

Article

Oil Sludge as a Rejuvenator for Aged Bitumen: Structural and Thermal Effect

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Abstract: This study explores the potential of oil sludge, a hazardous by-product of the oil industry, as a sustainable rejuvenator for restoring the physicochemical and rheological properties of aged bitumen. Aged binder samples were modified with different concentrations of oil sludge (1%, 3%, and 5%) and analyzed using dynamic shear rheometry (DSR), Fourier-transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM), and thermogravimetric analysis (TGA). The incorporation of 5% oil sludge increased penetration from 60 to 71 mm and the softening point from 55 °C to 72 °C, indicating enhanced flexibility. DSR measurements showed a ~10% decrease in complex modulus (G^*) and a slight increase in phase angle, confirming partial rheological recovery. FTIR spectra revealed partial restoration of aliphatic and aromatic functional groups, with a decrease in sulfoxide absorption bands, while SEM analysis indicated improved homogeneity and reduced microcracking. TGA confirmed enhanced thermal behavior and a reduction in residual mass. The novelty of this work lies in the first-time application of regionally sourced oil sludge as a rejuvenator, evaluated through a multiscale analytical framework. These findings demonstrate the dual benefits of performance recovery and hazardous waste valorization, contributing to sustainable road maintenance within a circular economy approach.

Keywords: oil sludge; bitumen rejuvenation; circular economy; waste valorization; sustainable pavement



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1. Introduction

Highways and related infrastructure are critical to national economic development, and asphalt pavements—due to their flexibility and durability—remain a key component of this network [1]. However, over time, exposure to oxidative aging, UV radiation, and environmental stressors such as moisture and traffic loading leads to severe degradation in the physicochemical properties of bitumen, the primary binder in asphalt concrete [2,3]. The aging process involves the transformation of light fractions into heavier ones—aromatics into resins and then into asphaltenes, eventually forming carbenes and carboids [4,5]. These changes increase binder stiffness, reduce flexibility, and impair pavement performance [6–8].

Bitumen aging is typically divided into short-term (technological) aging, which occurs during production and mixing with aggregates, and long-term (operational) aging, which takes place over time under environmental influences such as UV radiation, oxygen, and traffic loads [9]. To counteract these effects, rejuvenators are employed to reverse aging-induced alterations. An effective rejuvenator should not only soften aged bitumen by reducing viscosity and increasing elasticity, but also rebalance its molecular composition, particularly the ratio of maltenes to asphaltenes [10–12]. The restored binder should regain its flexibility and resistance to thermal and mechanical stresses, thereby prolonging pavement service life.

A wide range of rejuvenators has been explored, including petroleum-based oils, waste engine oils, and bio-based alternatives such as castor, soybean, and palm oils [13–16]. These additives have shown promise in improving the flexibility of aged asphalt, restoring functional group balance, and reducing chemical aging markers such as carbonyl and sulfoxide indices. More recent studies have focused on renewable sources like agricultural residues, wood chips, and waste cooking oils, aligning with global trends toward sustainability [17–21].

Other industrial by-products, including aromatic extracts, vacuum residues, and used lubricants, have also demonstrated rejuvenating potential [22–26]. Among them, oil sludge—a hazardous semi-solid residue from petroleum refining—has drawn increasing attention due to its similar hydrocarbon structure to bitumen [27]. Globally, petroleum operations generate substantial volumes of oil sludge: each U.S. refinery produces up to 30,000 tons annually, while Chinese petrochemical industries generate approximately 3 million tons per year [28]. In Kazakhstan, the annual production reaches around 120,000 tons. This growing volume of hazardous waste poses both environmental risks and economic losses, reinforcing the need for sustainable reuse solutions. While traditionally considered waste, oil sludge contains valuable light fractions and aromatic compounds that may enable its application as a low-cost, circular economy rejuvenator.

Building upon these findings, the present study seeks to deepen the understanding of oil sludge's role as a rejuvenator for aged bitumen. The primary objective is to evaluate its effectiveness in restoring the physicochemical and structural properties of aged binders and to explore the potential of utilizing this hazardous waste material within a sustainable circular economy framework. The novelty of this work lies in its focus on a regionally sourced oil sludge and its potential dual benefit of enhancing asphalt performance while mitigating environmental burden.

2. Materials and Methods

2.1. Raw Materials

In this study, primary bitumen grade BND 70/100 (unmodified bitumen with penetrating ability of 70/100) was used, the main characteristics of which are presented in Table 1.

Table 1. Initial characteristics of bitumen grade BND 70/100.

Name of the Characteristic	Penetration at 25 °C, 01 mm	Softening Temperature, °C	Ductility, cm
Indicators	71	48	>100

This bitumen was used to prepare artificially aged bitumen using a thin-film oven (RTFO, London, UK) at 163 °C for 85 min according to [29]. After aging, the bitumen was heated to 160 ± 5 °C to ensure a fluid state. Then, oil sludge was introduced at room temperature in amounts of 1%, 3%, and 5% by weight. The components were blended

using a high-speed laboratory mixer (IKA T25 Digital, Staufen, Germany) at 4000 rpm for 30 min to achieve homogeneous dispersion.

The oil sludge used in this study was obtained from petroleum processing plants in Kazakhstan and represents a semi-solid, dark brown hydrocarbon waste. It contains approximately 25% water and a complex mixture of paraffinic, aromatic, resinous, and asphaltenic components. Based on extended component analysis, the organic fraction includes $4.55 \pm 0.5\%$ asphaltenes, $7.2 \pm 1\%$ resins, $3.0 \pm 0.5\%$ paraffinic hydrocarbons, and $28.2 \pm 0.5\%$ aromatic compounds. In addition, residue analysis revealed that the sludge contains $31.2 \pm 1\%$ mechanical impurities, including silicates, oxides, and sulfates. According to previous investigation [30], oil sludge samples underwent pretreatment involving gravity separation, filtration, and hydrocarbon extraction using solvent blends such as white spirit and hexene, which were shown to be highly effective in recovering the bitumen-like fraction. This step ensured that the rejuvenator used in this study was representative of the recoverable hydrocarbon phase of the sludge. Owing to its chemical similarity to bitumen—particularly the presence of maltene-like and polar fractions—it was selected as a potential rejuvenating additive for restoring aged binder properties [31].

2.2. Physical Property Tests

The physical properties of both aged and reclaimed bitumen binders were assessed using a number of standard test methods.

2.2.1. Penetration Test

Penetration at 25 °C was measured in accordance with ASTM D5 to evaluate the consistency and softness of the binder.

2.2.2. Softening Point

The softening Point was determined using the ring-and-ball method according to ASTM D36, which reflects the thermal susceptibility of bitumen.

2.3. Rheological Properties

The rheological properties of the bituminous materials were measured using a dynamic shear rheometer (DSR, NETZSCH-Gerätebau GmbH, Selb, Germany) according to ASTM D7175 to determine the complex shear modulus (G^*) and phase angle (δ).

2.4. FTIR Spectroscopy

Fourier Transform Infrared (FTIR) spectroscopy was conducted using an ALPHA II spectrometer (Bruker, Ettlingen, Germany) in attenuated total reflectance (ATR) mode across the spectral range of 500–4000 cm^{-1} .

2.5. Scanning Electron Microscopy (SEM)

Surface morphology was examined using a JSM-6490LA scanning electron microscope (JEOL Ltd., Tokyo, Japan) at an acceleration voltage of 2–10 kV and up to 500× magnification.

2.6. Thermogravimetric Analysis (TGA)

TGA was carried out on a NETZSCH STA 409 instrument (NETZSCH-Gerätebau GmbH, Selb, Germany) to assess the thermal degradation and residue of each sample. The tests were performed from 50 to 600 °C at a heating rate of 20 K/min under a nitrogen atmosphere.

2.7. SARA Fractionation

The separation of bitumen into SARA fractions (Saturates, Aromatics, Resins, and Asphaltenes) was performed in accordance with ASTM D4124. The method involved the

precipitation of asphaltenes using n-heptane, followed by chromatographic separation of maltenes into saturates, aromatics, and resins using silica gel. Each fraction was dried and weighed to determine its mass percentage. The asphaltene-to-maltene ratio (A/M) was then calculated using the following formula:

$$A/M \text{ ratio} = \frac{\text{Mass of asphaltenes}}{\text{Mass of resins} + \text{aromatics} + \text{saturates}} \quad (1)$$

This ratio was used as an indicator of the colloidal balance of the bitumen and to assess the rejuvenating effect of oil sludge.

3. Results

3.1. Study of Physical Properties of Aged Bitumen After Introduction of Oil Sludge

The physical and mechanical properties such as penetration, softening temperature, and rheological properties of artificially aged bitumen were measured and compared with virgin bitumen to evaluate the efficiency of oil sludge recovery.

Table 2 presents the penetration and softening point values of virgin, aged, and oil sludge-modified bitumen samples. As expected, the aged asphalt binder exhibited reduced penetration and an increased softening point due to oxidative hardening and molecular rearrangement. The incorporation of oil sludge led to a notable enhancement in both parameters. At a 5% dosage, the penetration value even surpassed that of the virgin binder, indicating a pronounced rejuvenating effect.

Table 2. Effect of oil sludge application on the physical properties of bitumen.

Indicators	Aged Bitumen	Amount of Oil Sludge, %		
		1	3	5
Penetration, 0.1 mm at 25 °C	60	63	67	71
Softening point, °C	55	60	68	72

Note: All values represent the average of three replicate measurements.

During the aging process, bitumen typically requires higher temperatures to soften, resulting in elevated softening point values [32]. In this study, the softening point continued to increase with oil sludge addition, which may reflect improved cohesion and thermal stability within the modified binder. Similar effects of organic additives on penetration and softening behavior have been observed in other studies [33].

3.2. Effect of Oil Sludge on the Rheological Properties of Bitumen

The authors [34] note that the aging process is accompanied by the loss of volatile substances and the migration of oily components from the bitumen into the aggregate, as well as slow, reversible hardening upon heating, which is manifested in a change in the viscosity of the bitumen over time. For a deeper understanding of the structural changes in bitumen, dynamic shear rheometry (DSR) was employed as a key method to assess its viscoelastic behavior. These methods are widely used to analyze the properties of bitumen materials, since each of them provides unique information on molecular structure and rheological characteristics.

Dynamic shear rheometry (DSR) is an important method for the analysis of the mechanical properties of bitumen binders, allowing the evaluation of the characteristics of bitumen in practical applications [35]. The softening point is traditionally regarded as a temperature threshold at which the binder transitions from a high-viscosity to a more flowable, low-viscosity state, indirectly reflecting its thermal behavior during practical application [36].

The introduction of oil sludge (Table 3) into aged bitumen leads to a decrease in the complex modulus (G^*) and phase angle (δ), indicating the restoration of the material's elasticity and flexibility (Figure 1). According to the study by Liu et al. [37], a decrease in the $G^*/\sin \delta$ value indicates lower energy loss under repeated loading and improved fatigue properties of the material. Thus, the use of oil sludge helps improve the fatigue properties of bitumen, which is similar to the effect of rejuvenators that restore the molecular structure of bitumen.

Table 3. DSR results for bitumen samples.

DSR Results	δ [°]	G^* [Pa]	$G^*/\sin \delta$ [Pa]	T [°C]
Aged bitumen	84.70 ± 0.1	652.2 ± 0.5	655 ± 0.5	66.5 ± 0.3
1% oil sludge	84.55 ± 0.1	594.8 ± 0.8	597.5 ± 1.0	71.6 ± 0.5
3% oil sludge	84.52 ± 0.1	581.5 ± 0.8	584.2 ± 0.8	71.6 ± 0.3
5% oil sludge	84.12 ± 0.1	582.1 ± 0.9	584.5 ± 1.0	71.4 ± 0.3

T [°C] indicates the actual test temperature used in the DSR measurements to evaluate the high-temperature rheological behavior of bitumen.

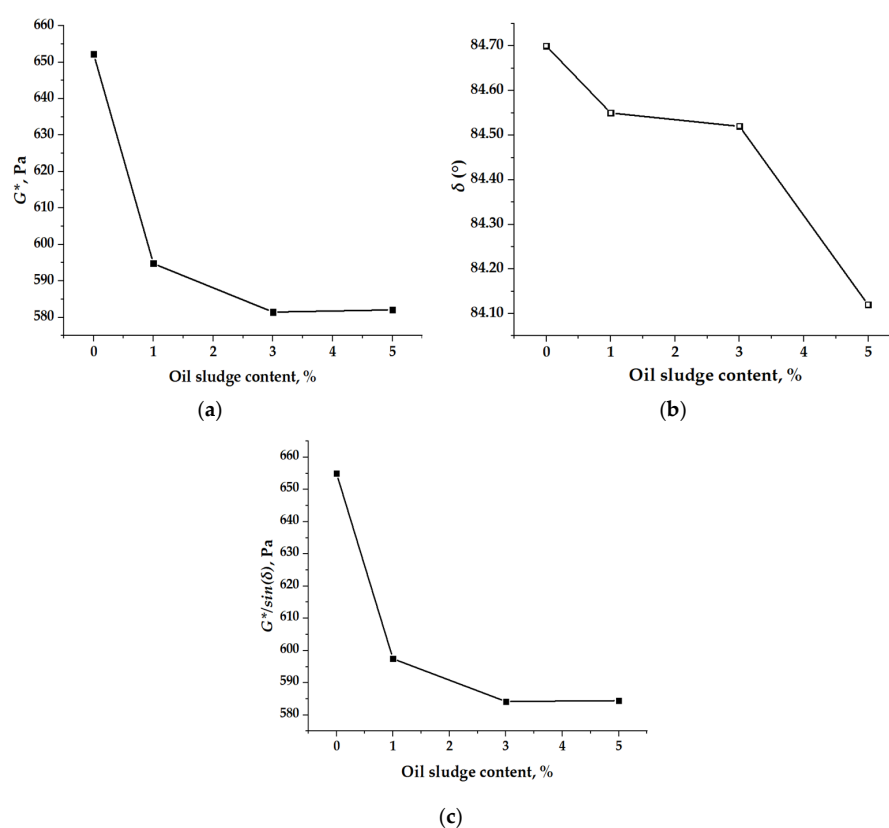


Figure 1. Variation in rheological parameters {(a) G , (b) δ , and (c) $G/\sin \delta$ } with oil sludge content in aged bitumen.

According to the Superpave performance grading specifications (AASHTO M320: Standard Specification for Performance-Graded Asphalt Binder), the rutting factor ($G^*/\sin \delta$) of RTFOT-aged binders must not be lower than 2.20 kPa at the designated high temperature to ensure sufficient resistance to permanent deformation. In this study, the aged bitumen exhibited a $G^*/\sin \delta$ value of 655 Pa at 66.5 °C, indicating excessive stiffness due to oxidative aging. Upon modification with 1–5% oil sludge, $G^*/\sin \delta$ values decreased to 597.5, 584.2, and 584.5 Pa, respectively, at elevated test temperatures (71.4–71.6 °C). Although these values remain above the Superpave threshold, their gradual decline confirms the partial restoration of rheological balance and improved high-temperature workability. These

results suggest that oil sludge contributes to enhanced fatigue tolerance and reduced rutting susceptibility, aligning with the performance criteria outlined in AASHTO M320.

3.3. Analysis of the Impact of Oil Sludge on the Chemical Composition and Structure of Bitumen

One of the most important tools for recording changes in the chemical composition of a bitumen sample is considered to be IR Fourier spectrum (FTIR). The FTIR spectrum of aged bitumen is shown in Figure 2.

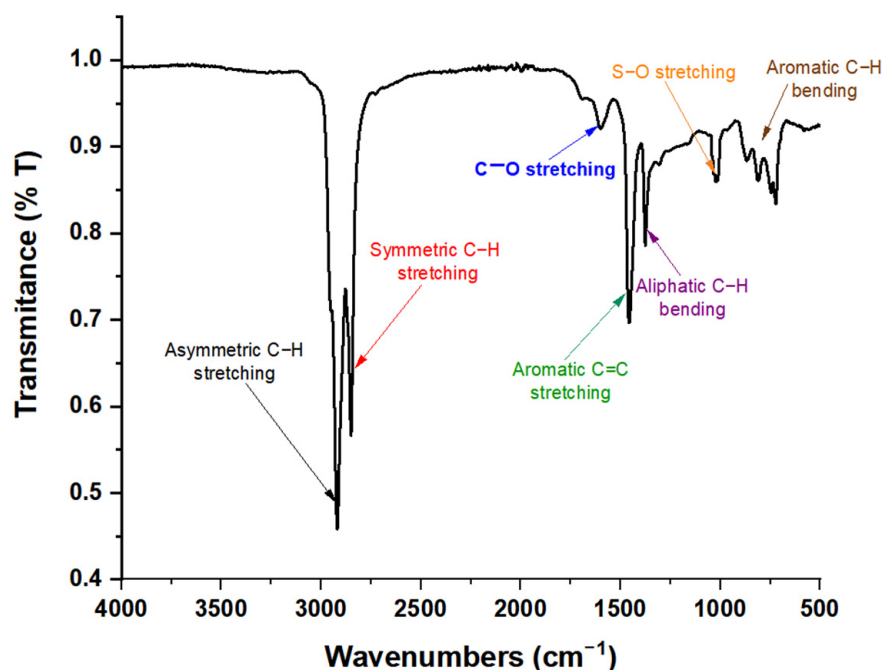


Figure 2. FTIR spectrum of aged bitumen.

The main chemical functional groups for determining the class of bitumen binders are considered to be vibrations of aliphatic and aromatic bonds at wave numbers of about $3000\text{--}2880\text{ cm}^{-1}$, $1350\text{--}1500\text{ cm}^{-1}$, and $750\text{--}900\text{ cm}^{-1}$ [38]. The authors of the work note that the most important are the bands in the range of $1800\text{--}900\text{ cm}^{-1}$, which show chemical changes in functional groups based on heteroatoms. At the same time, in studies [39], the main signs of bitumen oxidation and the confirmation of the process of its aging have been directly associated with the formation of carbonyl (at 1700 cm^{-1}) and sulfoxide (at 1030 cm^{-1}) groups and aromatic groups. A number of scientists [38,40] confirm the successful use of peaks corresponding to sulfoxides and carbonyls to assess the degree of aging and their correlation with the physical and rheological properties of bitumen. At the same time, studies [41] have shown that changes in aromaticity depending on the source of bitumen and its state of aging are not convincing enough, since the evolution of aromatic bands with age is not always obvious, and therefore this parameter is less often used as an indicator of aging, despite the general recognition of an increase in aromaticity [42].

The FTIR spectra of aged bitumen showed the presence of absorption bands corresponding to the sulfoxide bond ($\text{S}=\text{O}$), detected at 1030 cm^{-1} . According to Figure 3, adding oil sludge to aged bitumen leads to a decrease in the sulfoxide bond values. To support this observation, a quantitative FTIR analysis was conducted by calculating the normalized absorbance ratios for the sulfoxide band ($\sim 1030\text{ cm}^{-1}$), using the aliphatic C-H deformation band at 1456 cm^{-1} as an internal reference.

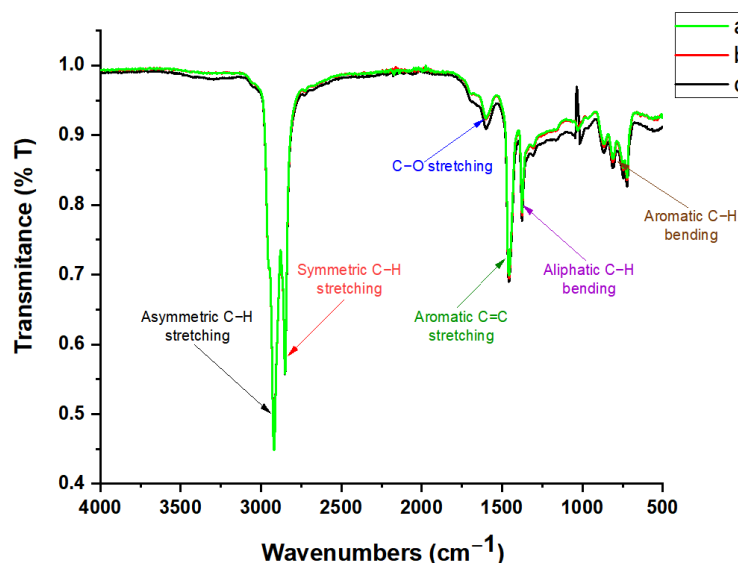


Figure 3. FTIR spectra of aged bitumen samples after the introduction of oil sludge into the aged bitumen in the amount of 1% (a), 3% (b), and 5% (c).

As summarized in Table 4, the sulfoxide index (A_{1030}/A_{1456}) decreased with increasing oil sludge dosage (from 0.395 for aged bitumen to 0.259 for 5% OS), indicating a gradual reduction in sulfoxide group content. The carbonyl band at 1700 cm^{-1} , while present, exhibited only minor changes and was therefore excluded from the quantitative comparison at this stage. In [36], the decrease in sulfoxide and carbonyl bonds is associated with an increase in the ratio of maltenes to asphaltenes. It is important to point out that the symmetric and antisymmetric stretching bands of CH_2 and CH_3 , occurring in the $2600\text{--}3000\text{ cm}^{-1}$ region, are practically unchanged. Since the relative intensities of these contributions can be considered as indicators of the lateral packing of nearby alkyl chains in organic molecules [43], the unchanged intensity profile in this region suggests that no evident change in the short-range interactions of alkyls chains (mostly located within the maltene phase) takes place.

Table 4. Sulfoxide index of aged and rejuvenated bitumen.

Sample	A_{1030}/A_{1456}
Aged bitumen	0.395
1% OS	0.352
3% OS	0.265
5% OS	0.259

The FTIR spectroscopy results confirmed the presence of oxygen-containing functional groups, such as carbonyl and sulfoxide, in the structure of RTFOT-aged bitumen, indicating oxidation and molecular degradation, while the maltenic alkylic region remained largely unaffected. These findings are supported by the SEM images shown in Figure 4. The aged bitumen (Figure 4a) exhibits a rough, irregular surface with microcracks and phase separation, reflecting the destruction of the colloidal structure due to oxidative aging. According to [44], bitumen-rich domains typically appear darker and more compact in SEM micrographs, but these features are largely diminished in aged samples. In contrast, Figure 4b–d depict the morphology of aged bitumen rejuvenated with 1%, 3%, and 5% oil sludge, respectively. With increasing sludge content, the surface becomes progressively smoother and more homogeneous. Notably, at 5% sludge (Figure 4d), the structure shows restored continuity and compactness, indicating improved compatibility and partial reconstruction

of the dispersed binder matrix. These observations confirm the microstructural healing effect of oil sludge, complementing the chemical evidence from FTIR and supporting the assumption that the sludge does not form a separate phase but is integrated at the colloidal or molecular level—similarly to the behavior observed by Demchuk et al. [45], where phenol–cresol–formaldehyde resin demonstrated uniform morphology without distinct polymer-rich domains.

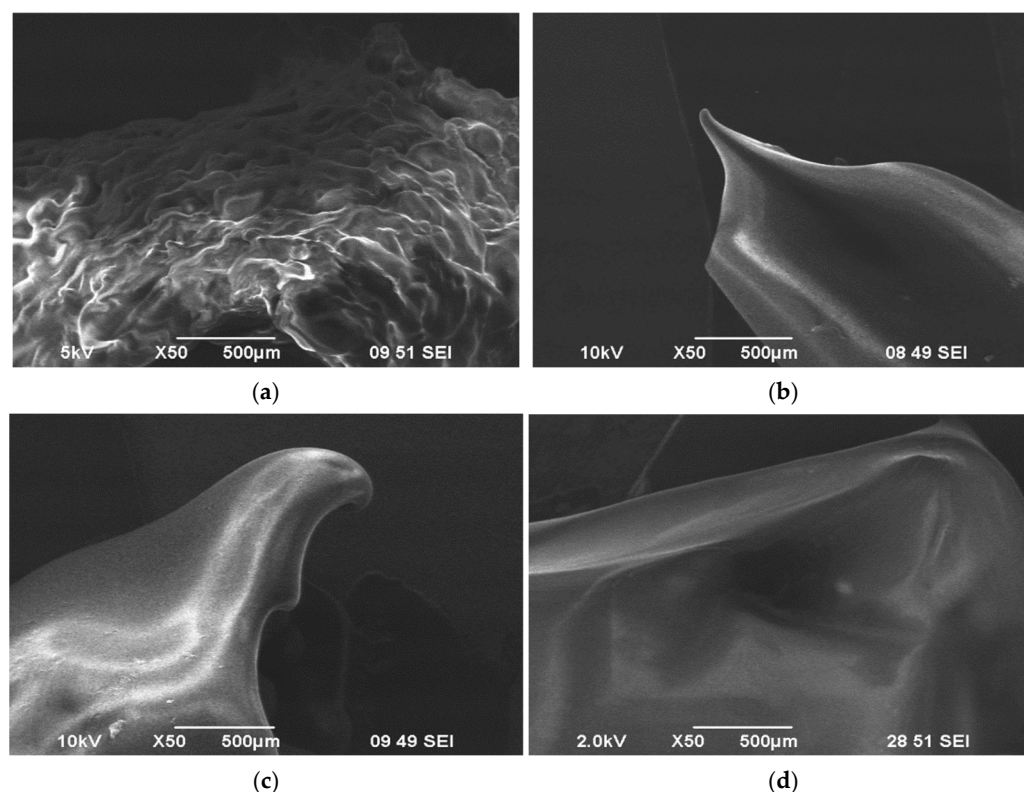


Figure 4. SEM images of aged bitumen (a) and after the introduction of oil sludge in the amount of 1% (b), 3% (c), and 5% (d). Images were acquired using accelerating voltages of 5 kV (a), 10 kV (b,c), and 2.0 kV (d), selected to optimize contrast and minimize surface charging based on sample composition and morphology. Standard post-processing (brightness/contrast adjustment) was applied uniformly across images without altering structural features.

Figure 5 presents the thermogravimetric curves of aged and rejuvenated bitumen samples.

Notably, no significant mass loss was observed up to 200 °C, confirming the thermal stability of the samples under typical conditions of hot mixing and rejuvenation, which are usually conducted at temperatures below 160 °C. In the range of 200–400 °C, a gradual mass loss occurred, associated with the volatilization of maltenes and light aromatic compounds.

The most intensive decomposition was observed between 450 and 500 °C, corresponding to the thermal degradation of heavier components such as resins and asphaltenes. Beyond 500 °C, the remaining mass mainly consisted of thermally stable substances, including both heavy organic residues and inorganic constituents introduced by the oil sludge, such as silicates, metal oxides, and sulfates.

The sample containing 5% oil sludge exhibited more intensive thermal degradation, with a total mass loss of 90.84% and a significantly lower residual mass (9.16%) compared to the aged bitumen (19.25%). This considerable reduction suggests a decrease in thermally stable asphaltene structures due to the disaggregation and partial dissolution of asphaltene clusters by aromatic hydrocarbons present in the sludge.

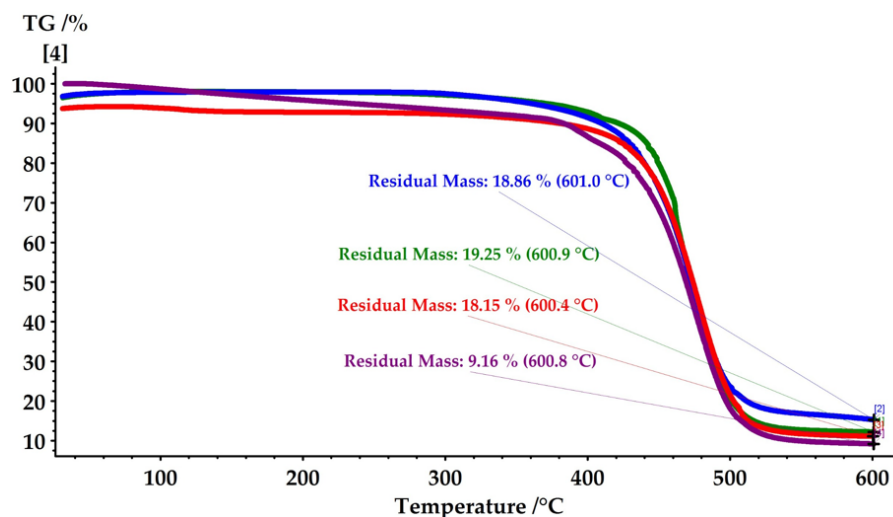


Figure 5. TGA results for aged bitumen (1) and bitumen rejuvenated with oil sludge at 1% (2), 3% (3), and 5% (4) dosages.

While a minor contribution of inorganic components to the final residue is acknowledged, the overall trend clearly indicates that the dominant mechanism is the suppression of coke-forming structures, resulting in a binder that is less rigid and more thermally responsive. These findings suggest that oil sludge contributes not only to softening but also to the molecular-level rejuvenation of aged bitumen, confirming its role as an active restorative agent rather than a simple fluxing additive [46,47].

4. Discussion

While previous studies have explored oil sludge in asphalt mixtures, this research is the first to systematically examine its rejuvenating effect on aged bitumen at the molecular and microstructural levels. By combining rheological, spectroscopic, morphological, and thermogravimetric analyses, the study demonstrates that oil sludge not only enhances flexibility and thermal stability but also alters the colloidal balance of the binder. This multiscale insight into its rejuvenating mechanism underscores the potential of oil sludge as a sustainable alternative to conventional rejuvenators.

The rejuvenating mechanism of oil sludge in aged bitumen systems is primarily attributed to its content of maltenic fractions, including light aromatic and resinous components, which facilitate the redistribution of the colloidal structure of bitumen. These components enhance the solubility and dispersion of asphaltenes and restore the balance between solid and liquid phases within the binder. According to the asphaltene colloidal theory, bitumen is viewed as a micellar system where asphaltenes are stabilized by surrounding aromatics and resins [48,49]. Aging disrupts this equilibrium by depleting stabilizing aromatics, which leads to the formation of dense, fractal-like asphaltene aggregates, as confirmed by SAXS and SARA studies [50]. Rejuvenation with oil sludge reintroduces these key fractions, promoting the fragmentation and redispersion of aged asphaltene clusters.

This trend is quantitatively confirmed by SARA fractionation results. In our previous study [31], the asphaltene-to-maltene (A/M) ratio in virgin bitumen was 0.355. Upon treatment with 1%, 3%, and 5% oil sludge, this ratio progressively decreased to 0.320, 0.280, and 0.257, respectively. Such a shift suggests a partial reversal of oxidative aging and restoration of the colloidal balance. Similar observations were reported by Hu et al. [50], where rejuvenation narrowed the size distribution of asphaltene aggregates and increased the aromatic/resin content, improving colloidal stability.

FTIR spectra revealed a reduction in sulfoxide group intensities with increasing oil sludge content, suggesting partial reversal of oxidation processes. This aligns with the concept that maltenes from oil sludge dilute oxidized polar components, thus increasing the maltene-to-asphaltene ratio. SEM micrographs confirmed the structural homogenization of the binder, indicating uniform dispersion and effective interaction between oil sludge and the aged matrix.

Asphaltenes are known for their rigid, polar, and aggregation-prone molecular structure, which significantly contributes to increased stiffness and loss of flexibility in aged bitumen. Therefore, the observed reduction in their relative content provides compelling evidence of colloidal rebalancing and structural rejuvenation of the binder [46]. This mechanism is schematically illustrated in Figure 6, which visually demonstrates the transition from an aged, asphaltene-rich structure to a rejuvenated and more homogeneous bituminous matrix with improved dispersion, elasticity, and colloidal stability.

In the figure, dark grey clusters represent aggregated asphaltenes; blue circles correspond to maltenes; black curly lines indicate oxidative fragments; green elements show light aromatics and resins derived from oil sludge.

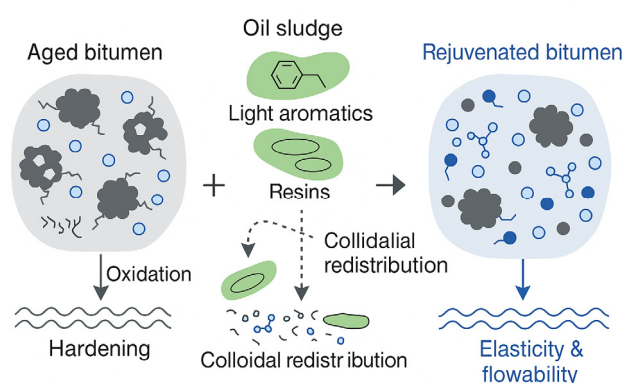


Figure 6. Schematic representation of the rejuvenating mechanism of oil sludge in aged bitumen.

Thermogravimetric analysis (TGA) further supported the mechanism by showing a decrease in residual mass with the addition of oil sludge, especially at a 5% concentration. This may imply a disaggregation or partial dissolution of thermally stable asphaltenes and other high-molecular-weight structures, improving the thermal responsiveness and plasticity of the bitumen.

Collectively, these findings demonstrate that oil sludge functions as more than a mere softening agent. It acts as a structurally interactive rejuvenator, capable of partially reversing the aging process through multiscale restoration of chemical, physical, and rheological properties. This study not only provides mechanistic insight but also underscores the potential of oil sludge as a sustainable and technically effective alternative to conventional rejuvenators.

5. Conclusions

This study has demonstrated that oil sludge can be effectively used as a rejuvenator for aged bitumen. The incorporation of 1–5% oil sludge led to increased penetration values (from 60 to 71 $\times$ 0.1 mm) and softening points (from 55 $^{\circ}$ C to 72 $^{\circ}$ C), indicating enhanced pliability and thermal performance. FTIR spectroscopy revealed a decrease in oxidation-related groups (notably sulfoxides), SEM showed improved surface uniformity, and TGA confirmed reduced thermal residue. Additionally, the asphaltene-to-maltene ratio dropped from 0.355 in aged bitumen to 0.320, 0.280, and 0.257, suggesting improved colloidal balance and partial structural recovery.

Beyond technical performance, this approach offers environmental benefits by repurposing hazardous oil refinery waste into a valuable material for road construction. The findings support the integration of oil sludge into sustainable pavement technologies. Future research will focus on the long-term durability of treated binders under field conditions and further elucidation of molecular interactions using advanced analytical tools such as GC-MS and NMR.

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