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Decreasing polycyclic aromatic hydrocarbons emission from bitumen using alternative bitumen production process

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ABSTRACT

In 1988, the National Institute for Occupational Safety and Health (NIOSH) recommended that bitumen fumes should also be considered a potential occupational carcinogen and management practices such as engineering controls should be implemented. Changing the production process of bitumen, as a source control method, was investigated in our study. For the first time, a novel alternative process was used to produce paving grade bitumen with decreased PAH emissions as well as improved bitumen performance grade (PG). Post-consumer latex and natural bitumen (NB) were used as additives to obtain 60/70 modified bitumen directly from the vacuum bottom (VB) without any need for air-blowing. The emissions were produced by a laboratory fume generation rig and were sampled and analyzed by GC-Mass and GC-FID as described in NIOSH method 5515. The PG of the resulting modified 60/70 bitumen in this study covers a wider range of climatic conditions and has higher total resistance against deformation than conventional 60/70 bitumen. The total PAH emissions from modified 60/70 bitumen (100.2619 ng/g) were decreased approximately to 50% of PAHs emitted from conventional 60/70 bitumen (197.696 ng/g). Therefore, it is possible to obtain modified bitumen with lower PAH emissions and better quality than conventional bitumen via additives and without air-blowing.

1. Introduction

Bitumen is a dark brown to black petroleum-derived binder that is used for paving (87%), roofing (11%), water proofing, bitumen-based paints, among other applications (2%) [1]. Bitumen composition is divided into four generic groups (SARAs): saturates, aromatics, resins, and asphaltenes [2,3]. The bitumen composition and consequently the physical and mechanical properties of bitumen vary depending on crude oil sources and their refining processes [2]. Due to high temperature processing of bitumen in refineries (up to 288 °C) and bitumen-contained materials (such as roofing products and asphalt) in factories (160–250 °C), the bitumen fumes and vapors may be released, and as a result, workers involved in these processes are exposed to emissions. Bitumen fumes and vapors may contain very small quantities of polycyclic aromatic compounds (PACs), which are comprised of polycyclic aromatic hydrocarbons (PAHs).

Some PACs and derivatives are known to have mutagenic and carcinogenic effects, and certain 4- and 5-ring PACs (i.e., benz[a]anthracene, chrysene, and benz[a]pyrene) are mutagenic and possibly carcinogenic [4]. The carcinogenicity of bitumen fumes has been investigated in several epidemiological studies, and some of these studies have suggested that exposure to bitumen fumes increases the risk of developing lung cancer as well as other cancers [5]. The International Agency for Research on Cancer (IARC) have evaluated the carcinogenicity of bitumen and classified air-refined bitumen as a possible human carcinogen (IARC group 2B) [6,7]. In 1988, NIOSH (National Institute of Occupational Safety and Health) advised asphalt fumes should also be considered a potential occupational carcinogen and governing practices such as engineering controls should be employed [8].

Several laboratory and field studies have investigated the quality and quantity of bitumen emissions and their occupational and environmental impacts [9]. Also, some descriptive studies have investigated PAH emissions from asphalts produced with conventional and modified paving grade bitumen. NIOSH investigated the occupational exposures and acute health effects of crumb-rubber modified (CRM) asphalt in comparison with conventional asphalt [10]. This study reported increased concentrations of emissions in personnel breathing zones for CRM asphalt. Virpi Vaananen

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Table 1
Composition of VB and 60/70 bitumen from Tehran Refinery (crude from Asmari reservoir of Ahwaz in Khuzestan, Iran).

Material	Saturates (wt%)	Naphthene aromatics (wt%)	Polar aromatics (wt%)	Asphaltenes (wt%)
VB	12.61	43.31	40.09	3.99
60/70 bitumen	15.83	39.58	35.43	10.26

et al. studied the road pavers' occupational exposure to asphalt containing waste plastic and tall oil pitch and reported similar concentrations of PAHs in conventional and modified asphalt exposures [11]. Saarela et al. studied the composition of bitumen emissions in laboratory and field conditions when organic and inorganic, recycled industrial by-products were used in asphalt mixes. The results of the exposure measurements show that the use of inorganic recycled material, coal fly ash, did not affect the concentrations of the exposure indices [12]. Hugener et al. measured the laboratory emissions from tar-containing bitumen and reported increased PAH emissions from tar-containing bitumen in comparison with conventional bitumen [13]. Brandt and Groot investigated the factors affecting worker exposure to PAHs in bitumen fumes and reported the similarity of PAH profiles between bitumen fumes emitted during road paving generated in laboratory [14]. Kurek et al. studied the potential of carcinogenicity of SHRP asphalt fumes and NIOSH fume fraction and found that the earlier has lower carcinogenic potential [15]. The most of these studies have reported high levels of emissions that exceed occupational exposures and environmental releases and have recommended the importance and necessity of control measures [5]. Thus, changing the production process as a source control method and as an effective measure in order to eliminate or minimize the PAH emissions of bitumen was implemented in our study.

VB is the heaviest cut of crude oil obtained from the bottom of a vacuum distillation column in oil refineries that is the base material for producing bitumen [16]. Blowing warm air on hot VB is a widely utilized process to obtain penetration grade bitumen used for paving. During this process, exothermic chemical reactions such as oxidation and de-hydrogenation take place, and the process temperature is controlled at high ranges (up to 288 °C) may change VB to more or less toxic material. Unfortunately, the neat bitumen achieved in this process is no longer suitable for paving applications because it is not able to resist against dynamic mechanical loads of passing traffic, which results in permanent deformation (rutting) and stripping under longtime loading at high temperatures [17], fatigue and cracking under longtime loading at moderate and low temperatures due to thermal stresses, aging, etc. [18]. Due to these inherent weaknesses of bitumen, many studies have been conducted to modify bitumen itself with various additives [19]. According to our knowledge, few researchers have focused on VB, such as Yousefi et al. [20]. Meanwhile, these studies have only considered improvements in physical and mechanical properties of modified bitumen. In our laboratory scale study, for the first time, a novel alternative process was used to produce paving grade bitumen with decreased PAH emissions (as a source control method) as well as improved bitumen performance grade. For this purpose, post-consumer latex and natural bitumen (NB) were used to obtain 60/70[1] penetration grade modified bitumen directly from VB without any need for the air-blowing process. Then, the quality and quantity of PAH emissions from the modified bitumen achieved were compared with conventional 60/70 bitumen to evaluate the effectiveness of the alternative production method.

Table 2
Selected physical properties of waste latex gloves.

Melting point (°C)	N/A
Density (g/cm^3)	0.91
Particle size (mm)	1–5
Transfer point (°C)	191

Table 3
The characteristics of NB.

Ash content (%)	13
Moisture content (%)	1–5
Volatile matter	60
Fixed carbon	28
Specific gravity	1.1–1.2
Color	Black
Softening point	119

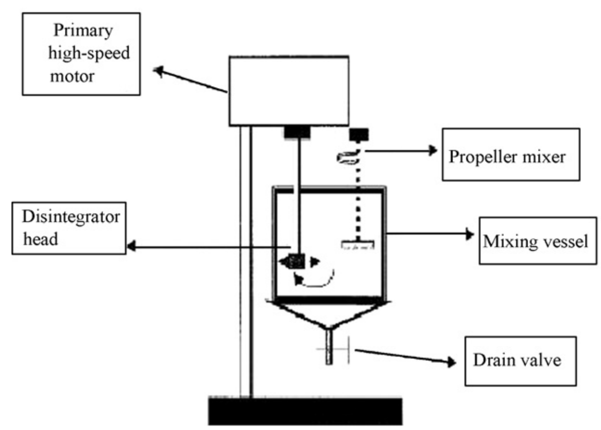

Fig. 1. High shear double-mixer set.

2. Materials and methods

2.1. Materials

Materials used in this study were VB and 60/70 bitumen (a paving-grade bitumen widely employed in Iran) from Tehran oil refineries (Table 1),[2] post-consumer waste latex gloves used in laboratories (Table 2), and NB from the Iranian Sormak Company (Table 3).

2.2. Experiment

2.2.1. Blends preparation and characterization

During the first phase of experimentation, the various blends of VB with latex and NB were prepared in order to obtain 60/70 modified bitumen without an air blowing process. To mix the components with VB, VB was first melted in an oven at 160 °C for 1 h and transferred into a preheated mixing chamber at 180 °C (Fig. 1). All conditioning and mixing process parameters and component percentages affecting the properties of blends were investigated

[1] 60/70 grade means the bitumen with penetration between 60 and 70 dmm at 25 °C according to ASTM –D5 that is used for producing of paving asphalt.

[2] For paving-grade bitumen, a PI between 2 and −2 is suitable for the temperature susceptibility of bitumen (value of approximately 1 is ideal) [21].

Fig. 2. Bitumen fume generation rig and sampling devices.

(detailed information available in Ref. [16]). The necessary samples were taken for different tests including penetration, softening point, Frass breaking point, and ductility according to ASTM D5, ASTM D36, IP 80, and ASTM D113, respectively. The penetration index (PI) and performance grade (PG)[3] of the prepared blends were estimated using semi-empirical relations. PI is calculated as follows [21]:

$$\mathrm{PI} = \frac{20(1-25A)}{1+50A}, \quad A = \mathrm{Log}(800) - \mathrm{Log}\left(\frac{P@25\,^{\circ}\mathrm{C}}{T_{\mathrm{R\&B}} - 25}\right)$$

In fact, PI is the slope of change in the bitumen consistency curve; P is penetration, and $T_{\mathrm{R\&B}}$ is the softening point temperature. The consistency of change in bitumen depends on the sign and magnitude of PI. For paving-grade asphalt, a PI between 2 and −2 is suitable for the temperature susceptibility of bitumen (value of approximately 1 is ideal) [21]. The performance grade (PG) of the obtained samples was estimated using the following semi-empirical relations [17]:

$$T_{\mathrm{DSR}} \equiv T_{\mathrm{R\&B}} + 20, \quad T_{\mathrm{BBR}} \equiv 2(T_{\mathrm{Frass}}), \quad PG = T_{\mathrm{DSR}} + T_{\mathrm{BBR}},$$

where T_{DSR} is the high temperature performance criterion; $T_{\mathrm{R\&B}}$ is the bitumen softening point; T_{BBR} is the low temperature performance criterion, and T_{Frass} is the Frass breaking point.

In order to determine the size distribution and homogeneity of blends, the morphology of the blends was viewed through a Zeiss FX optical microscope, Jenapol model. Also, the visco-elastic properties of selected blends were measured on a Paar-Physica MCR300 Rheometer in dynamic oscillatory mode using 25 mm parallel-plate geometry at 30 °C and 1–500 Hz.

2.2.2. Bitumen fume generation and sampling

Bitumen emissions were produced by a laboratory fume generation rig shown in Fig. 2. This fume generator consists of a 1500 ml round-bottomed and double-walled metal reaction chamber, which is heated by a Haake oil-circulation heater. The reported maximum temperature during the asphalt paving process (mixing paving-grade bitumen with mineral aggregates) is 180 °C, which was chosen as thermal set point of the heater. The emissions were produced by heating 400 g bitumen to 180 ± 2 °C. During heating and sampling, the bitumen was stirred with a mechanical stirrer at 200 ± 3 rpm. The same sampling equipment that is used for field monitoring studies as described in NIOSH method 5515, was used to collect the emissions [22]. The PAH components were emitted in two forms from bitumen: particles and semi-volatile vapors [23]. The particle form includes 4- to 6-ring PAH components and the semi-volatile vapors include 2- and 3-ring PAH components. Based on this fact, the particles and vapors emitted from bitumen should be collected separately using filters and sorbent tubes, respectively. The partial collection of 4-ring PAHs in sorbent tubes or 3-ring PAHs on filters is possible [24]. An integrated air sampling network was prepared according to NIOSH method 5515 as shown in Fig. 2. A 37-mm open-face filter cassette containing a pre-weighted PTFE filter (SKC-225-1709) supported with a back-up pad was used to collect the particles, and a sorbent tube (XAD-2 Cat.NO.226-30-04) that backed up the filter cassette was used to retain the semi-volatiles. The PTFE filter itself was conditioned at constant temperature and humidity for 24 h before and after sampling. A battery-operated sampling pump (SKC. S.S.S.NO-SSA-86027X) was used to conduct air sampling. The filter cassette, sorbent tube, and pump were connected via Tygon tubing. The Tygon tube between the filter cassette and the sorbent tube was wrapped with aluminum foil to minimize thermal radiation effects from the heating chamber. The sampling pump was calibrated using an electronic calibrator (BIOS Drycal DC-Lite High) before starting air sampling. In order to obtain reproducible and comparable results, the filter cassette positioning is very important; therefore, a simple holder was used to ensure exact positioning over the fume generation chamber (Fig. 2). Initially, the conventional 60/70 bitumen was tested twice and the modified 60/70 bitumen was tested the following day in similar conditions. The sampling began as soon as the bitumen reached a temperature of 130 °C, and the mechanical stirrer was turned on together with the sampling pump. Approximately 30 min after sampling commenced, the bitumen temperature was 180 °C. For all the experiments, the sampling time and flow rate were set to 2 h and 2 l min^{-1}, respectively. The conditional and process parameters were controlled, such as temperature and humidity encountered in air and flow rate sampling, storage, shipment, among other factors that can affect the efficiency of work or the quality and quantity of results.

The limits of detection (LOD) for 15 PAH compounds have been calculated via the standard deviation of blank values and the calibration curve gradient (LOD = 3 × SD/tang α). LODs for PAHs ranged from 0.02 ng/g to 0.08 ng/g. Based on the determined LOD, the limits of quantification (LOQ) were calculated. These limits are inserted in Table 5 as a separate column. In order to determine the precision of the analytical method, before any main sampling, 3 samples of blank VB emissions were taken under controlled conditions (process and environmental). The standard deviation of the obtained results equaled 106.612 ± 2.689 ng/g. All the other samples were taken under the same controlled conditions. The accuracy of the analytical method is measured and expressed as a recovery percentage. Although the true value (concentration) is rarely known for air samples, the accuracy is typically determined by spiking a sample with a known quantity of a standard. With respect to this, a sample with a known quantity of Anthracene was spiked, and % recovery for the method was calculated using the sampled value and spiked value, which is equal to 97.33%.

2.2.3. PAHs analysis

According to NIOSH method 5515, the filters containing samples were allowed to equilibrate 24 h with the laboratory atmosphere, then the total particulate mass (TPM) collected on the filters were determined gravimetrically. The quantitation limit for the gravi-

[3] PG is the high temperature and low temperature performance criterion for bitumen applied in paving asphalt.

Table 4
Some physical and mechanical properties of conventional and modified 60/70 bitumen.

Property	Conventional 60/70 bitumen	Modified 60/70 bitumen
Penetration (dmm @ 25 °C)	60	65
Softening point (°C)	50	53
Frass breaking point (°C)	−12	−21
Performance Grade (PG)	70–22	70–40
PI	−0.8	0.18

metric method was 0.01 mg. The filters were extracted twice with 5 ml of acetonitrile (Merck, p.a) for 15–20 min in an ultrasonic bath. In order to prevent the breakthrough (B.S.) values, the front-up and back-up sections of sorbent tubes were extracted separately with 5 ml toluene (Merck, p.a) for 30 min. Initially, all of the prepared extractions were analyzed with a gas chromatograph equipped with a mass spectrometer detector (GC/MSD, Agilent technology 6890N using selective ion monitoring). The analytical column, HP-5 (30 m × 0.32 mm with 0.25 μm film thickness and 5% phenyl methyl siloxane phase) was temperature programmed from 60 °C (2 min) to 290 °C (held for 5 min) at 7 °C/min. The temperature of the injector was 250 °C (splitless injection of 1 μl volume). After qualitative analysis, the extractions were analyzed with the same gas chromatograph equipped with a flame ionizing detector (GC/FID) at 300 °, using N_2 as the carrier gas at 1 ml/min to quantify the indentified PAHs with GC-MSD. Due to some analytical limitations, 15 types of PAHs were analyzed in this study.

3. Results and discussion

3.1. Engineering properties

Based on the obtained results, the blend containing 85% VB, 5% latex and 10% NB created modified 60/70 bitumen with an improved softening point, Frass breaking point, and PG in comparison with conventional 60/70 bitumen (Table 4). The PI of this blend is in the proper range, and its PG covers a wider range of climatic conditions than conventional 60/70 bitumen. Also, the complex viscosity of modified 60/70 bitumen has increased dramatically in comparison with that of the base VB, blank VB (base VB undergone mixing conditions without addition of modifier) and conventional 60/70 bitumen at 30 °C (Fig. 3). As reported by Yousefi, the lubricating constituents of VB can be absorbed by adding latex and natural bitumen, which results in an increased viscosity [25]. The storage modulus (Fig. 4) and the loss modulus (Fig. 5) of modified 60/70 bitumen have increased significantly in comparison with its conventional counterparts. The summation of storage and loss moduli gives the complex modulus of blend 10, which is a direct measure of its total resistance against deformation at 30 °C [26]. According to the Boltzmann principle, the increasing frequency acts similar to decreasing temperature and vice versa [27]; hence, the effect of increasing frequency on complex viscosity and modulus is similar to the effect of decreasing temperature, which increases complex modulus. Therefore, changes in the moduli of the modified 60/70 bitumen reveal a dramatic increase in the blend's resistance against permanent deformation. This result confirms the successfulness of using recycled latex and natural bitumen. The morphology of modified 60/70 bitumen shows homogenous dispersion of NB and latex in VB (Fig. 6). However, the NB particles seem coherent that can affect the ductility of the blend. It is possible that changing process conditions (e.g. increased mixing time or temperature) may be effective in finely dispersing NB and latex in VB.

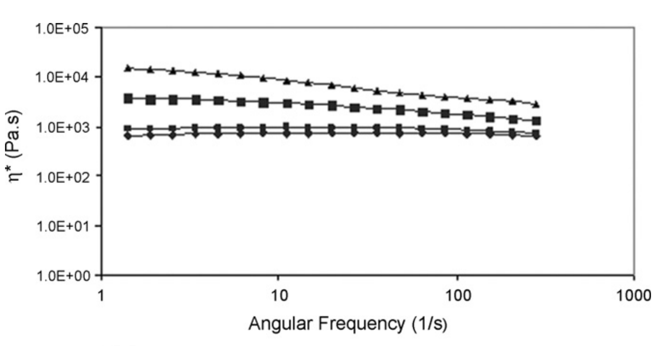

Fig. 3. Complex viscosity of samples in 30 °C.

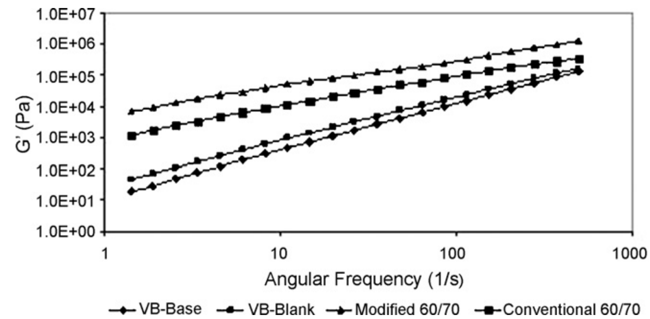

Fig. 4. Storage modulus of samples at 30 °C.

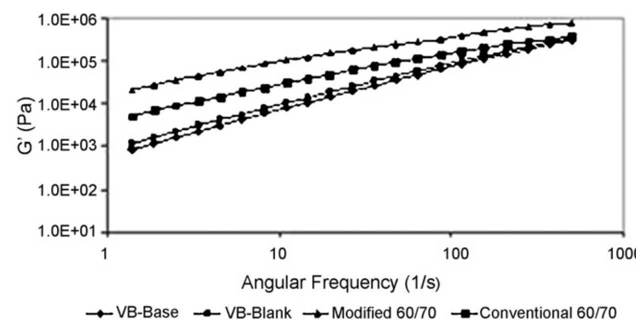

Fig. 5. Loss modulus of samples at 30 °C.

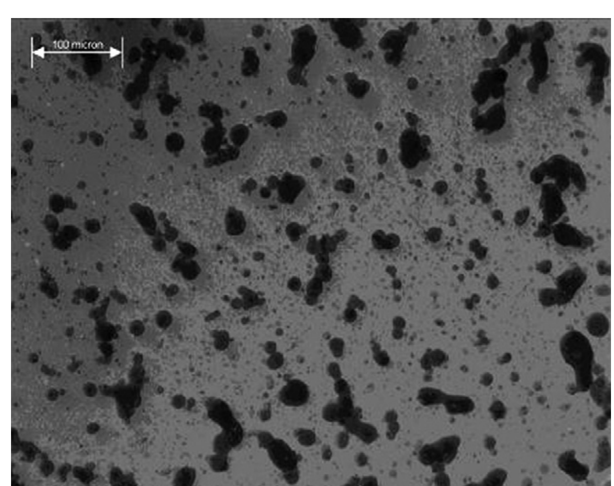

Fig. 6. Optical microscopic picture of modified 60/70 bitumen morphology.

Table 5
The identified PAHs emitted from conventional 60/70 bitumen.

PAHs	Limits of quantification (LOQ) (ng/g)	Collected on PTFE filters (ng/g)	Absorbed by XAD-2 sorbent tubes (ng/g)	Sum
Naphthalene	0.06	–	0.103	0.103
Acenaphthylene	0.2	–	12.089	12.089
Acenaphthene	0.2	–	61.017	61.017
Fluorene	0.2	–	7.497	7.497
Phenanthrene	0.2	–	57.558	57.558
Anthracene	0.2	–	33.019	33.019
Pyrene	0.2	–	10.779	10.779
Benzo[a] anthracene	0.2	–	1.226	1.226
Chrysene	0.2	–	0.591	0.591
Benzo[b] fluorantene	0.2	–	0.464	0.464
Benzo[k] fluorantene	0.2	0.377	5.926	6.304
Benzo[a]pyrene	0.26	1.710	1.167	2.878
Indeno [123-cd] pyrene	0.06	1.371	0.090	1.461
Dibenzo[ah] anthracene	0.14	0.169	0.269	0.438
Benzo[ghi] perylene	0.26	1.424	0.841	2.265
Total		5.052	192.643	197.696

3.2. Emission measurements

According to gravimetric measurements, on average, the TPM (total particulate mass) of bitumen emissions collected on PTFE were 10.418 and 10.363 μg/g for conventional 60/70 and the obtained modified 60/70 bitumen, respectively. It means that there is no considerable difference in TPM emissions between conventional bitumen and the modified bitumen, although the modified bitumen has lower emissions than conventional bitumen. In other words, the alternative method in this study has no meaningful effect on decreasing TPM emissions from bitumen. Since the particles are emitted from bitumen due to mechanical forces during mixing process, the changes conducted in bitumen structure and composition may not have significant effects on particulate emissions. The Tables 5 and 6 show the quality and quantity of all PAHs emitted as particle and vapor from conventional and modified bitumen. As seen in Table 5, the total PAHs emitted from conventional bitumen are 197.7 ng/g, 5.05 ng/g in particle form and 192.6 in vapor form. The total PAHs emitted from modified bitumen are 100.3 ng/g, 4.8 ng/g in particle form and 100.3 ng/g in vapor form (Table 6). Therefore, the quantity of PAHs emitted from modified 60/70 bitumen obtained in the alternative process is very low in comparison with conventional 60/70 bitumen.

According to these tables, all types of PAHs emitted from conventional bitumen were also emitted from modified bitumen, except naphthalene and benzo[b] fluorantene. Thus, two types of PAHs were eliminated from emissions using the alternative production process. Based on the scientific findings, the benzo[a]pyrene is the indicator component between PAHs from a carcinogenicity perspective [5]. From this point of view, the variations in benzo[a]pyrene emission are important. As seen in Table 5, the total benzo[a]pyrene emitted from conventional bitumen is 2.88 ng/g (1.71 ng/g in particle form and 1.17 ng/g in vapor form), while the total benzo[a]pyrene emitted from modified bitumen (Table 6) is 0.64 ng/g (only in particle form). Hence, the emission rate of benzo[a]pyrene from modified bitumen was decreased dramatically in comparison with that of conventional bitumen. Although, the total PAHs emitted as particles from modified bitumen were not decreased significantly in comparison with conventional bitumen, the benzo[a]pyrene emission in particle form was dramatically decreased. Based on this result, the alternative method has changed the compositions of particle form emissions of PAHs from bitumen and consequently, decreased its potential of carcinogenicity. Furthermore, the produced modified bitumen with improved quality can result in more durable paving asphalt with increased useful lifetime and consequently, reduced time-weighted average of emissions during the several years due to decreased re-paving and maintenance services.

The modified bitumens investigated by NIOSH [10], Vaananen et al. [11], and Saarela [12] during road paving were a blend of neat bitumen and various additives such as waste plastics, CRM and fly ash that are directly added to bitumen itself and therefore varies completely with the modified bitumen obtained from VB as described in the experimental section. Meanwhile, Hugener et al. [13], Brandt and Groot [14], and Kurek et al. [15] studied the PAH emissions from conventional paving grade bitumen in laboratory.

Table 6
The identified PAHs emitted from modified 60/70 bitumen.

PAHs	Collected on PTFE filters (ng/g)	Absorbed by XAD-2 sorbent tubes (ng/g)	Sum
Naphthalene	–	–	–
Acenaphthylene	–	2.558	2.558
Acenaphthene	–	15.016	15.016
Fluorene	–	12.635	12.635
Phenanthrene	–	6.896	6.896
Anthracene	––	54.563	54.563
Pyrene	–	0.527	0.527
Benzo[a] anthracene	–	1.786	1.786
Chrysene	–	0.235	0.235
Benzo[b] fluorantene	–	–	–
Benzo[k] fluorantene	0.864	1.274	2.139
Benzo[a] pyrene	0.641	–	0.641
Indeno[123-cd] pyrene	1.473	–	1.473
Dibenzo[ah] anthracene	0.390	–	0.392
Benzo[ghi] perylene	1.393	–	1.393
Total	4.766	95.495	100.261

According to our knowledge, there are no available published findings about PAH emissions of modified bitumen obtained from VB without air blowing; consequently, it is not possible to compare the results.

The argument of the authors about the decreasing effect of this method on PAH emissions from bitumen is twofold: (1) during the conventional process of bitumen production, hot air blowing leads to the oxidation of Maltene components of VB and the formation of higher molecular weight components [28]. These chemical reactions result in an increase of asphaltene that is a polar composition including high and very high molecular aromatic materials [29]; (2) in the alternative method, 15% non bituminous materials are replaced with VB. It means that 15% of original source of PAH emissions is eliminated in comparison with pure bitumen.

4. Conclusion

Based on the results, it is possible to obtain modified 60/70 bitumen directly from VB by adding waste recycled latex and natural bitumen without an air blowing process; this produces better quality bitumen and with lower PAH emissions than conventional bitumen. The PG of the resulting modified 60/70 bitumen covers a wider range of service conditions than those of conventional 60/70 bitumen, and its improved rheological properties indicates its higher total resistance against permanent deformation. Due to decreased PAH emissions from obtained modified 60/70 bitumen (approximately to 50% of PAHs emitted from conventional 60/70 bitumen), it can be concluded that the alternative method introduced by this study has decreased the mutagenic and carcinogenic potential of produced bitumen. In comparison with conventional methods, the alternative method has several advantages: (1) decreased PAH emissions from bitumen; (2) elimination of air blowing process and considerably decreased costs; (3) partial substitution of natural bitumen and recycled post-consumer latex with VB and consequently, contribution to the preservation of bitumen resources and avoiding environmental impacts of post-consumer latex; and finally (4) production of better quality bitumen.

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