

Article

Evaluation of the Engineering Properties of Powdered Activated Carbon Amendments in Porous Asphalt Pavement

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Abstract: Porous asphalt pavement (PAP) with a high drainage capacity was modified with powdered activated carbon (PAC) addition to produce permeable reactive pavement (PRP), which may exhibit the potential to reduce environmental non-point source (NPS) pollution. The experimental design mixtures used to produce and test the PRP incorporated with PAC (named PRP-PACs) were conducted as follows: first, the PACs were initially tested to determine their feasibility as an additive in PAP; second, different amounts of PAC were added during the preparation of PAP to produce PRP-PAC, and the unregulated and regulated physical characteristics for the mechanical performance of PRP-PACs were examined to ensure that they meet the regulatory specifications. Third, the aqueous contaminants, namely benzene, toluene, ethyl-benzene, and xylene (BTEX), column adsorption tests were preliminarily conducted to demonstrate their adsorption capacities compared to traditional PAP. The compositions of 0.8% and 1.5% PAC (by wt.) (PRP-PAC08 and PRP-PAC15) met all the regulated specifications. As compared to PAP, PRP-PAC08 exhibited higher BTEX adsorption capacities than PAP, which were 47%, 49%, 29% and 2%. PRP-PAC08 showed both superior physical properties and adsorption performance than PAP and may be recommended as an engineering application that reduces the potential for NPS contamination of air, soil, groundwater, and surface water.

Keywords: porous asphalt pavement (PAP); volatile organic compounds (VOCs); asphalt concrete; remediation; surface runoff



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1. Introduction

Non-point source (NPS) pollution from traffic or industry, such as volatile organic compounds (e.g., benzene, toluene, ethyl-benzene, and xylenes (BTEX)), are frequently detected in ambient air and rain (the tabulated literature-reported data pertaining to BTEX concentrations in ambient air and rain are shown in Tables S1 and S2, Supplementary Materials (SM)). For example, total BTEX concentrations were reported to be 106 $\mu\text{g}/\text{m}^3$ in ambient air (Ren-Wu, Taiwan) and 0.573 $\mu\text{g}/\text{L}$ in rain (Yokohama, Japan). A fugacity-based model developed by Iordache et al. [1] also indicated the potential of BTEX transfer from point sources such as wastewater discharges into the environment. Their model highlighted the trend of volatile organic compound transfer in the air, which may require a control of these emissions. Planting (park development, planting trees, etc.) is now the main method used to reduce NPS pollutants, but the effect is limited [2]. Since pollution originating from vehicle exhaust is one of the major NPS sources, if pollutant emissions produced while cars moving on the road could be retained by the asphalt pavement, migration of these hazardous materials could thereby be controlled. The sorbed pollutants on the pavement could be destroyed by natural photo-degradation [3] or when a severe pavement deterioration is reached, the asphalt could be excavated and sent back to the asphalt plant for recycling. The Taiwan Ministry of Transportation and Communications (MOTC) reported that the percentage of increase in the numbers of motor vehicles, the total length of asphalt roads, and the total area of asphalt pavement were 38%, 21% and 293%, respectively, between the years 2000 and 2017 [4]. Given the growing numbers of

vehicles, and increasing area of pavement, it may be beneficial to capture the pollution from vehicle exhaust on pavement, and thereby reduce the potential for these contaminants to be transported to air, soil, surface water, and groundwater. Successful control of vehicle exhaust chemicals on pavement will reduce human exposure to these compounds.

Porous asphalt pavement (PAP) has several important technical features, including good anti-slipping performance [5], low noise [6], reduced water splash and spray on rainy days [7], and mitigation of glaring lights when driving at night. These benefits are partly due to large air voids (~20%) [8] and the permeable structure of the interior of the PAP [9]. PAP with higher voids would allow faster drainage and reduce dangerous driving hazards, such as hydroplaning caused by puddles. This study is a continuation of our research on developing permeable reactive pavement (PRP), which was initially prepared using granular activated carbons (GACs) as an additive and incorporated into the design of PAP by Huang and Liang [10]. In this study, powdered activated carbons (PACs) were applied, instead of GACs, to prepare PRP (named PRP-PAC) and the objectives of this study included (1) evaluation and characterization of PRP-PAC; and (2) comparison of the BTEX adsorption capacities of PAP and PRP-PAC.

2. Material and Methods

2.1. Materials

Powdered activated carbons (<200 mesh) were purchased from China Activated Carbon Industries, Ltd., Taipei, Taiwan. Benzene (99.7% min.) and ethyl-benzene (99.0% min.) were purchased from Sigma-Aldrich (St. Louis, MO, USA). Toluene (99.0% min.) was purchased from Union Chemical Works, Ltd. (Hsinchu, Taiwan) and m-xylene (99%) was purchased from Alfa Aesar. Hydrochloric acid (37% min.) was obtained from Sigma-Aldrich. Pyrolith, granite, and metamorphic rocks were obtained from Hsin Hsu Tai Engineering Co., Ltd., Taipei, Taiwan. The cellulose fiber and modified asphalt were obtained from Chien Chung Engineering Co., Ltd., Taiwan.

2.2. Methodology

2.2.1. Design and Preparation of PAP and PRP-PAC

The PAP and PRP-PAC preparation procedures were on the basis of the road construction design criteria of porous asphalt pavement and the detailed design activities were described in Huang and Liang [10]. A 21% voids design criterion was used for the porous pavement in this study. PAC was washed with hydrochloric acid and then rinsed with reverse osmosis (RO) water [11]. Thereafter, PAC was oven-dried at 105 °C (6 h) and then stored in a desiccator. Different amounts of PAC (0.4%, 0.8%, and 1.5% by wt.) were supplemented during the course of the porous asphalt pavement (PAP) preparation. These three PRP-PAC compositions met the upper and lower specified gradation limits. The mixed aggregates were blended at a temperature of 180 °C for 6 s, and then blended with hot bitumen at 160–170 °C for 34 s, and thereafter, the designated quantities of the PAC were gradually sprayed onto the aggregates by hands and blended for 6 s prior to compaction into cylindrical PRP-PAC samples.

2.2.2. Aqueous BTEX Adsorption Capacity Experiments

Column adsorption tests were conducted to measure the adsorption capacity of PRP-PAC and to compare it with PAP. The PAP and PRP-PAC cylindrical samples (radius = 10 cm, height = 6 cm) were placed in a stainless-steel holder (see Figure 1 for experimental setup). It should be noted that, in these adsorption experiments, elevated BTEX concentrations, which are higher than those usually detected in rain or runoff water (the literature survey pertaining to BTEX concentrations in ambient air and rain is presented in Tables S1 and S2), were used in this study to ensure that each individual adsorption experiment could be completed within an acceptable time frame (~60 h). The BTEX solution (60 mg L⁻¹ in RO water (pH~6)) was prepared by adding pure BTEX, stirred for 48 h in a 2 L glass bottle without head space, and transferred into a Tedlar[®] bag (10 L). The prepared BTEX solution

was pumped (10 mL min^{-1}) upwards into the column using a peristaltic pump equipped with a PTFE head and tubing (Cole Parmer MASTERFLEX[®] model 77390-00). The BTEX concentrations in the influent and effluent were monitored at different time intervals until breakthrough of the BTEX was reached.

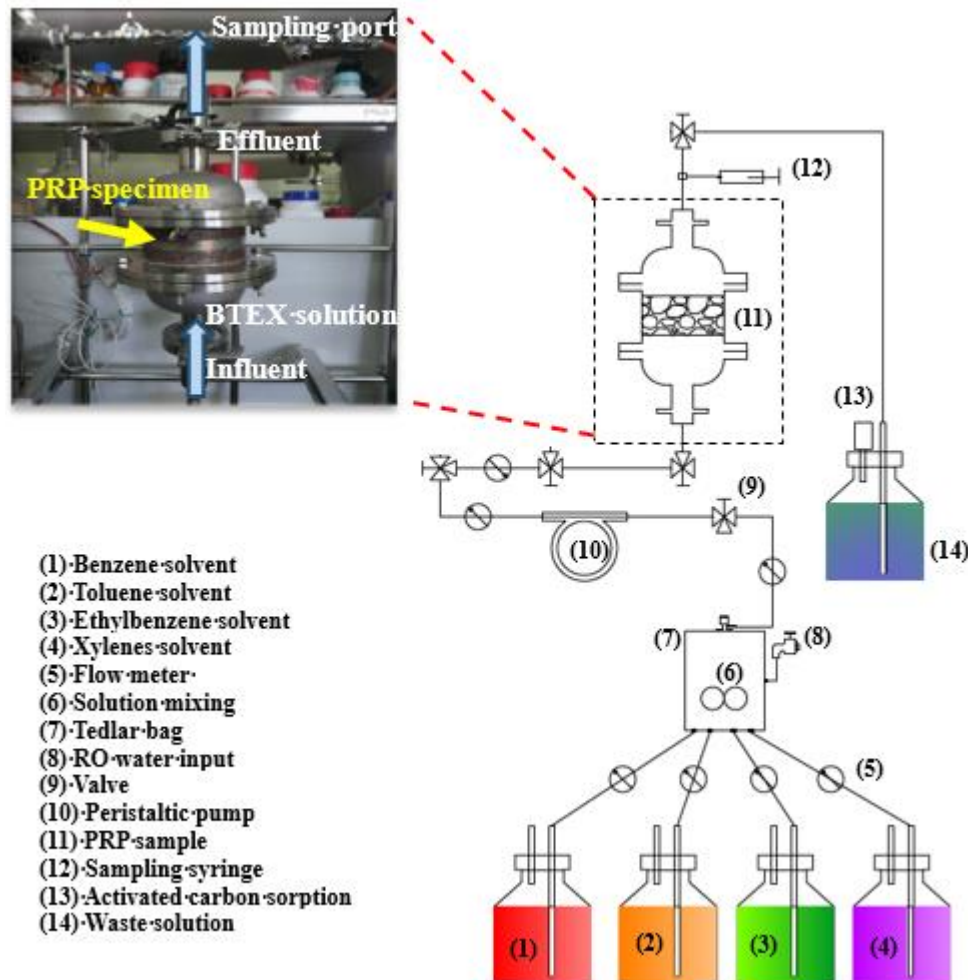


Figure 1. Experimental setup for porous asphalt pavement (PAP) and permeable reactive pavement-powdered activated carbon (PRP-PAC) adsorption.

2.3. Analysis

The mass loss of PACs was measured using a thermogravimetric analyzer (TGA) (LINSEIS, STA PT1600 model) over the temperature range of 25–1000 °C at a heating rate of 5 °C min^{-1} . The BET specific surface, pore volumes, and pore sizes of the PACs were determined by N_2 adsorption isotherms (77 K) using a BET sorptometer (Porous Materials, Inc., CBET-201A). Aqueous BTEX concentrations were analyzed using a high-performance liquid chromatograph/UV detector at a wavelength of 254 nm (HPLC, Agilent 1100) on a ZORBAX Eclipse XDB-C18 column. The mobile phase consisted of acetonitrile/water (70:30, *v/v*) and the flow rate was 1 mL min^{-1} . Characterization of aggregates and evaluation of the mechanical properties of PAP and PRP-PAC were in accordance with the relevant specified analytical methods, which are provided in Table S3 (SM).

3. Results and Discussion

3.1. Characterization of PACs

The results of the TGA analysis are presented in Figure 2, which show that little mass loss was detected during heating at 30–100 °C, presumably due to loss of water, and

nearly no change of mass was observed for the PAC in the range of 100–185 °C. However, when the temperature increased above 185 °C, significant mass losses were observed. PAC with sufficient thermogravimetric stability can be mixed with aggregates and asphalt in the mixer at a high temperature condition (170–185 °C) [12]. Additionally, PACs with a large surface area ($865 \text{ m}^2 \text{ g}^{-1}$) and pore volume ($0.51 \text{ cm}^3 \text{ g}^{-1}$) (see the inserted Table in Figure 2) appear to be good candidates for adsorbing contaminants.

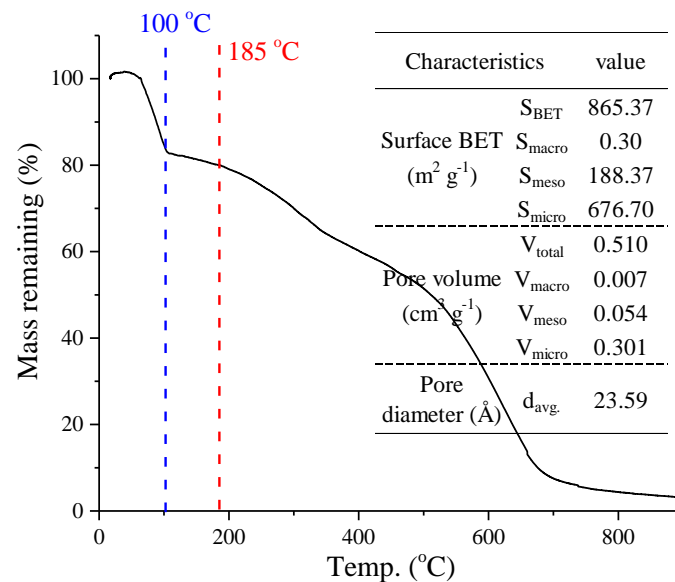


Figure 2. TGA and BET analysis of PAC.

3.2. Permeable Reactive Pavement

3.2.1. PAP and PRP-PAC Preparation

The properties of the aggregates (fine and coarse) and asphalt are presented in Tables S4–S6 (SM) and the physical property measurements were shown to meet the required specifications. The PAP void space design was 21% (note that 20% is usually set for porous pavement [13]) and the additional 1% void space was designed higher than traditional PAP in order to allow space for PAC addition without significant reduction of voids, which provide permeability. The 21% void design of PAP-PAC must be made up of 11% of the aggregates passing through a No. 8 sieve, which was determined by the linear relationship of percent void versus various compositions of the aggregate passing through a No. 8 sieve (see Figure S1 (SM)). The designed upper and lower gradation limitations for the 21% void in PAP are presented in Table S7 (SM). Abrasion and drain-down tests were conducted for samples with various blends of asphalt, which indicated an optimum asphalt content of 5.95% (by wt.) (shown in Figure S2 (SM)). The gradation curves for percent passing versus sieve size for different mixtures of PAC (i.e., 0.4, 0.8, and 1.5% by wt., noted as PRP-PAC04, PRP-PAC08, and PRP-PAC15, respectively) are presented in Table S7 (SM), which demonstrated that these PRP-PACs mixtures are within the upper and lower specification limits.

3.2.2. PAP and PRP-PAC Performance

The results of the physical performance testing for PAP and PRP-PAC are presented in Figures 3 and 4 for the regulated and non-regulated physical properties, respectively. Void, abrasion, and drain-down tests are three major parameters for constructing porous pavement. Void space criteria are applied to ensure that the PRP-PAC has enough space between the aggregate components for water drainage. Both abrasion and drain-down tests are there to confirm that the PRP-PAC has an appropriate amount of asphalt for adhesion and binding of aggregates. PAP and PRP-PACs met the specifications of MOTC with the

exception of PRP-PAC04, which showed 0.32% drain-down, which exceeded the maximum limit of 0.3%. Hence, 0.4% (by wt.) PAC would not be a suitable PRP composition.

Marshall stability (MS) and Marshall flow (MF), respectively, are the indications of pavement resistance to deformation, and are measures of the extent of deformation that can occur under traffic loadings. The added PAC could adsorb asphalt and reduce the tendency of asphalt to flow during warm temperature conditions [14]. It can be seen that the higher percentage of PAC added during our test resulted in higher MS and lower MF values. Both PRP-PAC08 and PRP-PAC15 reveal both higher strength and flexibility with superior MS (705–1031 kgf) and MF (2.91–3.09) performance as compared to those of PAP.

Retained Strength (RS) analysis is utilized to evaluate the durability of PAP and PRP-PACs due to aging of the asphalt, disintegration of the aggregate, and stripping of the asphalt film from the aggregate due to weather or traffic. The RS test results of both PRP-PAC08 (87.8%) and PRP-PAC15 (91.7%) show better anti-aging performances under long-term weathering than PAP (81.9%). Moreover, as compared to PAP, both PRP-PAC08 and PRP-PAC15 reveal a higher dynamic stability (DS) (4474–5581 passes/mm) and lower deformation rate (DR) (0.0075–0.0088 mm/min), which indicate PRP-PACs could better sustain the rolling loading of vehicles on the road and exhibit superior cumulative resistance to rutting deformation during heavy traffic loading periods. The addition of PAC to asphalt (e.g., asphalt sorption onto PAC) may result in a decreased thickness of the asphalt coating on the aggregates of PRP-PACs, and the increase of void space (e.g., 22.59% for PRP-PAC15 compared to 19.75% for PAP) could lead to an increase in permeability (e.g., 0.28 cm/sec for PRP-PAC15 compared to 0.14 cm/sec for PAP) of the PRP-PACs (see Figure 3).

The indirect tensile strength (ITS) analysis is used to determine the tensile properties of the PAP and PRP-PACs, which can further be related to the cracking properties of the pavement. A higher tensile strength corresponds to a stronger cracking resistance. Asphalt binder sorbed by PAC results in a higher void and seems to cause a slightly lower ITS value. The results obtained (6.4–8.3 kgf cm⁻² for PRP-PACs) are comparable to those reported for porous asphalt prepared using recycled crumb rubber (5.8–6.8 kgf cm⁻²) [15]. PRP-PACs, as compared to PAP, are capable of tolerating a higher strain prior to failure. In order to imitate the weather situation (a subtropical climate) in Taiwan, the unregulated properties of the wet dynamic stability (DS_w) and wet deformation rate (DR_w) were also examined under water-rinsing conditions. Both PRP-PAC08 (D_{sw} 2355 passes/mm) and PRP-PAC15 (D_{sw} 2869 passes/mm) revealed better wet dynamic loading resistance (see Figure 4) than PAP (D_{sw} 1181 passes/mm), based on the higher DS_w and lower DR_w values. Therefore, PRP-PACs potentially exhibit good mechanical performance under the weather conditions in Taiwan. Regarding the connected and closed porosities of PRP-PACs, the addition of PAC reduces the asphalt film covering on aggregates and induces slight increases in connected channels within the PRP-PAC. However, there are insignificant variations in closed porosity.

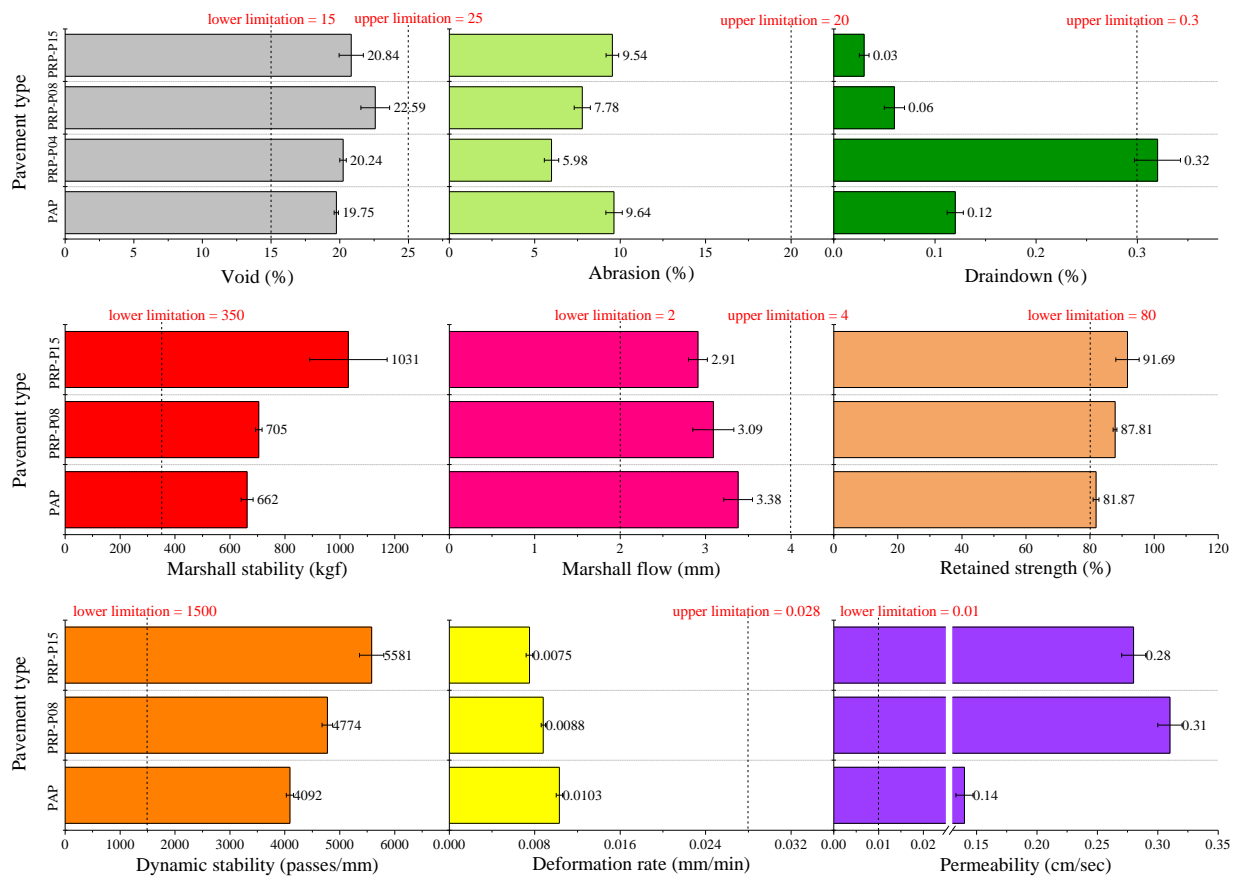


Figure 3. Comparison of the regulated properties (voids; abrasion; drain-down; Marshall stability; Marshall flow; retained strength; dynamic stability; deformation rate; and permeability) of the PAP and PRP-PACs (data expressed as the mean \pm one standard deviation, $n = 3$).

Voids in the mineral aggregates (VMA) include the air voids and asphalt film, which are the intergranular void spaces that exist between the aggregate particles in a compacted paving mixture. The more VMA in the PRP-PAC sample, the greater the space available for the asphalt film and the durability of the PRP-PAC mixture increases with the film thickness on the aggregate particles. This can be seen in the VMA of PRP-PAC08 and PRP-PAC15 (33.0% and 33.8%), which are higher than those obtained in PAP (31.01%). These results are similar to the values of 25–29% reported for porous asphalt mixtures incorporating crumb rubber [15], 31% for PAP with fiber addition, and 31.2–32.4% for PAP with granular activated carbon addition [10], and support the engineering quality of PRP-PACs.

Voids filled with asphalt (VFA) is the percentage of the inter-granular void spaces of VMA that are filled with asphalt. VFA is a measure of the effective asphalt binding among aggregates. It can be seen that the addition of PAC resulted in the VFA measurements for a PRP-PAC08 and PRP-PAC15 of 33.2 and 36.9%, respectively, which are similar to that of PAP (36.3%), implying that PAP with PAC addition did not cause significant variations in VFA, and therefore did not significantly alter the durability of the PRP-PACs.

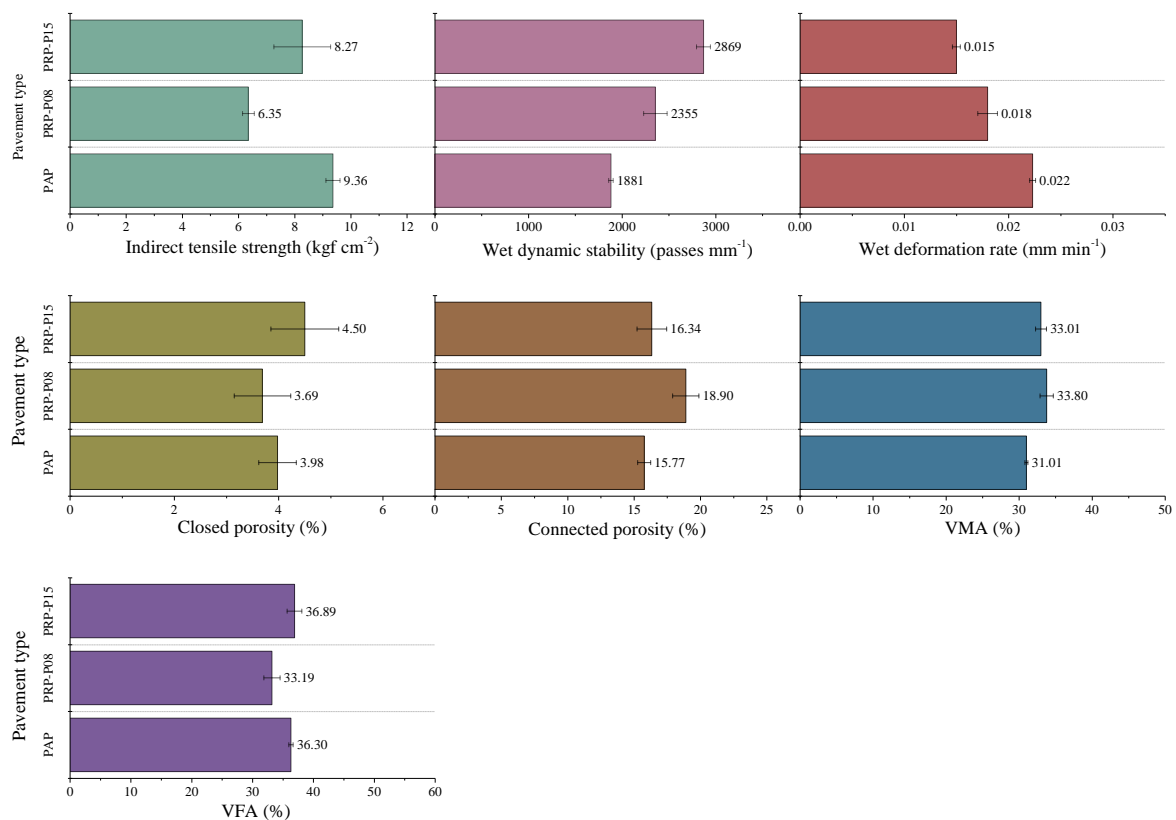


Figure 4. Comparison of the unregulated properties (indirect tensile strength; wet dynamic stability; wet deformation rate; closed porosity; connected porosity; voids in the mineral aggregates and voids filled with asphalt) of PAP and PRP-PACs (data expressed as the mean \pm one standard deviation, $n = 3$).

3.2.3. Comparison of Adsorption Capacity of PRP-PAC vs. PAP

Based on the results of physical performance evaluation, PRP-PAC04 did not meet the drain-down loss regulation limit, and hence was not further examined. BTEX column adsorption onto PAP, PRP-PAC08, and PRP-PAC15 were conducted to evaluate their adsorption capacities. The adsorption amounts of each sample were obtained from the breakthrough curves (see Figures S3–S5 (SM)) and comparison of adsorption variations using PAP as a basis are shown in Figure 5. It can be seen that the adsorption amounts are highly related to the solubility of BTEX in water (i.e., compounds with higher water solubilities (e.g., benzene) exhibited the greater adsorption capacity). Both PRP-PAC08 and PRP-PAC15 can effectively adsorb benzene, toluene, ethylbenzene, and xylene. The void and connected porosities of PRP-PAC08 are higher than those of PAP and PRP-PAC15, and it is possible that the pore space may be blocked by thick asphalt film and excess PAC, which could reduce the available adsorption sites. The increases in adsorption capacities of BTEX onto PRP-PAC08 were greater than those of PAP and PRP-PAC15, by 47, 49, 29, and 2%, respectively. In conclusion, as compared to PAP and PRP-PAC15, PRP-PAC08 showed both superior physical performance and adsorption capacities and revealed the potential for a successful engineering application for reducing aqueous organic contaminants from NPS pollution. These preliminary adsorption results may serve as references for a potential application of PRP-PAC. However, further evaluation with respect to the contaminant adsorption kinetics within the PRP-PAC, attenuation of contaminants under natural environmental conditions, long-term impact and performance, and eventually field demonstration should be considered.

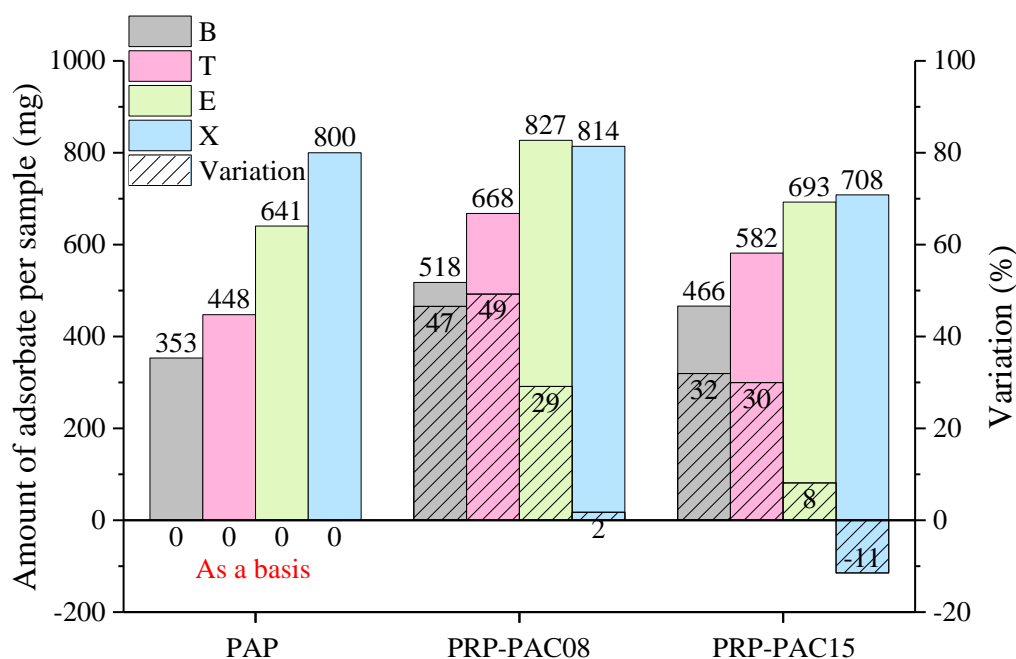


Figure 5. The mass of BTEX sorbed onto PAP and PRP-PACs and comparison of sorption variations using PAP as a basis.

4. Conclusions

Asphalt pavements were modified with additions of PAC to evaluate the ability to sorb BTEX from vehicle exhaust, and thereby reduce aqueous NPS pollution of air, soil, groundwater, and surface water. According to the BET and TGA analytical results, PAC with a high surface area and thermal stability can be applied to adsorb organic compounds by mixing with porous asphalt concrete in a high temperature mixer. Based on the measurements of voids, abrasion, and drain-down test, the voids of the PRP-PAC samples (0.8% and 1.5% (by wt.) of PAC additions) were approximately 20%, which met the void requirement for PAP, while PRP-PAC04 (i.e., with 0.4% PAC addition) exceeded the specification limit for abrasion loss, and therefore is not recommended. Based on physical performance evaluations, PRP-PAC08 and PRP-PAC15 met the regulated physical performance specifications and exhibited a higher loading strength, lower strain, and higher permeability, as compared to traditional PAP. Moreover, the adsorption capacities of PRP-PAC08 increased by 47%, 49%, 29%, and 2% for BTEX, respectively, as compared to PAP. These observations of both the physical and adsorption performance of PRP-PAC08 demonstrated a superior potential for engineering application as a replacement of PAP for reducing NPS contamination of air, soil, groundwater, and surface water.

Supplementary Materials: The following are available online at <https://www.mdpi.com/article/10.3390/pr9040582/s1>, Table S1: Summary of the BTEX concentrations in ambient air, Table S2: Summary of the BTEX concentrations in rainwater, Table S3: The test methods and equations for PAP physical performance, Table S4: The properties of coarse aggregates, Table S5: The properties of fine aggregates, Table S6: The properties of modified asphalt, Table S7: Grading specification and the gradation for PAP and PRP-PACs, Figure S1: Various compositions of aggregates passing through sieve No. 8 used for determining voids of PAP and PRP-PACs, Figure S2: Determination of optimum asphalt content dependent upon abrasion and drain-down tests, Figure S3: The breakthrough curves of BTEX sorption onto PAP, Figure S4: The breakthrough curves of BTEX sorption onto PRP-PAC08, Figure S5: The breakthrough curves of BTEX sorption onto PRP-PAC15.

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Conflicts of 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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