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Extraction of Asphalt from Asphalt Concrete Using Subcritical Water

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ABSTRACT

Although asphalt concrete is recyclable, recycled asphalt mix is less durable than virgin asphalt mix due to the mixing of deteriorated straight and polymer modifier asphalt, and various chemical additives. Therefore, to improve the quality of recycled asphalt mix, extraction of deteriorated asphalt from asphalt concrete prior to its recycling is essential. It could enable further adequate chemical processing of the extracted asphalt. Thus, the extraction of asphalt from the residues of pulsed power discharge inside asphalt concrete lumps was conducted in an autoclave. The effect of treatment time and cooling conditions on asphalt extraction was investigated at 300°C, 450 rpm, and 2mL/g as respective temperature, stirrer speed, and ratio of the amount of water to that of the residues. The authors found that 3h as treatment time, refraining from stirring during treatment, and continuous stirring at 450 rpm during natural cooling to a temperature corresponding to the softening point of the recovered pure asphalt represent the optimum asphalt extraction conditions. Finally, penetration, softening point, and ductility tests were performed on the recovered pure asphalt. Results of asphalt properties tests suggest that the extracted asphalt still contained a significant amount of polymer modifier and relatively small amount of oily components. For this reason, the extracted asphalt requires further adequate rejuvenation to improve its quality as construction material.

INTRODUCTION

Background

Recycling of construction and demolition waste (CDW) including asphalt concrete waste is essential to save financial resources and mitigate environmental issues such as deforestation, natural resources depletion, and air and water resources pollution [Pacheco-Torgal 2013]. Although asphalt concrete is recyclable, recycled asphalt mix is less durable than virgin asphalt mix due to the mixing of deteriorated straight and polymer modifier asphalt and various chemical additives [JMAA 2007; Watson 2011; Kubo 2009]. Asphalt concrete is composed of asphalt and aggregate. Asphalt, also known as bitumen derives from the fractional distillation of crude oils at temperature ranging from 300 to 350°C or from natural

deposits such as bituminous sands [McNally 2011]. Asphalt consists of asphaltenes, resins, aromatics, and paraffins. To satisfy increasing demands for energy in terms of light hydrocarbons, refineries upgrade part of asphaltenes-rich fractions into lighter ones [Wang and Anthony 2003]. In fact, consumption of fossil fuels is estimated to increase and stabilize from 2025 to 2075 [Mohr, Wang, Ellem, Ward, and Guirco 2015]. In addition, as a non-renewable natural resources [Höök and Tang 2013], oil reserves are expected to be exhausted by 2042 [Shafiee and Total 2009]. Generally, asphalt contained in asphalt concrete wastes has lost a substantial amount of its low molecular weight components namely the aromatics and paraffins and thus; consists mainly of asphaltenes and resins [McNally 2011].

Consequently, to promote sustainable management of asphalt concrete wastes and crude oils; and improve the quality of recycled asphalt it is essential to extract asphaltenes-rich asphalt from asphalt concrete waste and investigate its recyclability and upgradability into lighter hydrocarbons. To this end, while sub- and supercritical water has been used to process fuels materials such as bituminous sands [Brunner 2014]. Few pieces of research have investigated the application of subcritical water to the recycling of asphalt concrete. In one study, Kano and colleagues successfully removed asphalt from asphalt mixture using high-temperature and-pressure water [Kano, Akiba, and Kuriyagawa 2006]; in another, asphalt was shown extractable from asphalt concrete using a Soxhlet extraction set-up [Brunner 2014].

Objectives

The authors have proposed the application of pulsed power discharge [Shigeishi et al. 2013; Akiyama 2003; Amoussou, Ishimatsu, Oyama, and Shigeishi 2015], and subcritical water to extract asphalt from asphalt concrete lumps (Figure 1). This paper focuses on the extraction of asphalt from the residues of pulsed power discharge applied to asphalt concrete lumps in subcritical water. The two goals of this paper are to investigate the performance of subcritical water at extracting asphalt from the residues and to determine the optimum treatment time and cooling conditions.

MATERIALS AND METHODS

Production of the asphalt concrete residues

The authors have previously applied pulsed power discharge to separate asphalt concrete into recycled aggregate and residues containing asphalt [Amoussou et al. 2015]. Figure 2 shows production of residues due to pulsed power discharge to the interior of submerged pieces of asphalt concrete lumps. The residues were composed of fine aggregate and asphalt binder.

Subcritical water extraction

The critical temperature and pressure of water are 373.946°C and 22.064 MPa, respectively [Brunner 2014]. Water under either temperature below 373.946°C or pressure lower than 22.064 MPa, is considered to be in subcritical condition. Asphalt was recovered from the above residues using subcritical water in an autoclave (Figure 3) with internal volume of 500ml (OM LABTECH, CO, Ltd., Japan). At the start, about 50g of residues with an asphalt content of 7.86% and 100 ml of water were loaded to the autoclave, which was subsequently heated to 300 °C (Figure 4). After four different treatment times (1, 2, 3, and 4h) had elapsed, the heater was stopped. The stirrer was inactivated during heating while continuous stirring at 450 rpm was applied during cooling. The stirrer was inactivated again when the internal temperature of the autoclave reached the target temperatures. Cooling conditions of C0 and C1 were similar, with the target temperature for C0 60°C and that of C1 70°C.

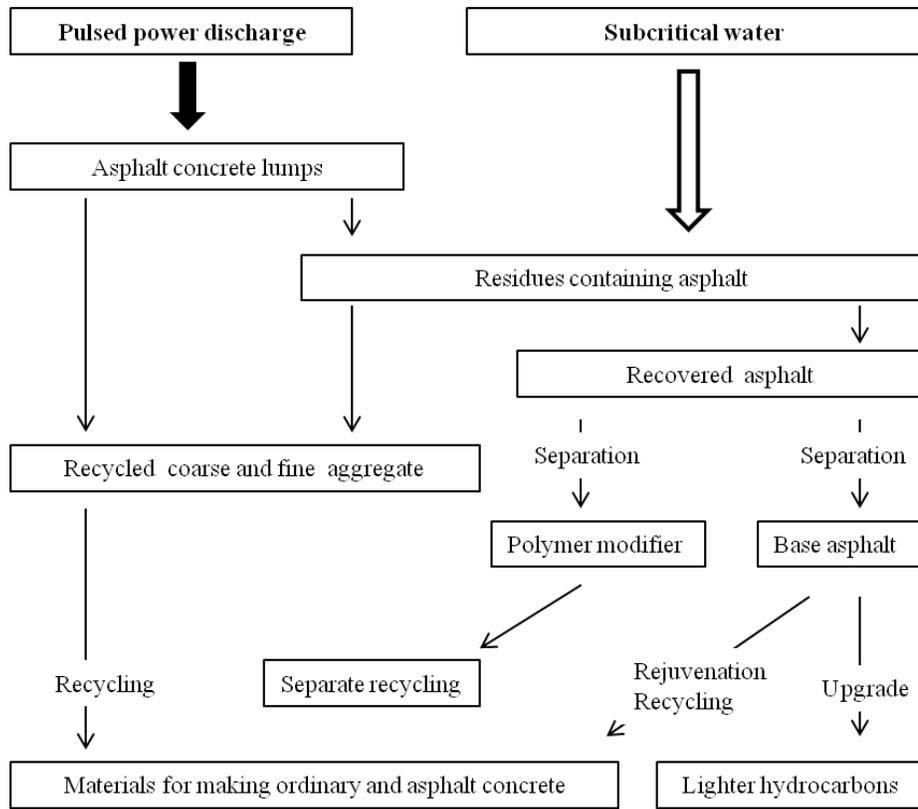


Figure 1. Proposed Asphalt Concrete Recycling

It is the softening point of the recovered pure asphalt produced under cooling condition C0. The five cooling and heating conditions utilized, C0, C1, C2, C3, and C4, are summarized in Table 1 and Figure 4.

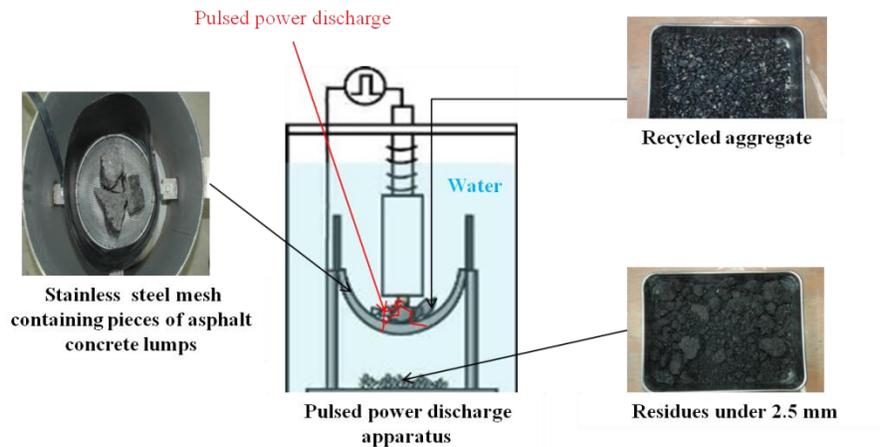


Figure 2. Production of Asphalt Concrete Residues

Cooling conditions C1, C2, and C3 were conducted to investigate the effects of cooling time on the quality of the recycled aggregate and recovered asphalt, in order to discern the optimum cooling conditions. C0 and C4 were applied to confirm the effects of a continuous stirring of the contents of the batch-type reactor of the autoclave on the pure asphalt recovery rate and quality of the recycled aggregate.

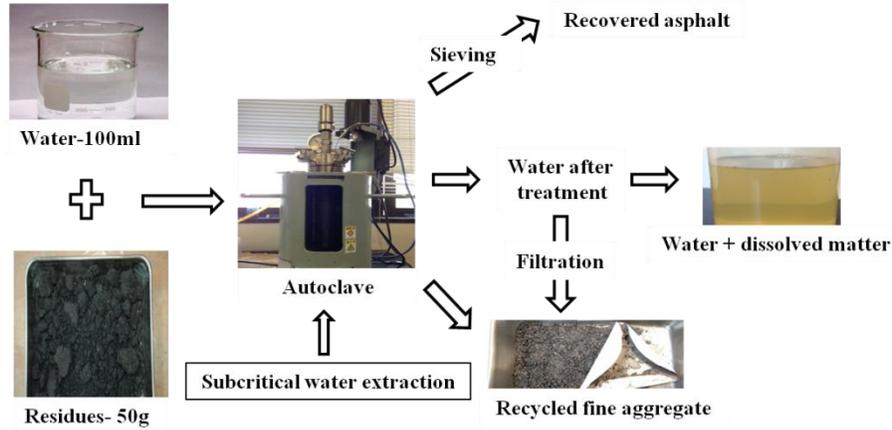


Figure 3. Subcritical Water Extraction

After cooling according to the conditions described in Table 1, products resulting from subcritical water extraction (Figure 3) were categorized as recovered asphalt, recycled aggregate, and water after treatment. The target temperature is the temperature at which the stirrer was turned off.

Table 1. Cooling Conditions

Conditions	C0	C1	C2	C3	C4
Target temperatures (°C)	60	70	70	70	55
Types of cooling	Natural	Natural	Rapid	Rapid	Natural

Exclusive of water resulting from the application of cooling conditions C1, C2, C3, and C4, suspended matter was collected by suction filtration as a part of the recycled fine aggregate for C0. To evaluate amount of recovered asphalt and recycled aggregate, they were dried at room temperature and $105 \pm 5^\circ\text{C}$, respectively and weighed. Recovered asphalt was sieved under 1.2, 2.5, 5, 9.5, 16, 19, and 26.5 mm apertures to determine size distribution.

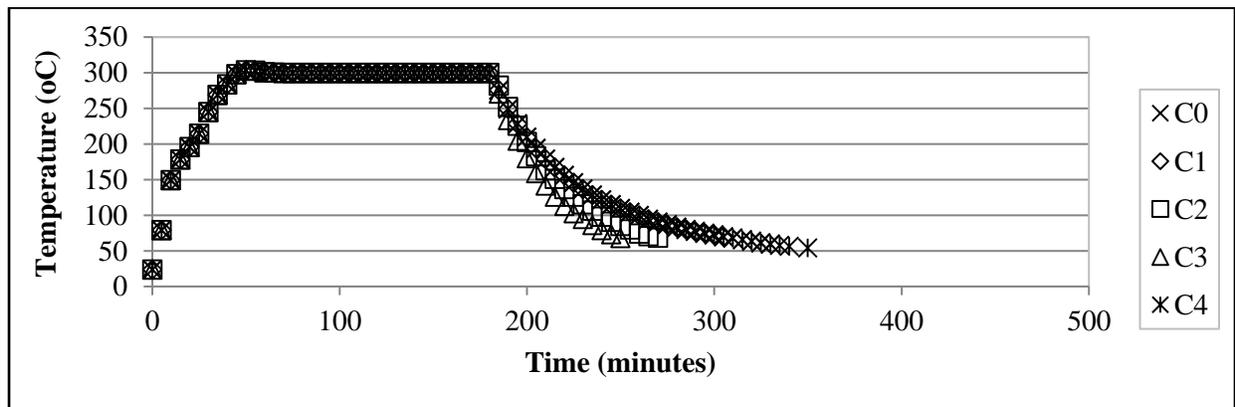


Figure 4. Heating and Cooling Conditions

Asphalt and aggregate purity and mass balances

The asphalt ignition method [ASTM D6307-10] was applied to evaluate pure asphalt and aggregate contents of residues, recycled aggregate, and recovered asphalt. Ignition tests were carried out at $540 \pm 5^\circ\text{C}$ for 1 h using an electrical furnace KBF626N1 (Koyo Thermo Systems Co., Ltd., Japan) to investigate pure asphalt and aggregate mass balances according to equations 1 and 2.

$$AS_o + AS_{W_o} = AS_{RAS} + AS_{RAG} + AS_{WL} \quad (1)$$

where AS_o is the pure asphalt content of residues (g), AS_{W_o} is the initial pure asphalt content of water (g), AS_{RAS} is the pure asphalt content of recovered asphalt (g), AS_{RAG} is the pure asphalt content of the recycled aggregate (g), and AS_{WL} is the asphalt content of water and lost portion of the residues (g).

$$AG_o + AG_{W_o} = AG_{RAS} + AG_{RAG} + AG_{WL} \quad (2)$$

where AG_o is the aggregate content of residues (g), AG_{W_o} is the initial aggregate content of water (g), AG_{RAS} is the aggregate content of recovered asphalt (g), AG_{RAG} is the aggregate content of the recycled aggregate (g), and AG_{WL} is the aggregate content of water and lost portion of the residues (g).

Recovered asphalt and recycled aggregate purity was computed using equations 3 and 4.

$$PRAS = \frac{AS_{RAS}}{RAS} * 100 \quad (3)$$

where $PRAS$ is the purity of the recovered asphalt (%), AS_{RAS} is the pure asphalt content of recovered asphalt (g), RAS is the mass of recovered asphalt (g).

$$PRAg = \frac{AG_{RAG}}{RAG} * 100 \quad (4)$$

where $PRAg$ is the purity of the recycled aggregate (%), AG_{RAG} is the pure aggregate content of the recycled aggregate (g); RAG is the mass of recycled aggregate (g).

Properties of recovered asphalt by cooling condition C0

Penetration, softening point, and ductility tests were performed at the Kumamoto Prefectural Center of Constructional Technology, Kumamoto, Japan on pure asphalt recovered under cooling condition C0.

RESULTS AND DISCUSSION

Cooling condition C0

Effect of treatment time. Figure 5 shows that the highest pure asphalt content of recovered asphalt (about 91%) and lowest pure asphalt content in recycled aggregate (about 2.1 %) were achieved at 3h. These results imply that 3h were required to enable an effective separation of asphalt from the recycled aggregate. However, treatment time of 4h resulting in mixing of aggregate and asphalt due to continuous stirring during cooling. For this reason, pure asphalt content in the recovered asphalt decreased from 90.75% to 75.42 % while that in the recycled aggregate increased to 17.8 %.

Properties of recovered asphalt by cooling condition C0 at 3h. Results of quality tests performed on the recovered pure asphalt produced under cooling condition C0 are listed in Table 1.

Table 1. Properties of Recovered Asphalt by Cooling Condition C0 at 3h

Penetration (1/10 mm)	Softening point (°C)	Ductility (cm)
12	70.0	1

A comparison of these results with standard properties of asphalt [JMAA 2007] indicates that the extracted asphalt still contains a significant amount of polymer modifier but a relatively small amount of oily components. Consequently, the recovered asphalt hardens, thus requiring proper rejuvenation before being recycled as material for asphalt concrete formation.

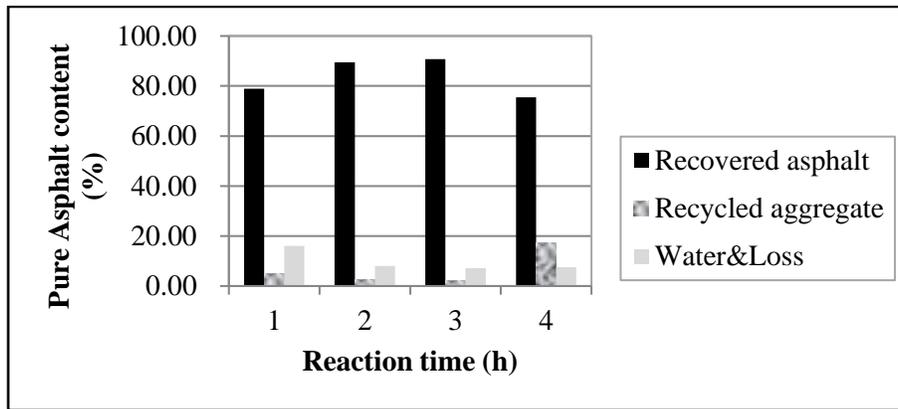


Figure 5. Effects of Treatment Time

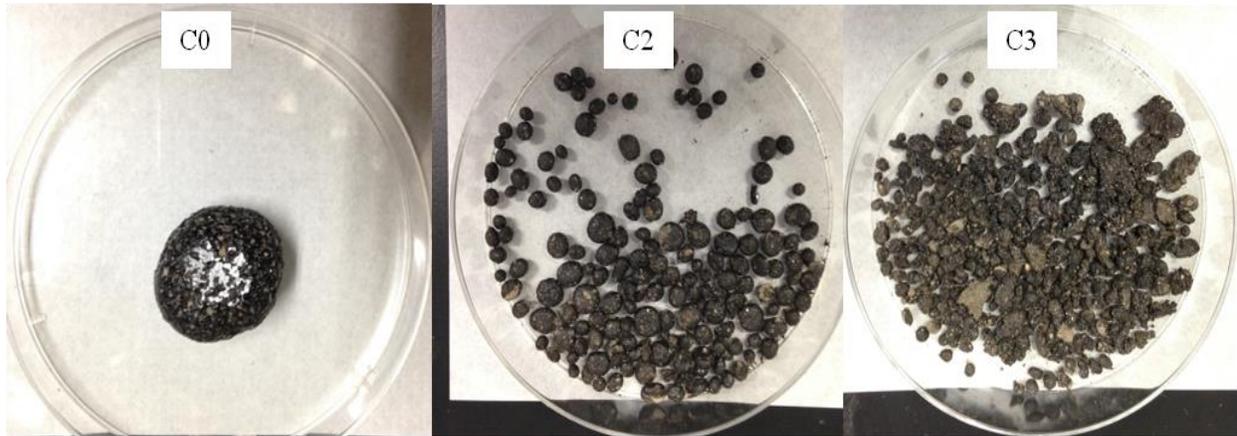


Figure 6. Recovered Asphalt at 3h under Cooling Conditions C0, C2, and C3

Effects of cooling conditions on recovered asphalt and recycled aggregate. Figure 4 shows that approximately 125, 90, and 70 minutes were required for C1, C2, and C3, respectively, to quench the contents of the autoclave to about 70 °C. Figure 7 illustrates size distribution for recovered asphalt under cooling conditions C0, C1, C2, C3, and C4. Conditions C0 and C4 produced mainly a spheroid (Figure 6) while C1 produced a piece of irregularly-shaped recovered asphalt (Figure 8), respectively with sizes smaller than or equal to 26.5 mm but larger than 19 mm. On the other hand, about 80 % of the granular pieces produced under cooling conditions C2 and C3 (Figure 6) were smaller than or equal to 5mm but

larger than 2.5 mm. When cooling time decreased from 125 to 70 mins, the amount of portions larger than 5mm but smaller than or equal to 26.5 mm decreased drastically from 100 % to about 20 %.

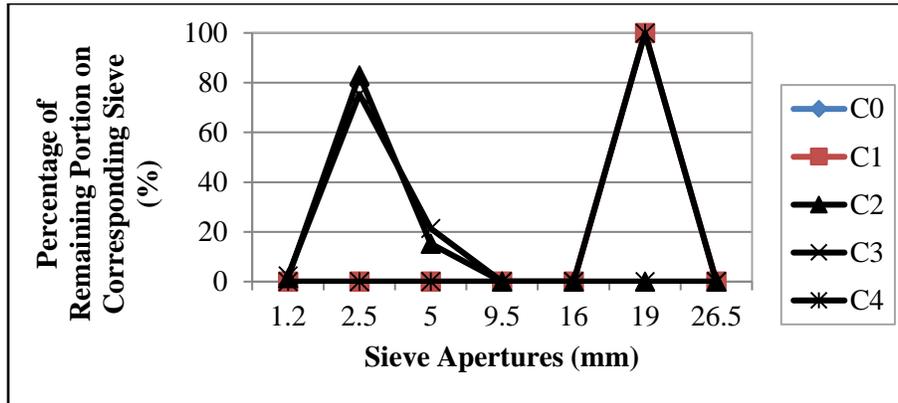


Figure 7. Effects of Cooling Conditions on Recovered Asphalt Size

These results provide compelling evidence that the size of recovered asphalt decreases with decreased cooling time. However, finer sizes of recovered asphalt result in more difficulty in its separation from the recycled aggregate.

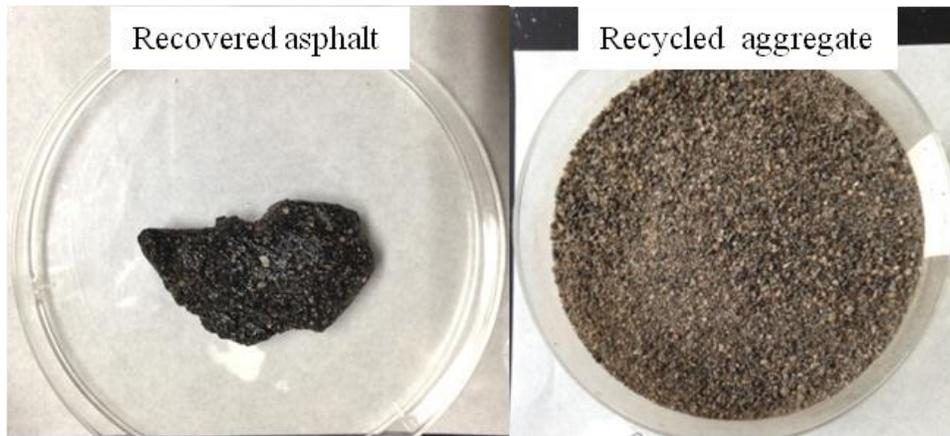


Figure 8. Recovered Asphalt and Recycled Aggregate under Cooling Condition C1

Cooling conditions apparently affected the quality of recycled aggregate and recovered asphalt. Cooling to the softening point of recovered pure asphalt improves the quality of the recycled aggregate by decreasing its asphalt content (Figure 9). Conditions C0 and C1 produced almost asphalt-free recycled aggregate compared to C2, C3, and C4. For instance, when the stirrer was turned off at 55°C (C4), pure asphalt content of recycled aggregate increased to 2.11%. However, asphalt content of recycled aggregate produced under C1 (0.05 %) was lower than that of recycled aggregate produced under C0 (0.22 %). On the whole, these results highlight the fact that asphalt content of recycled aggregate increases when cooling time decreases due to the decrease in recovered asphalt size. Furthermore, spheroidization occurred during cooling when the target temperature is inferior to 70°C (softening point of the recovered pure asphalt under C0).

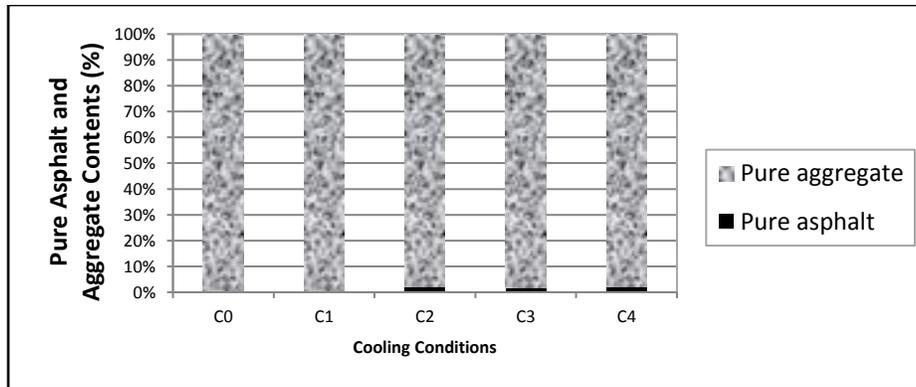


Figure 9. Effects of Cooling Conditions on Aggregate Quality

It is the result of the friction between the solidifying irregularly-shaped recovered asphalt (Figure 8), recycled aggregate (Figure 8), stirrer, and inner part of the batch-type reactor of the autoclave. We could infer that spheroidization required natural cooling to 70°C for at least 125 minutes and depended on the shape of the recovered asphalt at the same temperature corresponding to its softening point. The evidence suggests that spheroidization contributed to an increase in asphalt content of recycled aggregate when the contents of the autoclave are quenched to temperatures smaller than that of the softening point of the pure recovered asphalt under natural cooling namely 60 and 55°C.

Asphalt and aggregate purity and mass balances. Data in Figure 10 show that conditions under C1 helped to increase the recovery rate of pure asphalt; and to decrease the asphalt contents of recycled aggregate, water, and lost portions of the residues.

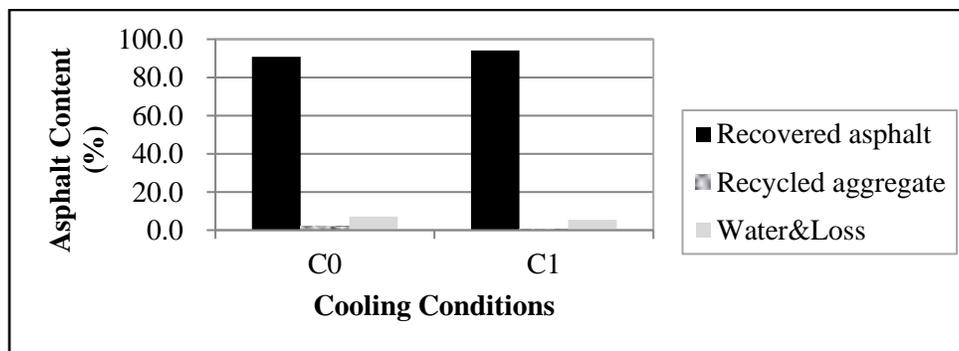


Figure 10. Pure Asphalt Mass Balance for Cooling Conditions C0 and C1

Cooling to about 70°C instead of 60°C resulted in an increase in the recovery rate of pure asphalt from about 91% to approximately 94%. While the asphalt content of recycled aggregate decreased significantly from about 2.1% to 0.4%. Similarly, asphalt content in water and lost portions of the residues decreased slightly from about 7.2% to 5.6%. These results point to an optimum treatment time and cooling conditions of 3h and C1, respectively. Figure 11 shows asphalt and aggregate purity and mass balances associated with the optimum subcritical water extraction of asphalt from 1000g of the residues.

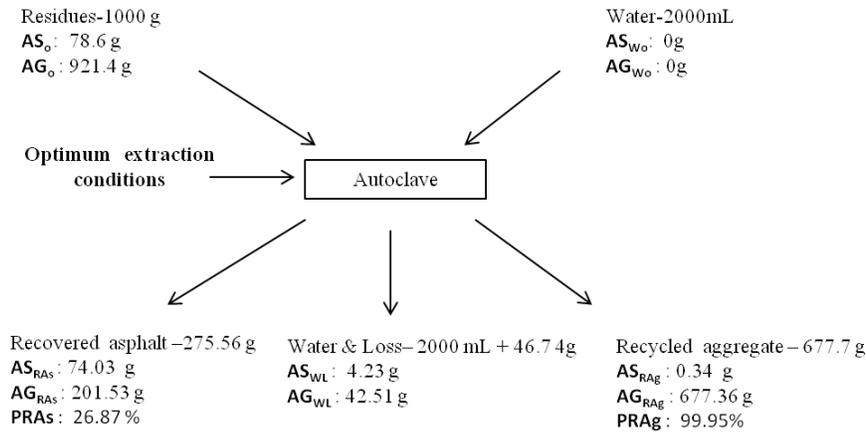


Figure 11. Purity and Mass Balances for Optimum Extraction Conditions

Though purity of recovered asphalt was only about 27%, it contained about 94% of the initial pure asphalt content of the residues. Recycled fine aggregate showed extremely high purity (99.95%), while its pure aggregate content was about 74 % of the initial content of the residues. Pure aggregate content in water and lost portions of the residues was about 42.51 g; this is because under condition C1, filtration was not carried out to separate residual fine aggregate from water after subcritical water extraction. According to these results, it is probable that residual fine aggregate obtained after filtration of water after subcritical water extraction might have contributed to the increase in the pure asphalt content of the recycled aggregate produced under cooling condition C0.

CONCLUSION

In this paper, asphalt was extracted from residues resulting from pulsed power discharge to pieces of asphalt concrete lumps using subcritical water. We found that optimum treatment time at 300°C was 3h with a stirring speed of 450 rpm and ratio of water amount to residues of 2mL/g. The optimum cooling condition was found to be natural cooling to the softening point of the recovered pure asphalt. Recycled aggregate quality improved by decreasing its pure asphalt content and that of water and lost portions of residues. As a result, pure asphalt recovery rate increased as well. At this point, results suggest that natural cooling up to the softening point of the recovered pure asphalt is essential to improve the recovery rate of pure asphalt and quality of recycled aggregate, and facilitate the separation of recovered asphalt from recycled aggregate after its subcritical water extraction from residues. Future work should investigate the effects of the deactivation of the stirrer at temperatures above that of the softening point of the recovered pure asphalt to determine the temperature range at which the stirrer could be inactivated to maximize the pure asphalt recovery rate and purity of the recycled aggregate; and to minimize the amount of lost portions of the residues.

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