Research Article

Comparison of Bitumen Asbuton Diluents: Kerosene vs. Diesel Oil

Zakijah Irfin*, Achmad Chumaidi and Dwina Moentamaria
Department of Chemical Engineering, State of Polytechnic Malang, East Java, Indonesia

Heri Septya Kusuma
Department of Chemical Engineering, Faculty of Industrial Technology, Universitas Pembangunan Nasional "Veteran" Yogyakarta, Indonesia

* Corresponding author. E-mail: zakijah.irfin@polinema.ac.id DOI: 10.14416/j.asep.2022.02.012
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Abstract
Asbuton (aspal Buton) is a vast natural asphalt reserve owned by Indonesia and located on the island of Buton. This substance was composed of bitumen and minerals. Bitumen is the major ingredient used as an adhesive in the construction of roads. Because Asbuton bitumen is tightly bonded to the mineral calcite, a suitable diluent is required for the process of releasing the bitumen. The purpose of this research is to compare the recovery percentage of Asbuton bitumen using kerosene or diesel as a diluent. The choice of kerosene and diesel diluents in this investigation was based on Fourier Transform Infra Red (FTIR) analysis, which revealed the proximity of the kerosene and diesel functional groups to Asbuton bitumen. The first stage is to conduct a viscosity analysis by dissolving Asbuton bitumen and diluent in a ratio of 30:70, 40:60, 50:50, 60:40, and 70:30 (%, w/w) at temperatures of 30, 40, 50, 60, 70, 80, and 90 °C. According to the results of the experiments, the viscosity of the combination of bitumen Asbuton – diesel oil/kerosene with range 0.5–2 Pas is related to the optimal percentage removal of bitumen in both diluents. This result indicates that the separation process was carried out at a temperature of 50–90 °C, with an Asbuton-diesel oil/kerosene ratio of 50:50 and 40:60 (%, w/w), respectively. The best bitumen separation results were obtained with kerosene as a diluent, with a removal percentage of 76.30% at a temperature of 90 °C and a ratio of 40:60 (%, w/w).

Keywords: Bitumen, Asbuton Kabungka, Kerosene, Diesel oil, Diluent

1 Introduction

The use of asphalt in Indonesia is expected to increase further. According to data from the Directorate General of Construction Development, Ministry of Public Works, and Public Housing, the national demand for asphalt in 2017 was 1.545 million tons. Meanwhile, local asphalt producers, such as Pertamina can only meet a small portion of the national asphalt demand of 344.153 t. The remaining 1.2 million tons were mainly imported from other countries. According to the Ministry of Industry, the volume of asphalt imports in 2021 has reached 900,000 tons, with a foreign exchange value of nearly US$450 million (assuming an asphalt price of US$500 per tonne), or more than Rp6 trillion. Asphalt is made up of 95% aggregate particles (stone, sand and gravel) and 5% bitumen. Bitumen is used to bind aggregates together to form a cohesive mixture. According to a survey conducted by the Ministry of Public Works and Public Housing in 2011, Indonesia has reserves of Asbuton deposits for 663 million tons on Buton Island in total, of which is purified form will be produced more than 150 million tons of bitumen. This amount is enough to meet the country's asphalt needs for up to 125 years. Indonesia will be self-sufficient in asphalt and may even become an exporter of this material.

There are several Asbuton deposit regions, but only the Kabungka mining sites have been mined and are currently in use; other mining areas, such as the Lawele region, are still in the exploring stage and have limited values. Because non-uniform mining products from Asbuton Kabungka provide unsatisfactory asphalting results, pure bitumen must be created by separating Asbuton bitumen from its minerals [1]. Because of the high calcite concentration of Asbuton Kabungka minerals, bitumen is securely bound to the mineral, necessitating the use of a diluent and an appropriate physical technique in the Asbuton bitumen separation process.

Kabungka was used in this study, with an estimated reserve of 60,000,000 tons and a bitumen content of 15.35% (w/w) [2]. The addition of a diluent in the form of diesel oil has been widely studied for Asbuton purification [3], [4], organic solvents [5]–[7] or kerosene [8]–[10]. Aromatic solvents are diluents with a high success rate. Because of their abilities to retain asphaltene molecules in solution, aromatic solvents, such as toluene and xylene have long been recognized as the best solvents for reducing heavy oil/bitumen viscosity. However, due to high operating costs, the use of organic solvents to separate asphalt from Asbuton has been long-abandoned. Meanwhile, the use of diesel oil and kerosene as asphalt solvents from Asbuton has been applied on an industrial scale as well as in research, such as naphtha, condensate, kerosene, or light paraffin, which are more environmentally friendly, cost-effective and can reduce asphaltene deposition problems from heavy oil/asphalt or other problems associated with asphaltene. Asbuton bitumen has a high viscosity due to the asphaltene content. Some researchers mix the diluent in the form of diesel oil or kerosene, then add surfactant and alkaline salt [3]. The preceding statement demonstrates the critical importance of selecting the proper diluent in keeping asphaltenes and other solids in solution during the bitumen separation process from Asbuton.

Kerosene or diesel oil is used in the purification process as a bitumen diluent from Asbuton Kabungka. The Asbuton Kabungka bitumen viscosity value of 15.4 Pa. s at 100 °C necessitates the use of a diluent [11]. According to Hupka *et al.* [12], this viscosity value is included in the oil sand class III category, indicating that a diluent is required to enhance the bitumen recovery percentage. This concept will be used in this study to separate bitumen from Asbuton Kabungka in terms of viscosity, process temperature and recovery bitumen percentage. The addition of diluent in the form of kerosene and diesel oil reduces the viscosity of the bitumen. Reducing bitumen viscosity to 0.5–2 Pa.s at digestion temperature, regardless of oil sand type, will increase the bitumen recovery percentage [12]. Another researcher claimed that by lowering bitumen viscosity by 6 Pa.s, bitumen removal can be increased to more than 80% [13]. It is possible to assert that viscosity plays an important role in the bitumen recovery percentage [14].

Based on the explanation above, this study first determines the viscosity of the Asbuton Kabungka bitumen, which is in the range of 0.5–2 Pa.s, by diluting the bitumen with kerosene or diesel oil in a specific ratio (%, w/w) at a temperature of 40–90 °C. The mixture comparison with temperature conditions that meet the viscosity criteria of 0.5–2 Pa.s will be clarified during the stirring and separating processes of the Kabungka bitumen to obtain the value of the bitumen recovery percentage.

Viscosity, process temperature and diluent type are all key factors that influence the value of the bitumen recovery percentage in general. This research offers updates on the range of viscosity and temperature and the kind of diluent that is acceptable for use in the bitumen separation process, particularly Asbuton, using a simple and straightforward procedure.

2 Methodology

2.1 *Materials*

Asbuton from Kabungka, Southeast Sulawesi, was used as the raw material. The Asbuton that will be used is crushed and sieved through a –20/+40 mesh sieve. SNI 03-3640-1994 was used to characterize the initial content of Asbuton bitumen using the soxhlet extraction method. The solvents used are PERTAMINA commercial kerosene and diesel oil. The initial study of bitumen concentration in Asbuton Kabungka was conducted using technical trichlorethylene (TCE) as the solvent, according to SNI 03-3640-1994.

2.2 *Analysis*

Analysis of the initial content of bitumen in Asbuton was carried out by extraction using Soxhlet and

TCE solvent. Sample (a g) was weighed and placed in a Soxhlet to begin the extraction procedure. This procedure is repeated until the TCE solvent is clear and the remaining dry minerals in the Soxhlet are weighed (b g). These results are used to determine the initial content of Asbuton bitumen (I (%)) using the formula [Equation (1)]:

$$I(\%) = \frac{a-b}{a} \cdot 100\% \tag{1}$$

SARA (Saturated Aromatic Resin Asphaltane) analysis of Kabungka bitumen was carried out using the gravimetric method in accordance with NIGOGA's (The Norwegian Industry Guide to Organic Geochemical Analyses, Edition 4.0, 30 May 2000) Liquid Chromatographic separation of deasphaltened oils or rock extracts [15]. Meanwhile, the mineral content was analyzed using X-Ray Fluoresence (XRF) Brand: PANalytical, Type: Minipal 4 and for the functional groups of Asbuton Kabungka bitumen, kerosene and diesel oil were analyzed using Fourier Transform Infra Red (FTIR) Brand Shimadzu, Type: IR Prestige 21.

2.3 *Determination of the viscosity of a mixture of bitumen and diluent*

At this stage, 5 g of extracted bitumen is combined in the following proportions with kerosene or diesel oil: 30:70, 40:60, 50:50 and 60:40%(w/w). At temperatures of 50, 60, 70, 80, and 90 °C, each of these mixtures was tested using a kinematic viscosity KV 1000 instrument. The analysis results are observed and calculated based on the resulting viscosity value. The bitumen separation from Asbuton will be done with high-speed stirring for viscosities less than 4 Pa.s.

2.4 *Separation process bitumen from Asbuton*

The separation was carried out in a 500 mL beaker glass. Asbuton (c g) is combined with diluent – kerosene or diesel oil – in a preset ratio depending on viscosity less than 4 Pa.s. This mix was stirred with a magnetic stirrer that is placed on a hot plate that has a temperature setting and a stirring speed. Stirring was done for 30 min at 1000 rpm and a temperature was determined by the viscosity value. These results will be blended again for 5 min with a portable capsule mixer set to 1200 rpm. To separate bitumen-diesel oil or kerosene from mineral Asbuton, water was added to the mixture, resulting in three layers. Using the separation flask, the top layer of bitumen-diesel oil or kerosene is separated, weighed (d) and its density determined.

The standard curve of 1/r versus concentration percentage of Asbuton Kabungka bitumen in kerosene or diesel oil is used to calculate the removal bitumen percentage. This standard curve is created by dissolving pure bitumen in diesel oil or kerosene at concentrations of 50, 40, 30, 20, 10, and 0% (w/w). The density of each mix was measured to generate a standard curve of 1/ r versus bitumen content percentage in the mixture. The standard curve line equation for diesel oil is $y = -336.05x + 428.03$, whereas for kerosene it is $y = -367.58x + 445.51$.

The amount of bitumen in the mixture's upper layer is estimated using a standard curve that already knows the line equation (x, %). The removal bitumen percentage may be computed using the formula [Equation (2)]:

$$\text{Removal of bitumen Asbuton } (\%) = \frac{d \cdot x\ (\%)}{c \cdot I\ (\%)} \cdot 100\% \tag{2}$$

3 Results

3.1 *Raw material characterization*

To obtain the initial content, Kabungka Asbuton that has been sifted with a mesh size of –20/+40 is extracted according to the SNI 03-3640-1994 standard. The initial average content of Asbuton Kabungka bitumen was 16.465% after three extraction repetitions.

3.2 *Analysis of SARA composition on Asbuton Kabungka bitumen*

Saturated (S), Aromatic (A), Resin (R) and Asphaltane (A) are heavy oil molecular compounds that can help to select the separation process and to characterize heavy oils. Asbuton bitumen typically has a SARA composition with a high asphaltene and resin content. In their research, Wenshen, Xiaowen and Jie found that resin and asphaltane from Asbuton were 62.77% [16], whereas Hao *et al.* [17] found that Indonesian oil sand in general had a saturated fraction of 14.05%, aromatic 38.23%, resin 24.51% and asphaltane 23.21%. In addition to the presence of naphtenic acid or carboxylic acid, the content of resin and asphaltane has a significant impact on bitumen viscosity.

The bitumen in Asbuton Kabungka that was analyzed using the gravimetric method in this study had a resin and asphaltene content of 33.95% (Table 1). The results of SARA can be used to determine the interaction between bitumen fraction and physical, mechanical properties, such as the Gaestel index (Ic). Asbuton Kabungka bitumen has an IC of 1.18. This value indicates that the colloid in Asbuton bitumen is not stable, allowing it to settle easily [18].

Table 1: Composition of SARA (Saturated Aromatic Resin Asphaltane) bitumen Asbuton Kabungka

Compositions	Fractionation (%)
Saturated	31.19
Aromatic	34.86
Polar compounds (NSO's) [Resin]	11.01
Asphaltane	22.94

3.3 *Analysis of elemental content in the mineral Asbuton Kabungka*

Mineral rocks are generally classified into two types: carbonate and silicate [19]. Asbuton is a type of oil sand that belongs to the oil-wet oil sand category and is highly hydrophobic [20], [21]. Table 2 shows the results of mineral analysis from Asbuton Kabungka by using XRF.

Table 2: Results of Analysis of Asbuton Kabungka Minerals by using XRF

Compounds	Concentration (%)
Al	1.40 ± 0.10
Si	4.95 ± 0.08
S	2.00 ± 0.07
K	0.24 ± 0.01
Ca	73.85 ± 0.11
Ti	0.57 ± 0.01
V	0.05 ± 0.004
Cr	0.18 ± 0.005
Mn	0.15 ± 0.006
Fe	0.35 ± 0.02
Ni	0.20 ± 0.003
Cu	0.062 ± 0.005
Sr	0.86 ± 0.03
Mo	3.40 ± 0.10
In	2.40 ± 0.09
Ba	0.20 ± 0.03
Yb	0.23 ± 0.01

The mineral Asbuton is made up of calcite, quartz and clay [22]. According to several researchers, the main mineral content of the largest is $CaCO_3$ with levels ranging from 77.67 to 86.66% [11], [23]–[25]. According to XRD analysis, the most abundant Asbuton mineral is $CaCO_3$ [16]. XRF analysis (Table 2) gives results that are similar to the previous research that the largest mineral component of Asbuton Kabungka is calcium with $CaCO_3$ or calcite compounds as much as 73.85%.

Calcite Asbuton is a type of carbonate rock that is naturally water-wet. Calcite absorbs organic [18] readily and forms an adsorbed layer on its surface even with simple organic molecules [26]. The bitumen's is strongly adsorbed on the carbonate surface, causing the rock wettability to become oil-wet [27]. It is easily ionized and accumulates at the oil-bitumen interface. Carboxylic acid is also a metal ion (e.g., Ca^{2+}) and fine solid absorber in asphalt oil. With the mineral composition and SARA owned by Asbuton Kabungka, it is indicated that it requires both physical (quick stirring) and chemical (addition of polar and hydrophobic solvent) treatment at the same time for the release of very hydrophobic bitumen Asbuton Kabungka with oil-wet oil sand properties.

3.4 *FTIR analysis of Asbuton Kabungka bitumen, kerosene and diesel oil*

Kerosene and diesel oil were used as diluents in this study. Figure 1 compares the FTIR analysis results of functional groups from kerosene, diesel oil and Kabungka bitumen. The purpose of this test was to distinguish qualitatively between Kabungka bitumen, kerosene and diesel oil.

The FTIR sample test described above was performed with pelletized KBr. The three samples showed similar absorption in the area above 3000 cm^{-1}, indicating the presence of CH bonds in aromatic compounds; the main absorption was close to 2935–2860 cm^{-1} and there was also absorption around 1470 and 720–725 cm^{-1}, indicating aliphatic chains. The absorption region at 1650–1600 cm^{-1} is a carboxylate bond (carboxylic acid salt) with an aromatic ring, the absorption region 1350–1000 cm^{-1} shows -CO and C=O bonds [28] and the absorption region 720–590 cm^{-1} shows OH bonds. There is no peak in diesel oil between 1350 and 720 cm^{-1}, while it presents in kerosene or Kabungka Asbuton bitumen. In the area

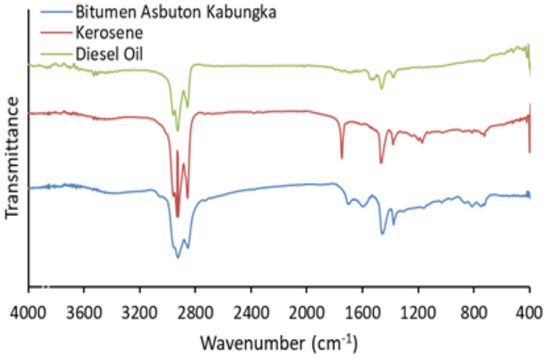

Figure 1: FTIR Spectra of Bitumen Asbuton Kabungka, kerosene and diesel oil.

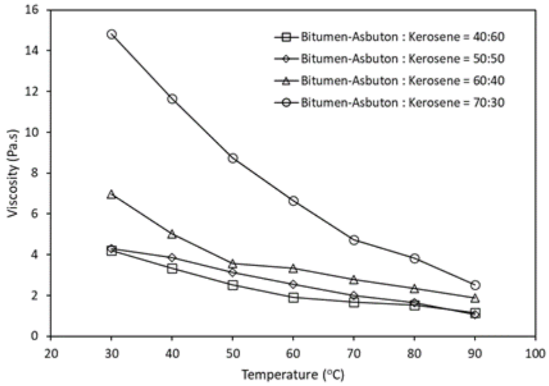

Figure 2: Viscosity of the mixture of bitumen-kerosene at a temperature of 30–90 °C.

around 1490–1410, 880–860 cm^{-1}, Kabungka Asbuton bitumen and kerosene showed the same characteristics, indicating the presence of carbonate ions [29]. In other words, the kerosene group is very similar to the Asbuton Kabungka bitumen group.

The diluent selected based on its polarity similarity to bitumen will be able to act as a penetrating agent to soften and remove bitumen from the pores of the rock. Because a material's solubility is influenced by a variety of factors, including the similarity of functional groups. Based on these findings, kerosene is a superior solvent to diesel oil. The use of kerosene diluents on White Rocks oil sand has also been tested and the bitumen forms a certain contact angle, indicating a tendency to release bitumen from the mineral [30].

Previous studies used diesel oil as a medium to facilitate the release of Asbuton Kabungka bitumen, resulting in a fairly high recovery percentage with the addition of surfactant and NaOH [28]. Based on the FTIR results, it will be determined how much influence the diluent kerosene and diesel oil have on the separation of bitumen from Asbuton Kabungka in this study.

3.5 *Separation bitumen from Asbuton*

The application of the bitumen separation method from the oil sand used is very dependent on the composition of the natural asphalt. This composition affects the physical properties of each natural asphalt. One of the important physical properties to know for the separation of bitumen is viscosity. Bitumen viscosity data is needed to handle the very viscous nature of natural asphalt, such as bitumen in Athabasca and Cold Lake, as well as Kabungka Asbuton, which has a viscosity of 15.4 Pa.s at 100 °C [11]. This value indicates that Asbuton bitumen from Kabungka has a heavy oil character and requires a diluent during the separation process [12], [13]. In order to achieve a low process temperature and high bitumen recovery, certain materials that reduce bitumen viscosity and mineral-bitumen adhesion must also be added [31]. However, a specific study on the separation of bitumen from Asbuton using fuel as a diluent revealed that after 254 days at room temperature, this diluent was unable to reach the bitumen in granular Asbuton, whereas at 90 °C bitumen was taken up about 55% of the time [8], [32]. Based on this, Figure 2 depicts the relationship between the diluent ratio (kerosene and diesel oil), temperature and the resulting viscosity value.

Temperature almost affects bitumen viscosity and solid-bitumen adhesion, which affects bitumen recovery, as shown in Figure 2. When the temperature of kerosene or diesel oil is raised, the viscosity decreases exponentially. The effect of bitumen addition on viscosity is clearly visible in both types of diluents. This suggests that the presence of asphaltene in the bitumen is to blame for the viscosity characteristics. Figures 2 and 3 also shows that at temperatures above 80 °C, there is no significant difference in viscosity in the 50:50, 40:60 ratios of Asbuton bitumen to kerosene/diesel oil (%, w/w). The use of molecular kerosene or diesel oil [12] .

The mixed viscosity of the two diluents in Asbuton Kabungka bitumen has the same tendency in general.

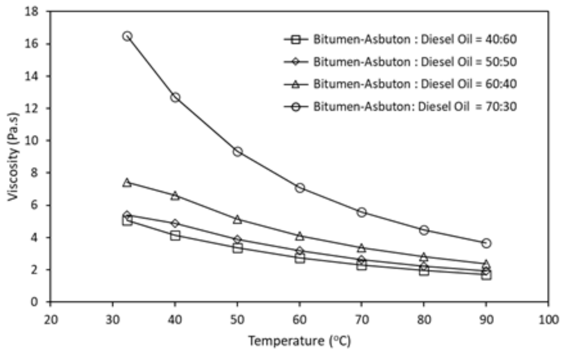

Figure 3: Viscosity of the mixture of bitumen-diesel oil at a temperature of 30–90 °C.

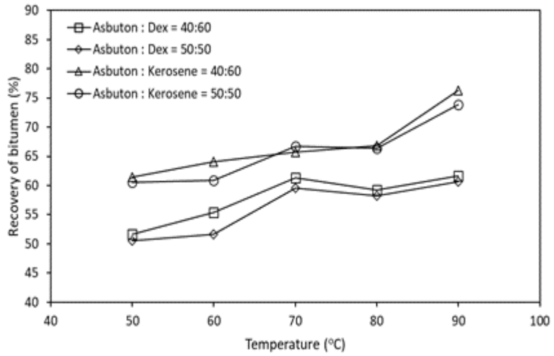

Figure 4: Removal of bitumen Asbuton Kabungka (%).

Kerosene has a lower specific gravity than diesel oil (BJ kerosene 0.80 g/cm^3, diesel oil 0.83 g/cm^3), so the viscosity value produced by kerosene is lower than the mixture containing the same ratio of diesel oil. To demonstrate that the viscosity range of 0.5–2 Pa.s recovery percentage will increase, this study will try the viscosity range of 4 Pa to obtain bitumen recovery percentage through the process of separating bitumen from Asbuton. According to Figure 4, the separation process will be carried out at temperatures above 50–90 °C in a ratio of 50:50 and 40:60 % (w/w).

3.6 *Removal of bitumen (%) in separation process*

The graph above shows that using kerosene and diesel oil as diluents and heating and two times stirring (magnetic stirrer and portable capsule mixer) resulted in the highest recovery percentage of bitumen at a ratio of 40:60 (%, w/w) at 90 °C. Kerosene as a diluent yielded the highest recovery percentage of 76.30%, while diesel oil yielded 61.65%. Stirring and heating are essential because it is necessary to change the viscosity and presence of shear force between Asbuton and the solvent in order to separate bitumen. In this regard, kerosene has a viscosity of 1.07 Pa.s at a ratio of 40:60 (%, w/w) at a temperature of 90 oC, whereas diesel oil has a viscosity of 1.72 Pa.s. According to Hupka *et al.* [12], the viscosity range 0.5–2 Pa.s is the viscosity condition that allows for the greatest recovery percentage. In this regard, kerosene has a viscosity of 1.07 Pa.s at a ratio of 40:60 (w/w %) at a temperature of 90 °C, whereas diesel oil has a viscosity of 1.72 Pa.s. As previously stated, Hupka *et al.* [12] stated that the viscosity range 0.5–2 Pa.s is the viscosity condition that allows for the greatest recovery percentage.

When compared to diesel oil, kerosene is the best diluent for Asbuton bitumen. This is consistent with the FTIR results, which show that kerosene shares more functional groups with Asbuton bitumen. These findings also show that when using kerosene with bitumen content greater than 50%, the relationship between viscosity and recovery percentage has the same tendency. However, at 90 °C, there was a significant increase in recovery percentage. This demonstrates that temperature has a significant impact on the bitumen separation process using diluent kerosene. The viscosity characteristics of bitumen-kerosene ratio of 40:60 % are almost identical to those of kerosene and it produces the highest recovery percentage, without any deposits.

Asphaltane is one of the components of bitumen that is responsible for the viscosity of bitumen. The physical bonding of the hydrocarbon components with high molecular weight will be affected by the temperature increase. The hydrocarbon components will be broken down, physical bonds will be disrupted and viscosity will be decreased. Asbuton's capacity to decrease bitumen viscosity in kerosene is superior to diesel oil. This is due to the interplay between the main functional groups' similarity and the strong hydrogen bonds between kerosene and asphaltene. This assertion is consistent with the FTIR data, which show that kerosene has a higher functional group similarity when compared to gasoline.

As a consequence of this finding, continuing the experiment using kerosene as a diluent will require additional research. Combining physical treatments such as grinding or mixing with the use of a specialized stirrer for homogenizing dispersion at high velocity makes it simple to achieve greater removal of bitumen Asbuton. The equipment used to make bitumen Asbuton at a pressure of 2 Pa.s. should have temperature control, according to the conclusions of this study.

4 Conclusion

This study provides diluent information that can be used to extract bitumen from Asbuton. According to the FTIR results, kerosene has a functional group that is similar to Asbuton bitumen. This similarity results in a higher recovery percentage of bitumen using kerosene than diesel oil in every comparison and process temperature. The bitumen recovery percentage is heavily influenced by viscosity and temperature. The maximum value of the bitumen recovery percentage for Asbuton-kerosene bitumen mixture is 76.30% when the viscosity is in the range of 2 Pa.s, the ratio is 40:60 (%, w/w) and the temperature is 90 °C. Kerosene performs well as a diluent for Asbuton bitumen when used in the simple method. A grinding or mixing tank with a specific stirrer for homogenizing dispersion and controlled temperatures to extract more bitumen from Asbuton is the best alternative for further study.

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