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Mechanism and Investment Analysis of Recycling Gasoline Solvent with Mineral Powder for Asphalt Cleaning

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Abstract: Gasoline is widely used as a powerful organic solvent to remove asphalt residuals in road engineering applications; however, it is also known as a non-renewable fuel resource. This research aims to employ mineral powder to mitigate the consumption of gasoline in asphalt binder cleaning process, and evaluate its mechanism, environmental and economic benefits. Based on X-Ray Diffraction (XRD) spectra, X-Ray Fluorescence (XRF) spectra and Atomic Absorption Spectroscopy (AAS) detection, the microstructure and composition of mineral powder was investigated after adsorbing asphalt components from gasoline solution. Fourier Transform Infrared spectra (FTIR) were used to calculate the adsorption efficiency. Moreover, the assessments of environmental and economic impacts of investigated approach were evaluated quantitatively. The results indicated that the interactions between mineral powder and asphalt binder have not changed the crystal phase, and the rough interface of mineral powder could adsorb approximately 4% asphalt binder from the gasoline solution according to the detection of Zn. The comparative analysis, regarding cost-effectiveness and environmental impacts, demonstrated that once 1 kg asphalt adsorbed by mineral powder, recycling gasoline with mineral powder, could reduce costs by 80% (CNY 57.10), energy use by 97% (352.88 MJ) and equivalent CO_2 emission by 93% (23.95 kg).

Keywords: gasoline consumption; mineral powder; asphalt binder; solvent; environmental impacts

1. Introduction

With the massive demand for infrastructural developments, the 21st century is witnessing a significant increase in asphalt pavement construction globally [1]. It is expected that 25 million kilometers of new pavements are going to be built before 2050 [2], more than 90% of which will be in developing countries [3]. Furthermore, according to the panel data from European Union, asphalt pavement construction has a prominent position for huge financial investments and resource consumption [4]. Consequently, researchers are increasingly focusing on environment-friendly and sustainable asphalt pavement constructions, especially in reducing both aggregate and fuel depletion [5], but ignoring the considerations involving the associated experimental operations and laboratory management.

From the perspective of laboratory management, associated administrators constantly highlight the operation safety and implementation disciplines in engineering education. Li assessed the risks in engineering laboratories by accident statistics and questionnaires, and proposing that people need to pay attention to the storage of inflammable and explosive substances and common safety training [6].

Gao advocated the workable strategies and practical considerations for vigorous developments of engineering laboratories under industry-oriented, world-oriented and future-oriented backgrounds [7]. For practical functions of laboratories, finishing the experiment should be set on a priority right; however, in the current context of sustainable development, it is simultaneously important to deal with experimental wastes environmentally. For example, the technicians in asphalt-related experiments not only take efforts to ensure experiment completion, but treat residuals also involved multiple works, such as cleaning and wiping out extra asphalt binder from experimental equipment.

Today, fossil fuels such as gasoline supply over 80% of the energy all over the world [8], but their reserves are expected to last for a limited period based on the current consumption rate [9]. Gasoline, besides being used as an energy supplier, is also commonly adopted in asphalt research laboratories as an organic solvent to remove asphalt binder from experimental devices, owing to its efficient and non-toxic characteristics [10]. Gasoline and bitumen are extracted from the petroleum crude oil as a distilled and residual product, respectively [11]. Therefore, asphalt binder displays good solubility in gasoline based on the 'like dissolves like' principle [12]. Gasoline can be used as an efficient organic solvent since it can attach to asphalt binder particles [13,14]. As for the chemical structure, gasoline is mainly composed of relatively lightweight hydrocarbons (C4–C12 molecules) [15], but the detailed content of different compositions usually varies based on the refining feed, operations and octane rating [16]. Considering the aforementioned trend for asphalt pavement construction, it is foreseeable that the consumption of gasoline would steadily become more significant.

Gasoline is commonly used worldwide in universities, enterprises and construction sites to remove the residuals of asphalt binder on laboratory equipment [17]. Furthermore, many other organic solvents are also available for this purpose, but most of them have heavily adverse effects to human health [18,19]. For example, carbon disulfide has a good asphalt dissolving effect, but constant exposure to this substance could lead to ischemic heart disease mortality [20]. Tetrachloromethane is also effective, but it is characterized by high cost, and negative impacts on human health are considerable [21]. Comparatively speaking, gasoline solvent is an appropriate solvent for asphalt cleaning with an acceptable price and low toxicity [22]. Nevertheless, gasoline is a valuable and valued resource in the current energy market. Therefore, recycling gasoline solvent not only saves the investments in purchasing chemical reagents, but also reduces the consumption of nonrenewable resources.

Mineral powder is an easily accessible raw material in asphalt laboratories with a low wholesale price (approximately CNY 450 per ton in China). It is widely used as a filler in asphalt mixture preparation [23], and plays a dominant role in enhancing binder adhesion to aggregates and increasing the stiffness module and stability of the asphalt mixture [24]. The main chemical components of mineral powder are calcium oxide and silicon oxide, and the particle size meets the specification for passing 200 mesh (0.075 mm) sieve. At low temperatures, mineral powder could affect the cohesive strengths of asphalt mortar [25]; at high temperatures, the rutting resistance and fatigue failure of the asphalt mixture would also be influenced by the mineral powder's addition [26].

The composite interaction between asphalt binder and mineral powder enhances the workability of the asphalt mixture. In contrast, this interaction can also be utilized in disposing waste asphalt binder. In this study, a novel approach is proposed to use the mineral powder adsorbing part of the dissolved asphalt binder in gasoline, aiming to recycle the gasoline solvent used in asphalt research institutions. X-Ray Diffraction (XRD) spectra and X-Ray Fluorescence (XRF) spectra were adopted to verify the crystal phase and qualitative composition; after that, Fourier Transform Infrared spectra (FTIR) and Atomic Absorption Spectroscope (AAS) were used to estimate the adsorption efficiency by comparing the peak intensity and content of Zn. Furthermore, a laboratory case study evidenced the annual gasoline usage, and related financial investments and environmental impacts for the proposed method were assessed for China and Norway. Overall, the investigated solution can make a significant difference in saving fossil fuel resources for asphalt disposal.

2. Materials and Methods

In this study, asphalt binder with a penetration grade of Pen 60/80 (Hubei Guochuang Hi-tech Material Co., Ltd., Wuhan, China) was investigated as the substance to be removed, the fundamental properties are listed in Table 1. This bituminous binder is largely employed in heavy traffic roads. The used #92 gasoline was supplied by Sinopec (Sinopec, Beijing, China). The mineral powder adopted was made of limestone with an average size of 48 μm, Table 2 shows its associated physical properties.

Table 1. Fundamental properties of Pen60/80 asphalt binder.

Property	Result	Standard
Specific gravity	1.034	ASTM D70
Penetration at 25 °C (0.1 mm)	63	ASTM D5
Ductility, 5 cm/min, 5 °C (mm)	37	ASTM D113
Softening point (°C)	48	ASTM D36
Apparent viscosity, 135 °C (Pa·s)	0.46	ASTM D4402
Loss on heating (wt.%)	+0.09	ASTM D6

Table 2. Physical properties of mineral powder.

Property	Result	Standard
Apparent density (t/m^3)	>2.701	T 0352-2000
Moisture content (%)	<0.1	T 0103-1993
Hydrophilic coefficient	0.8	T 0353-2000
Plasticity index (%)	3.4	T 0354-2000
Passing rate (0.075 mm sieve, %)	86.7	T 0351-2000

In order to analyze the absorbing strength, the experiment added mineral powder into mixed asphalt and gasoline solution whose sediment was then investigated further. Figure 1 depicts the sample preparation processes. Firstly, 10 g asphalt binder was dissolved in 150 mL gasoline contained in an Erlenmeyer flask, the process was accelerated by shaking the flask for 10 min and letting it rest for 60 min. Successively, 20 g of mineral was added into the solution, which rested for 30 min; afterwards, the sediment was extracted with suction filtration. Due to the complex composition of gasoline and asphalt, the components of both mineral powder and sediment were investigated as research objects for their relatively stable components and structures. Corresponding to the actual situation for asphalt cleaning works, all procedures for sample preparation were implemented under room temperature and normal pressure conditions. Pure mineral powder was labelled as sample A, the sediment from gasoline and mineral powder was labelled as sample B and the sediment from mineral powder, asphalt and gasoline was marked as Sample C.

There are three primary aims for this research: (1) verify the feasibility of using mineral powder to absorb asphalt; (2) explore the absorbance efficiency and mechanism of using mineral powder to absorb asphalt; (3) assess the environmental impacts and financial investment of using mineral powder. In order to address these problems, this study focused on the dissolving process and utilized XRD (BRUKER AXS GMBH, Karlsruhe, Germany) with D8 Advance specification to study samples' crystal compositions [27]. XRF (Malvern Panalytical, Almelo, Netherland) method with Zetium specification was adopted to integrate the contents and compositions of samples [28]. The specification of the FTIR instrument used in the experiment was Nicolet 6700 (Thermo Electron Scientific Instruments, Madison, WI, US), the conducted wavenumber range was 400–4000 cm^{-1} [29]. The AAS employed was CONTRAA-700 (Analytik Jena AG, Thuringia, Germany), and the samples were tested at 0.3 MPa under an argon atmosphere [30]. Additionally, a case study from Wuhan University of Technology was implemented to assess the cost and environmental impacts. Consulting the data from the China Energy

Statistical Yearbook 2013 [31] and published literature [32–36], the extraction processes of gasoline and mineral powder were considered, and their comprehensive benefits were calculated and compared.

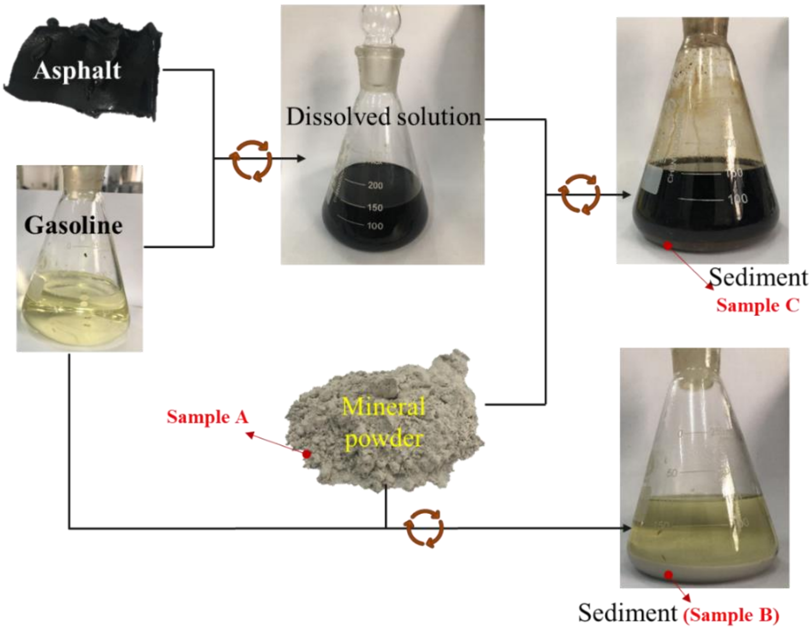

Figure 1. Schematic diagram of sample preparation.

3. Results and Discussion

3.1. Mechanism Performance

3.1.1. Crystal Phase of Absorbed Mineral Powder

In order to explore the changes after adsorbing the asphalt binder, the crystal phase was considered in the powder samples analyzed with XRD. Figure 2 shows the XRD spectra for three samples: A, B and C. The obtained peak (104°) belongs to calcite $Ca_6C_6O_{18}$. All the specimens had a peak in correspondence of the same degree value, as well as a similar flow trend. This means the crystal phase of applied mineral powder is calcite $Ca_6C_6O_{18}$. Moreover, the same regularities of three XRD spectra in Figure 2 indicate that after adding in pure gasoline (sample B) and adsorbing the asphalt binder from the solution (sample C), the mineral powder did not comprise any changes regarding the crystal phase.

3.1.2. Chemical Compositions of Absorbed Mineral Powder

The composition differences among the three samples were integrated by an XRF test. The detected substances and associated contents are listed in Table 3: CaO, SiO_2 and Al_2O_3 accounted for nearly 90% of the whole composition. The limestone mineral composition fulfils the standard values according to previous literatures [35,37]. Although the three specimens have similar compounds content distributions, an evident difference can be found in Table 2 that only sample C contains a small percentage of ZnO due to the distillation treatments and following transport operations [36]; on the other hand, this demonstrates that the applied mineral powder and gasoline were not related to ZnO.

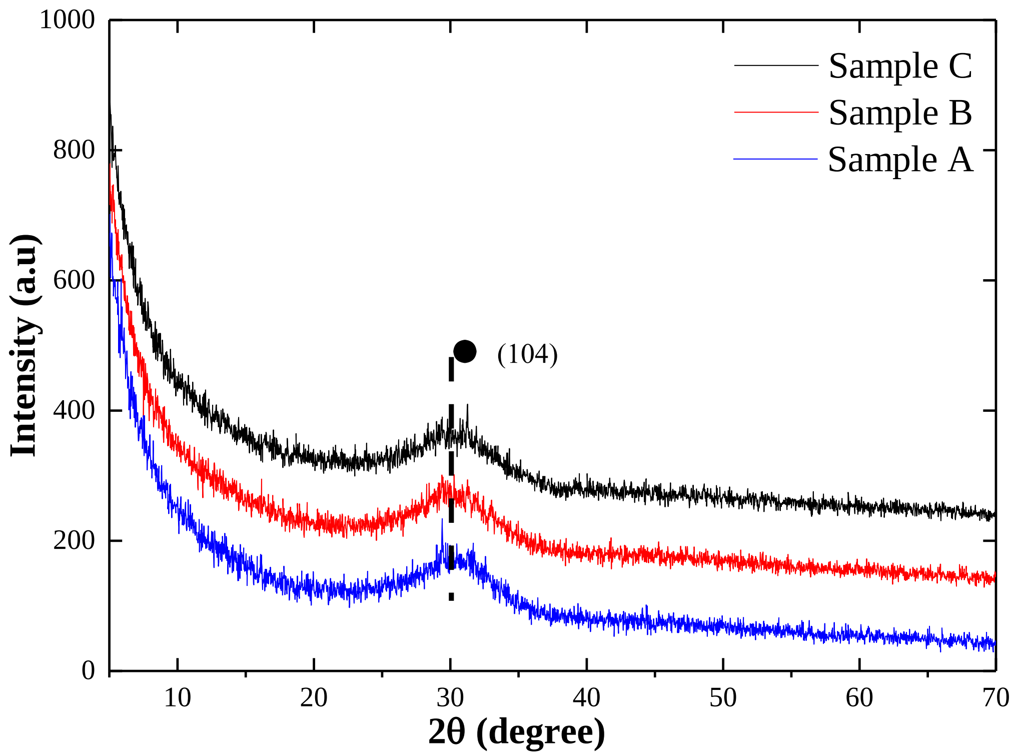

Figure 2. XRD spectra for three investigated samples.

Table 3. Composition and content for the three investigated samples.

Composition	Content (%)		
	Sample A	**Sample B**	**Sample C**
Na_2O	0.325	0.357	0.392
MgO	7.835	8.488	8.413
Al_2O_3	15.749	16.755	16.218
SiO_2	32.612	33.651	32.987
SO_3	2.373	2.315	2.674
K_2O	0.445	0.423	0.442
CaO	39.237	36.788	37.600
TiO_2	0.758	0.615	0.638
MnO	0.133	0.141	0.155
Fe_2O_3	0.241	0.218	0.194
SrO	0.102	0.085	0.094
Y_2O_3	0.012	0.010	0.012
ZrO_2	0.049	0.041	0.044
BaO	0.103	0.114	0.133
ZnO	0	0	0.005

3.1.3. Qualitative Analysis by Functional Groups of Mineral Powder

Given the small amount of ZnO detected by XRF, the study applied AAS to obtain Zn contents in a pure asphalt binder and sample C, according to the standard GB/T 15337-2008. Based on the atomic vapor for a given characteristic line, AAS analysis achieved qualitative and quantitative analysis by assessing the degree of attenuation [38]. The results show that the concentration of Zn in pure a asphalt binder and sample C were 2.75 mg/kg and 48.36 mg/kg, respectively; this approximately corresponds to 0.005% for sample C and agreed well with the XRF result. Therefore, the AAS test also proves that the mineral powder adsorbed part of asphalt binder from the solution.

Afterwards, the study attempted to calculate the adsorption ratio by comparing the peak intensity in FTIR spectra for a pure asphalt binder and the three samples (Figure 3). It can be found that, in the wavenumber range 2750–3000 cm^{-1}, both sample C and 70# asphalt binder clearly displayed two peaks (2919–2845 cm^{-1}), while no absorption peak appeared in sample A or sample B (Table 4) [39]. Based on the existing literature concerning gasoline [40], mineral powder [41] and asphalt binder [42], the two peaks registered in the range 2750–3000 cm^{-1} prove the mineral powder's adsorption capacity.

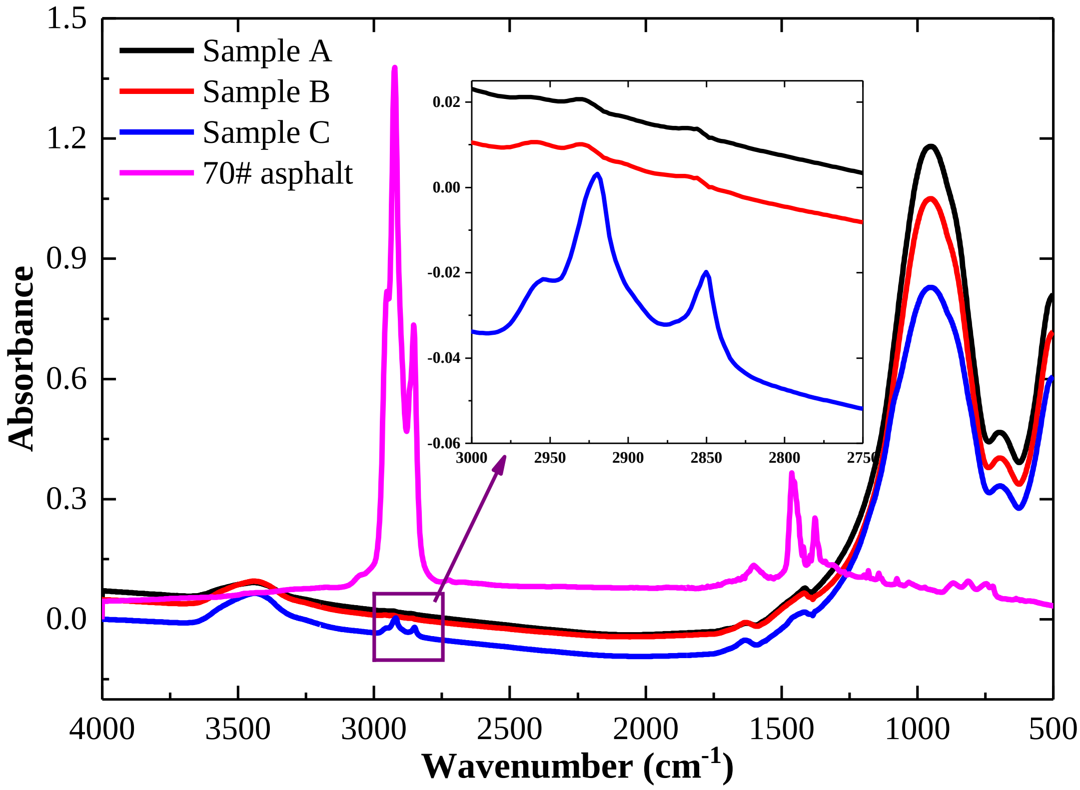

Figure 3. FTIR spectra for pure asphalt binder and the three samples.

Table 4. Peaks and assignments for the FTIR wavenumber range 2750–3000 cm^{-1}.

Wavenumber (cm^{-1})	Assignment	Origin of the Chemical Structure
2919	Methylene C–H anti-symmetry stretching	Methylene units
2845	Methyl C–H symmetry stretching	Methylene units

3.1.4. Adsorption Efficiency of Mineral Powder

The peak areas were calculated (OMNIC software) for sample C and asphalt binder in the wavenumber range 2750–3000 cm^{-1} (Figure 4), the results are shown in detail in Table 5. The intensity of the peak in 2919 cm^{-1} was connected to the biggest adsorption efficiency of 4.1%, while the peaks in correspondence of 2845 cm^{-1} and the range 2750–3000 cm^{-1} were 3.8%. Based on the peak areas for selected wavenumbers, the adoption ratio for mineral powder can be estimated as equal to 4%.

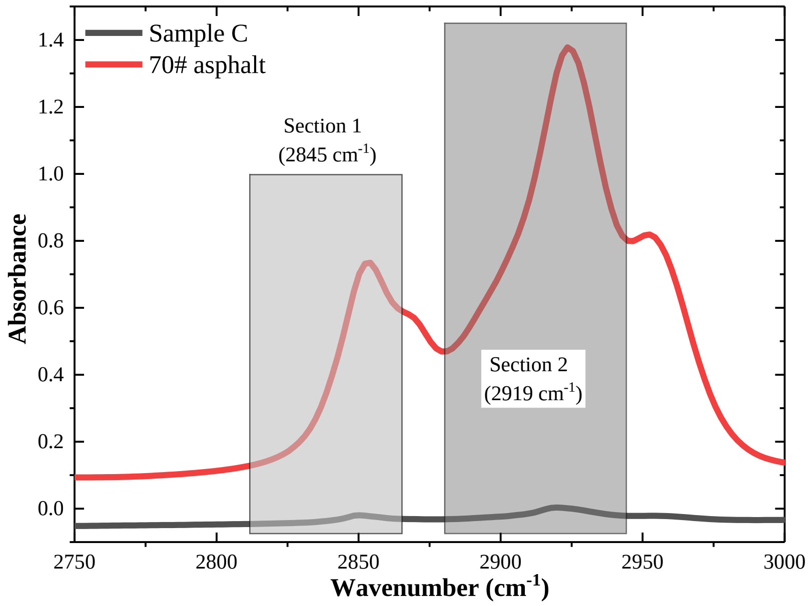

Figure 4. Selected sections for calculating peak areas.

Table 5. Peak areas for selected wavenumbers.

	Peak Wavenumber (cm^{-1}) and Areas		
	Section 1 (2919 cm^{-1})	**Section 2 (2845 cm^{-1})**	**2750–3000 cm^{-1}**
70# asphalt	17.385	7.845	62.362
Sample C	0.710	0.295	2.369
Percentage (%)	4.084	3.760	3.799

Considering the outcomes from FTIR and XRF analyses, no formation of new functional groups was detected, proving that the adsorption exerted by the mineral powder only involves physical processes and no chemical reactions. Previous research revealed that the surface of the mineral powder was quite rough, thus improving the adhesive effect to asphalt binder [43,44]. As shown in Figure 5, the adsorption mechanism for the mineral powder can be explained considering three phases. The addition of mineral powder in the solution promotes contact between asphalt compounds and the rough surface (a); then, part of the dissolved compounds sticks to the mineral powder's surface (b) and forms the sediment by constant accumulation (c). Therefore, the achievement of adsorption should be attributed to the contact between the mineral powder and asphalt binder phase, promoted by the rough interface of the former. These series of reactions are physical activities, which would not affect any compositions or ingredients of the original gasoline.

The dissolved asphalt binder did not reach the maximum solubility of gasoline in this experiment [45]. Increasing the amount of dissolved asphalt in the gasoline solution also increased the probability of mineral and asphalt components creating contacts. The practical situation occurring in asphalt laboratories usually corresponds to supersaturated solutions of massive waste asphalt binder handled by limited gasoline solvent. Therefore, it can be stated that a higher content of dissolved asphalt could entail an adsorption ratio that is better than the one investigated (4%).

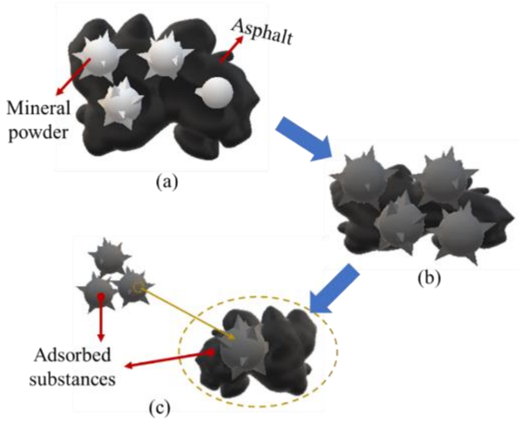

Figure 5. Adsorption mechanism for mineral powder: (**a**) adsorption; (**b**) adhesion; (**c**) sediment.

3.2. Cost and Environmental Impact Analyses

In order to achieve a real estimation of gasoline consumption in laboratories, the annual use of this resource in Wuhan University of Technology was taken into consideration. The total amount corresponded to 740 L, (approximately CNY 5200), 95% of which was used for asphalt cleaning related to experimental devices. Assuming that the gasoline consumption rate of a common car is 9.2 L/km, the annual laboratory usage corresponded to an 80-km drive [32].

Furthermore, the research referred to the data available in the literature concerning both the energy consumption for mineral aggregates (25.59–32.18 MJ/ton) [33] and for gasoline production (3.04–3.90 MJ/L) [34]. According to the aforementioned chemical detections and interpretive mechanisms, 15 g asphalt binder was dissolved in 150 mL gasoline, 4% of which was adsorbed by 20 g mineral powder at room temperature. Therefore, under experimental conditions, 20 g mineral powder adsorbed 4% and/or 0.6 g asphalt binder. Specifically, removing 1 kg asphalt would consume 10 L gasoline, while adsorbing 1 kg asphalt from solution would demand 33.33 kg mineral powder.

3.2.1. Economic Evaluations

The study considered the market prices for 92# gasoline (7.12 CNY/L) and mineral powder (425 CNY/t) in China according to the Numbeo database on 8 February 2020 [46]. Based on Equation (1), the study estimated the savings connected to the application of mineral powder to recycle gasoline in the laboratory. The cost of the gasoline necessary to dissolve 1 kg of bituminous binder is CNY 71.20. For the same purpose, the cost of the added mineral powder is 14.10 CNY; as a result, the difference is 57.10 CNY/L.

$$C = p_g * v_g - p_m * m_m \tag{1}$$

where C represents the saving cost(CNY), p_g is the reference unit price for gasoline (CNY/L), p_m is the unit price for mineral powder (CNY/kg), v_g is the volume of gasoline (l) and m_m is the mass of mineral powder (kg).

3.2.2. Environmental Impacts

Given the growing concerns regarding sustainability, recycling gasoline in the laboratory would not only reduce economic investments, but also entail a positive effect regarding environmental impacts. This study investigated the energy consumption and equivalent CO_2 emission of employing mineral powder to adsorb asphalt binder. According to the China Energy Statistical Yearbook 2013 [31], 1 L of gasoline could release 31.75 MJ energy and 2.21 kg equivalent CO_2, while its production would consume 4.54 MJ/L energy and emit 0.37 kg/L equivalent CO_2 [47]. On the other hand, the extraction of 1 kg limestone from a quarry would consume 80.00 kJ energy and emit 5.95 g equivalent CO_2 [48], and the following crushing and grinding operations need 220.60 kJ/kg energy and emit 59.63 g equivalent CO_2 [49].

Equations (2) and (3) are applied to integrate the saved energy and reduced equivalent CO_2. According to the data from referenced literatures, the removal of 1 kg of asphalt binder using mineral powder would cause 10.02 MJ energy consumption and 1.85 kg equivalent CO_2 emission, while dissolving asphalt binder by gasoline directly would result in 362.90 MJ energy consumption and 25.80 kg equivalent CO_2 emission.

$$E = (E_g + E_p) * v_g - E_m * m_m \tag{2}$$

$$G = (G_g + G_p) * v_g - G_m * m_m \tag{3}$$

where, E and G represent the saved energy (MJ) and reduced equivalent CO_2 (kg), respectively; E_g and E_p stand for the energy consumption (MJ/L) generated by gasoline combustion and production, respectively; G_g and G_p are equivalent CO_2 (kg/L) generated by gasoline combustion and production, respectively; E_m and G_m mean the energy consumption (MJ/kg) and CO_2 emission (kg/kg) during manufacturing mineral powder, respectively; vg represents the volume of gasoline (l) and mm is the mass of mineral powder (kg).

3.2.3. Comparative Interpretations

Combining the cost and the environmental impacts related to gasoline and mineral powder, Table 6 depict the results connected to the removal of 1 kg asphalt binder, after that, Figure 6 shows the cumulative percentage distribution for these two approaches. For the case study, treating 1 kg of bituminous binder merely with gasoline required CNY 71.20, caused 362.90 MJ of energy consumption and 25.80 kg equivalent CO_2 emission. On the other hand, using mineral powder to adsorb asphalt reduced the cost by 80%, energy consumption by 97% and equivalent CO_2 emission by 93%. However, the accessible asphalt binder adsorbed by mineral powder accounts for a small part in the dissolved solution. After adsorbing 1 kg asphalt binder with mineral powder, approximately 25 kg asphalt binder should be dissolved in the gasoline solvent.

Table 6. Cost and environmental impacts of cleaning 1 kg asphalt with 2 approaches.

	Cost (CNY)	**Energy Consumption (MJ)**	**Equivalent CO_2 EMISSION (kg)**
Gasoline	71.20	362.90	25.80
Mineral powder	14.10	10.02	1.85
Reducing percentage	80%	97%	93%

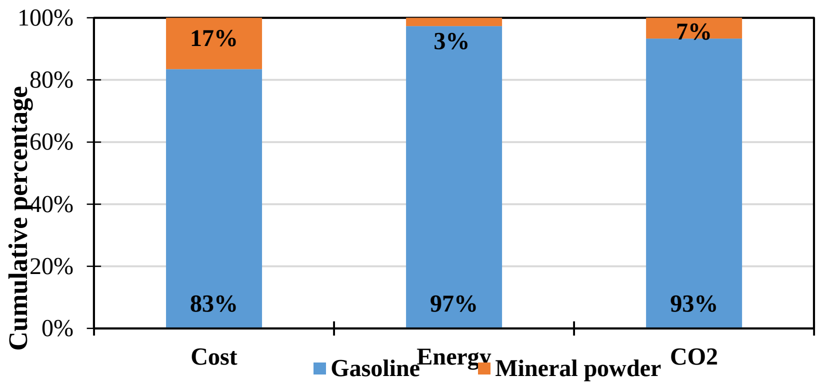

Figure 6. Cumulative percentage distribution for the investigated approaches according to cost and environmental impacts.

4. Conclusions

Gasoline is a valued and valuable energy source in the current energy market globally, and this resource is also frequently used as a solvent to remove bituminous binder in asphalt laboratories. This study investigated the use of mineral powder to adsorb part of the asphalt binder. In order to investigate the adsorption performance, X-ray Diffraction (XRD), X-ray Fluorescence (XRF), Atomic Absorption Spectroscopy (AAS) and Fourier Transform Infrared (FTIR) were employed to analyze the compositions of adsorbed sediments. Eventually, a case study compared the investigated solutions for bituminous binder removal according to costs and environmental impacts. The following conclusions can be drawn:

1. Mineral powder mixed with gasoline is an effective solution to remove bituminous binder commonly found in experimental devices in asphalt laboratories. The mechanism can be explained as a physical interaction between the mineral powder and asphalt binder.
2. Almost 4% of the binder can be adsorbed by the gasoline solution, thus proving the beneficial effect generated by the use of mineral powder.
3. The investigated case suggests that removing 1 kg asphalt binder with pure gasoline costs CNY 71.20, consumes 362.90 MJ energy and emits 25.80 kg equivalent $CO_{2.}$, and that adding a mineral powder can reduce these quantities by 80%, 97% and 93%, respectively.
4. Although the rough surface of the mineral powder determines the physical adsorption strength, the low adsorption rate limits the benefits in practical application. Future research should focus on the improvement of the adsorption rate.

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Data Availability: The data that support the findings of this study are available within the article.

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