

# An environmental-friendly procedure for recycling automotive paint sludge

Hossein Jalaei Salmani <sup>1</sup>, Mahdi Yousefi <sup>2</sup>, Abolfazl Sanjarifard <sup>1</sup>, S. Mostafa Nowee <sup>1</sup>, Aliasghar Asgari <sup>3</sup>, Emad Sedaghat <sup>4</sup>, Kazem Lakzian <sup>5,\*</sup>

<sup>1</sup> Department of Chemical Engineering, Faculty of Engineering, Ferdowsi University of Mashhad, Mashhad, 91779-48974, Iran. email: jalaei.chem@yahoo.com

<sup>2</sup> PRC for Frontier Energy Technologies and Utilization, University of Newcastle, Callaghan, NSW 2308, Australia

<sup>3</sup> School of Mechanical Engineering, Shiraz University, Shiraz, Iran

<sup>4</sup> Sanatbam waterproofing membrane Co., Mashhad, Iran

<sup>5</sup> Department of Safety, Health, and Environmental Engineering, National Kaohsiung University of Science and Technology, Kaohsiung, Taiwan

\* Correspondence: Kazem Lakzian emails: kazem.lakzian@gmail.com. kazemlakzian@nkust.edu.tw

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## Abstract

Paint sludge is one of the non-biodegradable and undesirable wastes which are annually produced in large quantities by automotive manufacturing plants. The current study investigates the possibility of producing basic modified bitumen by mixing three different types of solvent-based paint sludge (primer, base, and clear coat) with bitumen. The findings showed, that as the amount of all three types of paint sludge increases, ductility and penetration decrease while softening point remains nearly unchanged. The modified bitumen can be used directly in non-extreme stress applications such as bituminous waterproofing membranes or sealing. It can be modified further with other polymers for more serious applications such as asphalt. Therefore, the paint sludge, hazardous yet valuable waste in the automotive industry, could be used either as a bitumen additive or bitumen filler rather than being discharged into the environment. This approach contributes the efficient management of energy resources. The benefits associated with the proposed approach are enhanced in countries with abundant oil resources.

**Keywords** Automotive paint sludge, environment, bitumen, recycling, energy resources

## 1. Introduction

Natural bitumen has been used as an adhesive and a waterproofing agent for a very long time [1, 2], and it has variety of applications, including roofing coats, road paving, and bitumen paint [3, 4]. Thus, it is necessary to improve the properties of this substance due to its various applications. Several types of polymer could be used for the improvement of bitumen properties such as SBS (styrene butadiene styrene) [5-7], SBR (styrene butadiene rubber) [8, 9], PE (polyethylene) [10, 11], and EVA (ethylene-vinyl acetate) [7, 12]. These additives have been proven to improve bitumen's mechanical properties and characteristics [2, 13]. Due to the high cost of virgin polymers, recycled materials can be considered as a viable alternative from a cost standpoint. The development of recycled and waste polymers has a significant environmental benefit because non-biodegradable materials are removed from the environment [14]. Various

types of waste have been used in modified bitumen; for example, Moghadas Nejad et al. [15] demonstrated that crumb rubber reduced penetration and ductility, but increased softening point. Similarly, Soudani et al. [14] showed that waste nitrile rubber (NBR) extracted from shoe soles could reduce the penetration while the softening point increment was unavoidable, and Behl et al. [16] indicated that, unlike the softening point, the penetration decreased with the addition of 5% of waste polyvinylchloride (PVC). Polyethylene terephthalate (PET) as waste plastic bottles [17] and recycled PE from grocery bags [18] are two other wastes that have been used in bitumen.

One of the major challenges currently confronting most industrial plants is the generation of undesirable waste while producing desirable products. Wastes, such as paint sludge produced by automotive plants, have serious environmental consequences that may endanger the lives of many species. The demand for

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automobiles, and thus their production, has risen dramatically in recent years. For instance, the estimated global vehicle production was 90, 97, 92, and 78 million units in 2014, 2018, 2019, and 2020, respectively, whereas this number in 2000 was 58 million units [11]. The main source of hazardous wastes at an automotive assembly plant is originated from painting units which bring serious environmental consequences [19]. Painting an automobile involves several stages, starting with the application of a premier coat, which is the first layer to be applied and makes the application of the base coat easier. Sequentially, the base coat which brings color to the vehicle is laid on. Finally, the clear coat is applied to the surface as the final layer, providing protection against direct sunlight and UV radiation, abrasion, humidity, and other types of chemical aggression. The body is then dried completely. During the process, in order to meet the target, paint is oversprayed and a significant amount of waste is generated, which is washed with circulating water and collected in a sludge pit. The waste mixture of paint (primer, base, and clear coat) and water accumulated in each stage is called “paint sludge”. Consequently, the massive amount of accumulated paint sludge, which contains uncured polymer resins, pigments, organic solvents, and other chemical components, poses serious managerial and environmental challenges that must be addressed properly [20-22]. Furthermore, paint sludges have hazardous properties such as low flash points and high concentrations of heavy metals, volatile organic compounds (VOCs), and other toxic substances, resulting in a higher pollution possibility [23-25]. Some patents have been issued as a result of the use of paint

sludge in the sealant industry [26], composite materials [27], and concrete additives [28, 29]. Segala also used paint waste to create a type of a latex pigment (PLP) [30]. In the cement kiln, paint sludge was used as an additional fuel in a few studies to completely destroy the sludge [31, 32]. Avci and colleagues used recycled paint sludge as an aggregate in the production of construction materials [33]. An experimental study was conducted by Ruffino et al. [34] to compare the performance of traditional asphalt concrete mixtures with those that were modified with paint sludge (10% on the base bitumen). The recycling technique for every paint sludge requires its own investigation because the composition of paint sludge from each individual plant differs from that of others [33]. Li et al. (2018) recently investigated the use of paint sludge as a pore forming agent in the preparation of sewage sludge-derived carbon for the adsorption of various contaminants [35]. In another study, in order to produce bio-oil, Jayakishan et al. (2019) co-liquefied paint sludge rich in hydrocarbons with *Prosopis juliflora* biomass [36]. In one study [37], the dried and powdered paint sludge samples were mixed with the base bitumen to create modified bitumen. Water-based paint sludge samples were used in that investigation. Table 1 summarizes the aforementioned methods in various studies and their implemented approaches for recycling or disposal of paint wastes.

Biological processes for recovering water-based paint sludge have been proposed by some authors [32, 38, 39]. The high amounts of organic carbon and nitrogen in water-based paint sludge, as well as the low solvent content, made composting or bio-drying this waste product appealing [23].

**Table 1** Various approaches for managing paint sludge acquired from automotive plants.

Method	Approach	Final product application
Recycle	Removing volatile organic components by increasing temperature	Sealant industry [26]
Recycle	Removing volatile organic components by increasing temperature	Composite materials [27]
Recycle	Addition of dried polymer powder to cement for preventing cracks	Concrete additives [28]
Recycle	Addition of dried polymer powder in the production of construction materials	Tiles, blocks, and bricks [29]
Recycle	Combining recyclable materials and agglomeration agents	Create a type of a latex pigment (PLP) for cement industries [30]
Disposal	Co-processing in burning with other fuels	Cement kiln [31, 32]
Recycle	Addition of dried paint sludge in the production process of construction materials	Production of cement and lime [33]
Recycle	Addition of dried and milled paint sludge in the production of construction materials	Production of asphalt concrete mixtures [34]
Recycle	Making porous structures for adsorption by mixing dried paint sludge and dried sewage sludge	Production of a macro-pores forming agent [35]
Recycle	Mixing of paint sludge with woody biomass of <i>Prosopis juliflora</i> plant	Production a new bio-oil [36]
Recycle	Addition of dried and milled paint sludge in the production of construction materials	Production of modified bitumen [37]

**Table 2** Physical properties of bitumen.

Test	Standard	Result
Softening point (°C)	ASTM D36	55
Ductility at 25°C (mm)	ASTM D113	930
Penetration at 25°C (0.1 mm)	ASTM D5	65

## 2. Experimental

### 2.1 Materials

Bitumen with a standard penetration grade of 60/70 was used in the current study which was supplied from an Iranian refinery. The main physical characteristics of the base bitumen are given in Table 2. Additionally, three types of paint sludge were used including a primer coat, base coat, and clear coat which all were wastes produced in an automotive manufacturing plant. Due to an agreement between the automotive manufacturing plant and researchers of this study, the properties of paint sludge remained under cover. Meanwhile, the primer coat, base coat, and clear coat were commercial solvent-based materials.

### 2.2 Storage stability and blends design

Storage stability test determines the possibility of separating modified bitumen mixtures under storage. The test is performed by pouring the bitumen mixtures into vertical aluminum foil tubes with 3 cm in diameter and 30 cm in height which is kept for 48 h in an oven at 163°C without any disruption. Afterward, the sample is placed in a freezer for 4 h at -7°C to solidify and then cut into three equal pieces horizontally. Softening points of the bottom and the top of each tube are measured, in the case where the difference is less than 2.5°C, it indicates good storage stability [14, 40]. This test has been done to determine the maximum amount of each additive (Table 3).

### 2.3 Preparation of samples

The paint sludge samples included primer, base, and clear coat that were obtained from different stages of the painting process. Moreover, after each stage of painting, a sprayed aqueous solution was used to clean the painted surfaces. Therefore, the acquired paint sludge samples initially contained primer, base, or clear coat, as well as a small amount of water in each sample. One of the most important aspects of this method was creating a homogeneous mixture by combining neat bitumen and various paint sludge samples. Thus, the water in the sample was removed using an oven set to 30°C, and the sample was kept in this condition for 48 hours. The temperature was set at 30°C because high temperatures may alter the chemical structure of polymers, reducing the efficiency of the final modified bitumen. Therefore, this temperature will remove water from the samples while also preserving the chemical structure of polymers as much as possible prior to the production of modified bitumen. In some studies [28, 29, 33-35, 37], high-temperature processes were used to dry samples and make a fine powder of the samples, and then dried powder paint sludge samples from the previous step were used to make modified bitumen. However, in the current study, the paint sludge samples were dough-formed before being mixed with neat bitumen. As a result, the obtained solvent-based paint sludge was

applicable to create a homogenous mixture with neat bitumen. One of the advantages of this study over the previous studies is that less energy may be required for the sample preparation prior to mixing with neat bitumen.

In the next step, a certain amount of bitumen was heated to melt, then a predetermined amount of the paint sludge was added and the mixture was blended at 160°C by a high shear mixer with a shearing speed of 1000 rpm for 45 minutes.

### 2.4 Physical properties test

Since the main objective of this study was to recycle industrial paint sludge (not producing modified bitumen), just conventional tests were done to determine the characteristics of the modified bitumen. The performed tests of bitumen and paint sludge mixtures were namely, softening point (ASTM D36), penetration (25°C, ASTM D5), and ductility (25°C, ASTM D113) [14].

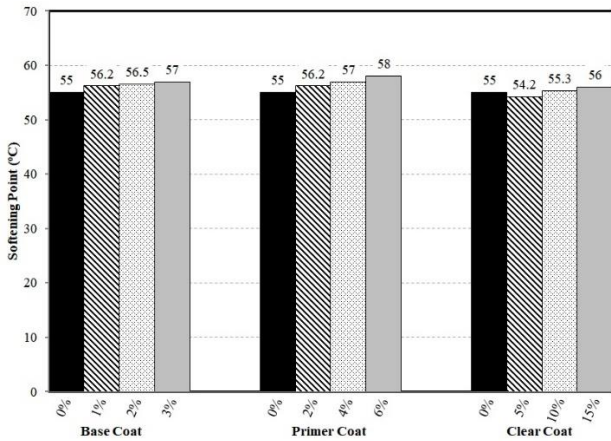
## 3. Results and discussion

The main goal of this research was to find a way to avoid dumping paint sludge into the environment rather than producing modified bitumen. All of the results presented in this section are based on a comparison of the properties of base bitumen with obtained modified bitumen. The maximum composition for each paint sludge type has been determined with respect to its miscibility which was accessible by storage stability (Table 3). According to Table 3, clear coat and base coat had the maximum and the minimum miscibility, respectively, and the miscibility of primer coat was in between.

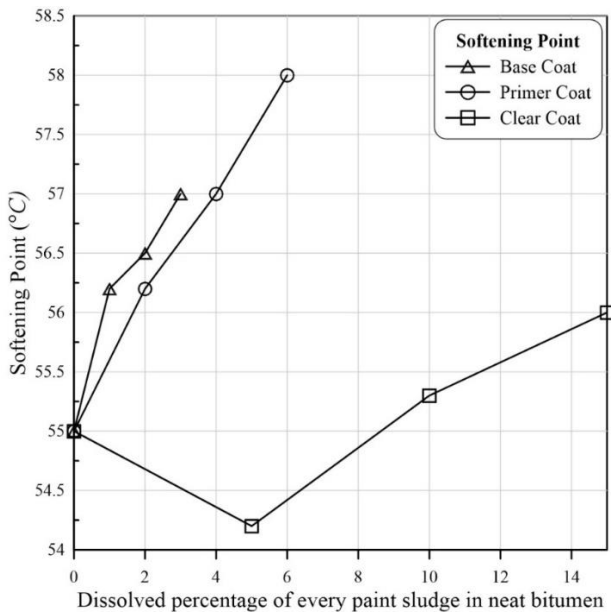
The bitumen softening point test is used to determine the consistency of bitumen. Furthermore, this test determines the temperature at which the bitumen attains a specific viscosity (ISIRI 3868-2<sup>nd</sup> reversion, originally adopted from ASTM D36-95 (2005) and AASHTO T53-96 (2000)).

In all figures, bitumen without additive is presented as 0%. It can be seen that from Figure 1, all types of paint sludge had no substantive effect on the softening point and remained almost unchanged. The percentage of each paint sludge sample that was miscible in neat bitumen and the corresponding softening points are shown in Figure 2. The base coat demonstrated the lowest miscibility with neat bitumen (up to 3%), in which the softening point changed by 2°C. The primer coat exhibited more miscibility compared to the base coat (up to 6%), changing the softening point from 55 to 58°C. The highest miscibility was observed in the mixture of clear coat with neat bitumen (up to 15%); however, it only varied by  $\pm 1^\circ\text{C}$  across all compositions. Therefore, the addition of clear coat to neat bitumen showed the lowest change in softening point when compared to pure bitumen, while a higher percentage of clear coat can mix with pure bitumen when compared to the other two types of paint sludge samples. Furthermore, the clear coat is the only paint

sludge that could lower the softening point (by nearly 1°C), whereas the other two types of samples can only rise the softening point.



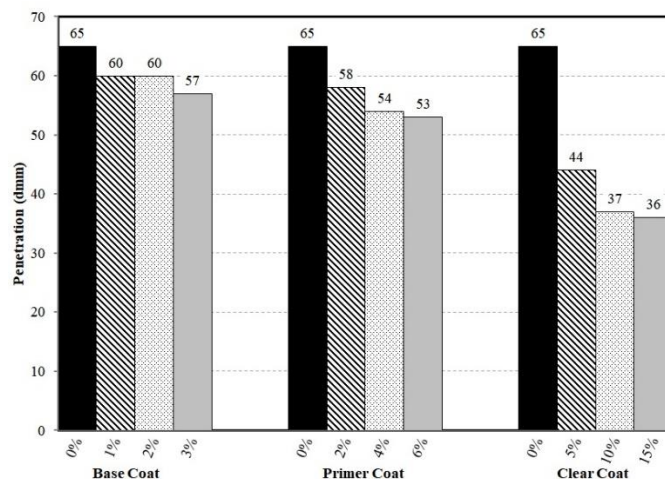
**Figure 1** Effect of each paint sludge addition on softening point.



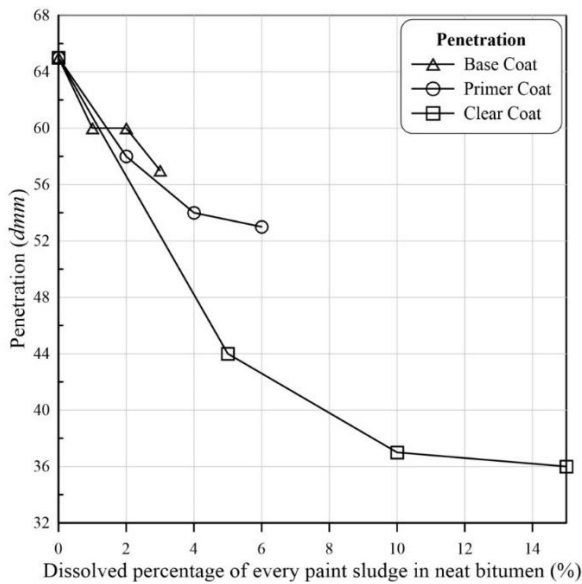
**Figure 2** Comparison of addition of different percentages of paint sludge samples on the softening point of modified bitumen.

The penetration test is used to evaluate the resistance of bitumen samples; this means samples with higher penetration values have less resistance. In this test, the penetration rate is determined by inserting a needle into a bitumen sample (ISIRI 2950 – 2<sup>nd</sup> revision, originally adopted from ASTM D5-05 (2005) and AASHTO T49-97 (2000)). The deeper a needle penetrates into a bitumen sample, the greater the penetration value. As a result, this test is used to evaluate the resistance of a bitumen sample.

According to Figure 3 and Figure 4, a declining trend was observed for penetration by adding up the amount of sludge additives. Comparatively, 2% primer coat had more impact than 2% base coat and 5% clear coat caused more reduction than 4% and 6% primer coat. Furthermore, the decreasing trend for clear coat persisted until the maximum miscible composition of this chemical was reached (i.e., 15%). The addition of a clear coat to neat bitumen reduced the penetration value to 32 dmm, whereas neat bitumen had a penetration value of 65 dmm. Overall, the clear coat had the most influence on penetration, amongst which the most decline and miscibility was related to 15% clear coat. This suggests that the interaction between clear coat and bitumen results in a more rigid structure, likely due to the nature of the polymeric components in the clear coat, which enhance stiffness and reduce the penetration value. The observed trend indicates that increasing the concentration of clear coat leads to progressive changes in bitumen's consistency, making it less susceptible to deformation under applied force. Additionally, variations in the penetration values across different sludge additives highlight the role of their distinct chemical compositions and interactions with bitumen. While the primer and base coats also contributed to penetration reduction, their effects were comparatively lower, suggesting differences in their compatibility and dispersion within the bitumen matrix. The findings emphasize that the choice and proportion of sludge additives play a crucial role in determining the final properties of modified bitumen, which can be tailored to meet specific performance requirements in various applications.



**Figure 3** Effect of each paint sludge addition on penetration.



**Figure 4** Comparison of addition of different percentages of paint sludge samples on the penetration of modified bitumen.

The ductility test determines the flexibility of a bitumen sample. This test measures the maximum flexibility of a sample before separation by stretching the bitumen-based samples (ISIRI 3866 – 2<sup>nd</sup> revision, originally adopted from ASTM D113-99 (2005) and AASHTO T51-00 (2000)). In the present study, the ductility was the most affected physical property by the addition of paint sludge samples (Figure 5). Among all paint sludge samples, the clear coat at its most weight percentage (15%) led to the most differences. Figure 6 depicted the ductility reduction trend

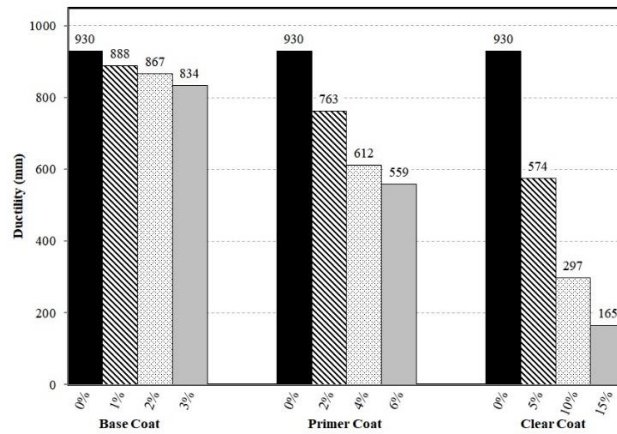
line for three types of paint sludge samples in different compositions.

From these figures, it is concluded that though clear coat had enhanced the penetration by reduction, the ductility has also dropped dramatically, while the softening point remained relatively unchanged. The addition of the clear coat significantly reduced bitumen penetration, which could be attributed to better mixing of clear coat polymers and the formation of stronger bonds with neat bitumen. It is worth noting that a higher percentage of clear coat could mix with neat bitumen than the other two paint waste types. By selecting the appropriate amount of paint sludge, a trade-off in the properties of the final mixture should be considered depending on the application of bitumen.

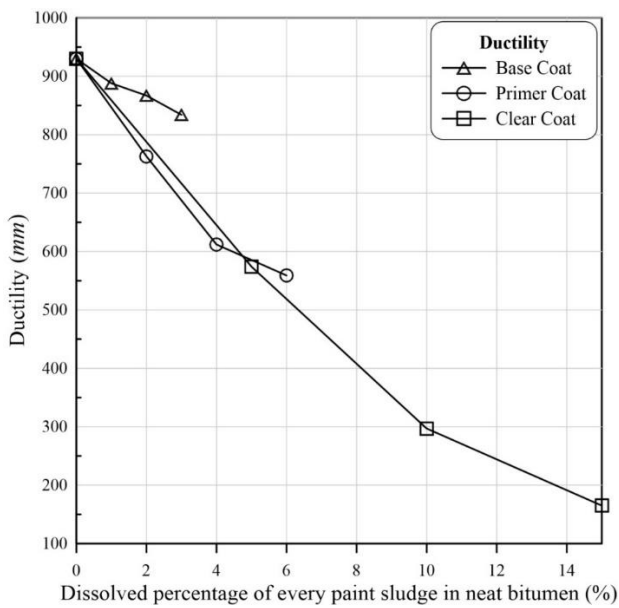
The low miscibility of the base coat, on the other hand, restored the bitumen properties to those of pure original bitumen. In general, it could be stated that the addition of all three types of paint wastes had a negligible impact on the softening point, but they partially reduced penetration and ductility drastically. Consequently, the applicability of the obtained basic modified bitumen depends on its intended use. For instance, if bitumen with lower penetration and ductility is required, blending clear coat with neat bitumen may be the optimal choice. In contrast, combining base coat with neat bitumen retains the three key properties of modified bitumen, making it comparable to neat bitumen. Furthermore, the modified bitumen can be treated as neat bitumen, allowing for additional modifications to achieve the desired properties for various applications.

**Table 3** Results of storage stability test.

Paint sludge content (% by total weight)	Top softening point (°C)	Bottom softening point (°C)	Softening point difference (°C)
Base coat (A)	-	-	-
Bitumen + 2% A	55.8	56.9	1.1
Bitumen + 3% A	56	58.1	2.1
Bitumen + 4% A	56.7	59.9	3.2
Primer coat (B)	-	-	-
Bitumen + 2% B	56.2	55.9	0.6
Bitumen + 4% B	56.1	58	1.9
Bitumen + 6% B	57.3	59.7	2.4
Bitumen + 8% B	58.6	62.2	3.6
Clear coat (C)	-	-	-
Bitumen + 2% C	54	54.2	0.2
Bitumen + 10% C	55.2	56.2	1.0
Bitumen + 14% C	56.2	55.4	0.8
Bitumen + 15% C	55.5	57.3	1.8
Bitumen + 16% C	58.6	56.1	2.5



**Figure 5** Effect of each paint sludge addition on ductility.



**Figure 6** Comparison of addition of different percentages of paint sludge samples on the ductility of modified bitumen.

The primary objective of this study was to identify and propose a viable approach to mitigating environmental concerns associated with paint waste generated in the automotive industry. Additionally, the findings demonstrated that this method is not only effective in recycling paint sludge but also in maintaining the quality of bitumen while producing basic modified bitumen suitable for various applications.

Ultimately, the suitability of the obtained modified bitumen mixtures depends on their intended applications, and further modifications can be made to enhance their compatibility for specific uses. Moreover, the proposed approach offers a sustainable solution for managing paint sludge in the automotive industry, thereby minimizing its environmental impact. It is important to emphasize that the primary objective of this research was to propose a feasible method for recycling paint sludge rather than to develop optimized modified bitumen.

#### 4. Conclusions

In this study, three types of paint sludge namely, primer coat, base coat, and clear coat were blended with

60/70 bitumen. Conventional and basic tests such as ductility, penetration, and softening point were performed on the final mixture. The miscibility of three wastes was investigated by storage stability test, and determined that miscibility of clear coat with 15 percent by weight was more than other wastes, followed by primer and base coat with 6% and 3%, respectively. The properties of paint sludge remained unknown due to an agreement between the automotive manufacturing plant and the researchers of this study. While it should be noted that the primer coat, base coat, and clear coat were all commercial solvent-based materials.

The following findings help to clarify some of the research's key points:

- For all types of solvent-based paint sludge samples, increasing the added amount reduced ductility and penetration, while had showed little effect on the softening point,
- The solvent-based nature of the used paint sludge samples aided in the production of homogeneous modified bitumen blends,
- One of the advantages of this study over the previous studies is that less energy may be required for the sample preparation prior to mixing with neat bitumen,
- Due to the high cost of virgin polymers, recycling paint sludge can be considered a cost-effective alternative,
- The obtained modified bitumen can be used directly or with further modifications for a variety of applications,
- The proposed method can be used in countries with abundant bitumen sources to minimize the negative environmental impacts of these chemicals.

Conclusively, this experimental investigation proved that paint sludge which is a hazardous waste in the automotive industry could be used as an additive to bitumen rather than discharge to the environment. The findings demonstrated that the proposed method could not only recycle paint sludge but also produce usable modified bitumen for other applications. It should be stated that the primary goal of this study was to propose a practical method for recycling the

paint sludge rather than to provide versatile modified bitumen. Another significant advantage of this approach is its role in enhancing the efficient management of energy resources. By repurposing paint sludge as a bitumen additive or filler, this strategy not only mitigates environmental pollution but also promotes sustainable resource utilization in the automotive and construction industries.

Generally, the findings of this study can be further evaluated through advanced testing methods to identify the most suitable components of each type of paint sludge for specific applications. Conducting sophisticated analyses, such as rheological, mechanical, and chemical characterization, would provide deeper insights into the interactions between paint sludge and bitumen, allowing for a more precise determination of optimal compositions. Moreover, long-term performance assessments, including aging resistance, thermal stability, and durability tests, could help refine the applicability of modified bitumen in various industrial settings. These additional investigations would enhance the understanding of how different paint sludge components influence bitumen properties, facilitating the development of more tailored and efficient recycling strategies.

### Author Contributions

Dr. Hossein Jalaei Salmani was responsible for conceptualization, methodology, validation, writing - original draft, and supervision.

Dr. Mahdi Yousefi was responsible for writing - review & editing.

Mr. Abolfazl Sanjarifard was responsible for investigation and writing - original draft.

Dr. S. Mostafa Nowee was responsible for writing - original draft and supervision.

Mr. Aliasghar Asgari was responsible for writing - review & editing.

Mr. Emad Sedaghat was responsible for investigation.

Dr. Kazem Lakzian was responsible for formal analysis, writing - original draft & Review & Editing, visualization, and supervision.

### Competing Interests

The authors report no declarations of interest.

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