nearby well is drinkable. However, in actual pavement, there are possibilities of heavy metal elements released from the surface water pressure in the tyre-water-pavement system (Duong and Lee, 2011). However, this study focused on the binder material which plays an important role in pavement. This study initiates the possibilities of more complex source of drinking water contamination due to road construction.

3.5.5. Heavy metal evaluation index (HEI), Heavy Metal Pollution Index (HPI), degree of contamination (C_d), and metal quality index (MQI)

Heavy metal evaluation index (HEI), Heavy metal pollution index (HPI), Degree of contamination (C_d), and Metal quality index (MQI) are practical indices used to assess heavy metal contamination. These indices give a composite influence of selected metals on the overall groundwater quality. The calculated indexes for the PG76-Rediset combinations are summarised in Table 7. The calculated heavy metal evaluation index which gives an overall quality of

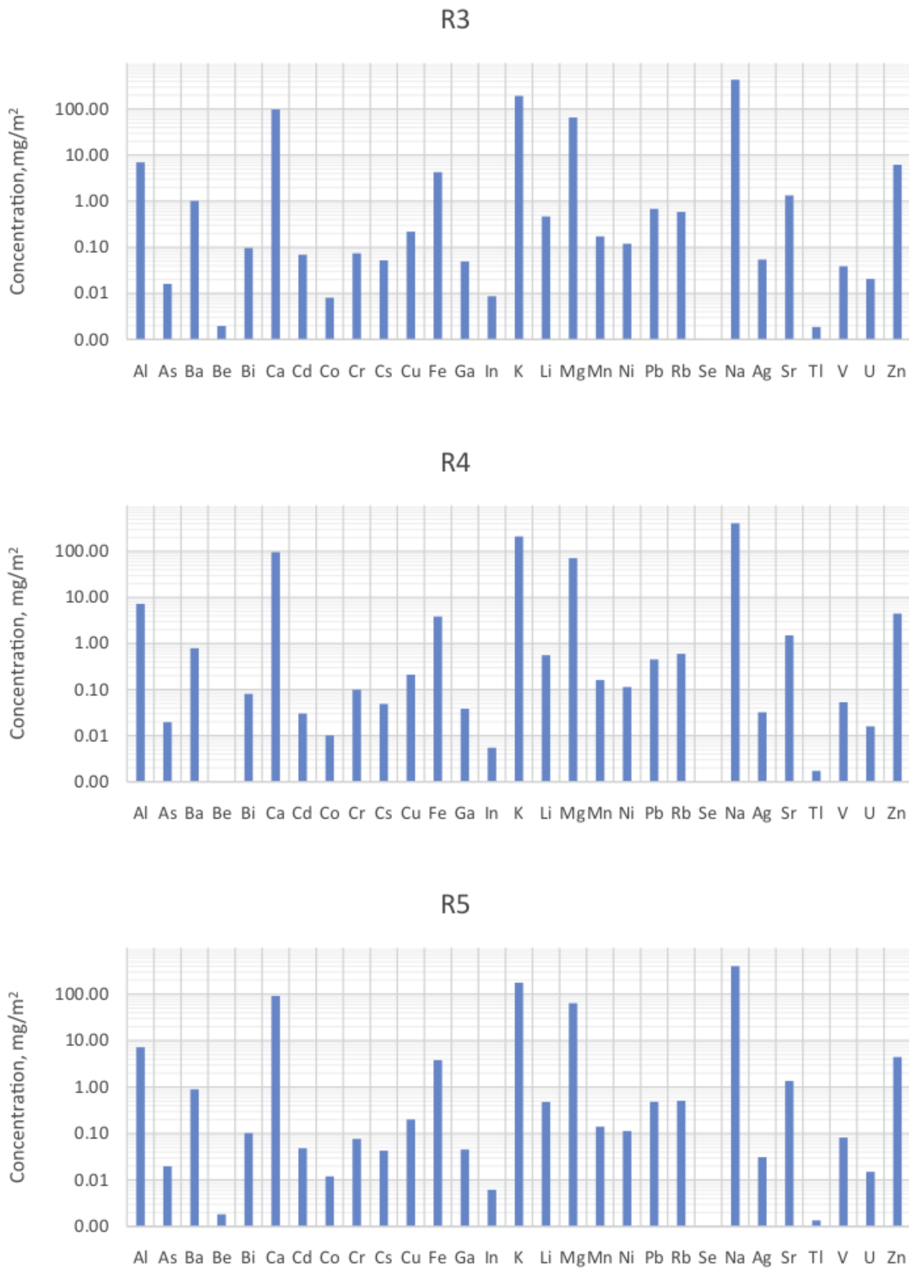

Fig. 6. (*continued*).

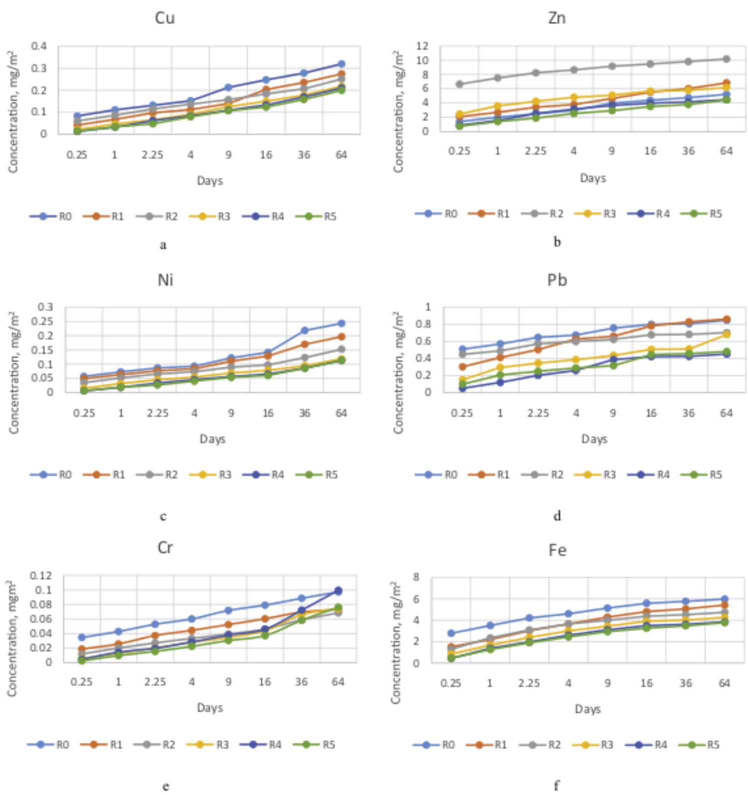

Fig. 7. Six selected elements of (a) Cu, (b) Zn, (c) Ni, (d) Pb, (e) Cr and (f) Fe in cumulative values for every blend combination.

Table 5
Pearson's Correlation Coefficient for each element obtained from each sample.

Sample	Pearson Correlation Coefficient					
	Cr	Cu	Ni	Pb	Zn	Fe
R0	0.83	0.84	0.92	0.57	0.98	0.67
R1	0.84	0.89	0.90	0.71	0.81	0.76
R2	0.92	0.85	0.89	0.53	0.51	0.70
R3	0.93	0.89	0.89	0.80	0.64	0.74
R4	0.97	0.91	0.92	0.73	0.72	0.74
R5	0.95	0.91	0.94	0.77	0.82	0.77

the water with respect to heavy metals. Of all the samples, R2 owns the minimum value that is 0.262, while R3 gives the maximum value of 1.090. However, the values obtained for this index are below 5 which indicates that for every percentage weight of Rediset added into PG 76 is in low degree of pollution.

Meanwhile, the HPI which based on the weighted arithmetic quality mean method represent the total quality of water with respect to heavy metals. For this index, any number less than 90 is considering as low degree of pollution. In this study, the ranges of value calculated are between 71.584 and 74.118. With mean value of 73.479, the blending combinations are all in low degree of pollution.

The degree of contamination (C_d) is a calculation method to evaluate the contamination index of water quality. Each sample is computed separately and all samples show value less than 1.0. This means that even after 64 days being submerged in acidic water, the combination of blendings in this study are in low degree of pollution.

Finally, for this study, the concentration of a metal was compared to its respective maximum permissible limit to get the metal quality index (MQI). For the water to be able for consumption, MQI value should be > 1 (Tamasi and Cini, 2004). For this study, all samples showed the value < 1. Thus, the blending was safe to be used near water source.

4. Conclusion

From the physical tests data, i.e. penetration, softening point and ductility tests, the addition of Rediset maintained the physical properties of PG76. The quality specification for all Rediset-modified PG76 samples did not show any significant decrease in their physical performances, thus indicating they are suitable for practical application. Thermal analysis via TGA showed that the new materials decomposed at temperature more than 300 °C. Even though WMA additive had influence in lowering the compacting and mixing temperatures, the materials were not susceptible to high temperature. Thus, they are suitable for the application in tropical countries as well. Wetting and spreading behaviours of the blending were also perceived by contact angle method. An increasing amount of WMA additive content allowed the spreading of water on the surface. The surface was deemed to be hydrophilic or poorly hydrophobic. However, further investigation on leaching test showed that these behaviours did not affect the chemical reaction during the test. As for the environmental effect from the blending, the results showed that the present of all leached heavy metals were complied with WHO and local MOH drinking water

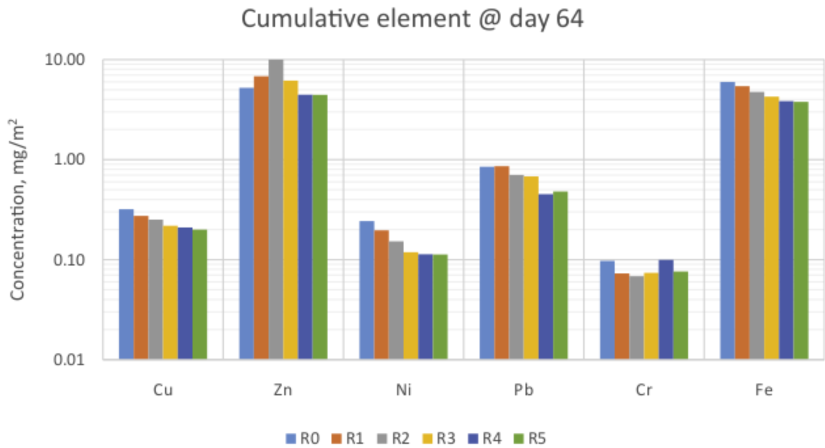

Fig. 8. Cumulative readings of Cu, Zn, Ni, Pb, Cr, and Fe at day 64.

Table 6
Concentrations of heavy metals detected in comparison to WHO and MOH Standard.

Sample	Analyte	Concentration			Standards		
		mg/L	Conc. SD	Conc. RSD	World Health Organization, 2017 Drinking Water	MOH Raw Water	MOH Drinking Water
R0	Cr	0.000297	0.11	38.60	0.05	0.05	0.05
	Cu	0.001425	0.19	13.30	2.00	1.00	1.00
	Fe	0.007316	1.10	15.00	0.30	1.00	0.30
	Ni	0.000851	0.02	1.90	0.07	–	0.02
	Pb	0.001421	0.10	7.20	0.01	0.05	0.01
	Zn	0.015875	2.21	13.90	3.00	3.00	3.00
R1	Cr	0.000115	0.01	6.20	0.05	0.05	0.05
	Cu	0.001334	0.08	5.70	2.00	1.00	1.00
	Fe	0.012328	0.24	2.00	0.30	1.00	0.30
	Ni	0.000907	0.08	9.00	0.07	–	0.02
	Pb	0.001057	0.04	3.50	0.01	0.05	0.01
	Zn	0.02743	0.06	0.20	3.00	3.00	3.00
R2	Cr	0.000261	0.07	27.20	0.05	0.05	0.05
	Cu	0.001207	0.30	24.70	2.00	1.00	1.00
	Fe	0.005976	1.04	17.40	0.30	1.00	0.30
	Ni	0.000791	0.10	13.20	0.07	–	0.02
	Pb	0.000563	0.02	2.90	0.01	0.05	0.01
	Zn	0.009853	0.33	3.40	3.00	3.00	3.00
R3	Cr	0.000228	0.03	15.20	0.05	0.05	0.05
	Cu	0.001134	0.16	14.00	2.00	1.00	1.00
	Fe	0.007042	1.04	14.80	0.30	1.00	0.30
	Ni	0.000689	0.05	7.60	0.07	–	0.02
	Pb	0.004733	0.11	2.20	0.01	0.05	0.01
	Zn	0.011006	0.06	0.60	3.00	3.00	3.00
R4	Cr	0.00078	0.03	4.00	0.05	0.05	0.05
	Cu	0.001154	0.04	3.70	2.00	1.00	1.00
	Fe	0.006452	0.49	7.60	0.30	1.00	0.30
	Ni	0.000765	0.10	13.10	0.07	–	0.02
	Pb	0.000743	0.02	2.50	0.01	0.05	0.01
	Zn	0.008674	0.04	0.40	3.00	3.00	3.00
R5	Cr	0.000566	0.05	9.60	0.05	0.05	0.05
	Cu	0.001297	0.05	4.10	2.00	1.00	1.00
	Fe	0.009202	1.16	12.60	0.30	1.00	0.30
	Ni	0.000831	0.07	7.90	0.07	–	0.02
	Pb	0.00075	0.03	4.40	0.01	0.05	0.01
	Zn	0.020293	0.67	3.30	3.00	3.00	3.00

* Note: Conc. SD: Standard Deviation of concentration.
Conc. RSD: Relative Standard Deviation of concentration.

standards. The blending has no threat to the drinking water even though at worst case scenario of flood.

CRediT authorship contribution statement

Suzielah Rahmad: Writing - original draft, Writing - review & editing. **Nur Izzi Md Yusoff:** Supervision, Funding acquisition. **Syazwani Mohd Fadzil:** Investigation. **Khairiah Haji Badri:**

Table 7
Summary of Heavy Metal Evaluation index (HEI), Heavy Metal Pollution Index (HPI), Degree of Contamination (C_d), and Metal Quality Index (MQI).

Sample ID	HEI	HPI	Cd	MQI
R0	0.458	73.517	−5.971	0.029
R1	0.421	73.690	−5.972	0.028
R2	0.262	74.118	−5.982	0.018
R3	1.090	71.584	−5.941	0.059
R4	0.309	74.016	−5.979	0.021
R5	0.339	73.950	−5.977	0.023
Minimum	**0.262**	**71.584**	**−5.982**	**0.018**
Maximum	**1.090**	**74.118**	**−5.941**	**0.059**
Mean	**0.480**	**73.479**	**−5.970**	**0.030**

*All values in mg/m^2.

Writing - review & editing, Supervision. **Ahmad Kamil Arshad:** Formal analysis. **Iswandaru Widyatmoko:** Writing - review & editing, Supervision. **Sri Atmaja P. Rosyidi:** Formal analysis.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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