Rheology of Virgin Asphalt Binder Combined with High Percentages of RAP Binder Rejuvenated with Waste Vegetable Oil

Connor R. Dugan, Chris R. Sumter, Shivani Rani, S. Ashik Ali, Edgar A. O’Rear,* and Musharraf Zaman

ABSTRACT: Waste cooking oils (WCOs) show promise as a rejuvenator for reclaimed asphalt pavement (RAP) binders. Their use helps to make RAP a renewable resource and to address environmental concerns related to WCO disposal. While studies suggest that 100% RAP for pavement is feasible, RAP will likely be combined with a virgin binder and aggregate incrementally in the field. In this study, the rheological properties of the virgin binder blended with a simulated RAP binder and 10% waste vegetable oil (WVO) as a rejuvenator were examined. Viscosities below that of a PG 64-22 virgin binder were observed with WVO in blends of 40 or 60% RAP and the virgin binder. The virgin-60% RAP/WVO binder blend retained a Superpave grade of the virgin binder and was found to satisfy requirements for both rutting parameter and fatigue resistance. Results indicated that WVO significantly reduced the effects of long-term ageing, making the blend containing RAP durable. The effect of RAP content on WVO/virgin binder blends was most pronounced on the rutting parameter. A "molecular lubrication" model is suggested as a mechanism for the viscosity reduction with WVO.

INTRODUCTION

Transportation contributes to the functioning of societies in many ways, with significant economic impact through its roles in commerce and with a higher quality of life for citizens as a result of greater mobility. The benefits to the public justify the huge investment in time, money, and energy that go toward the materials acquisition, handling, and disposal required for road construction and maintenance. With over 80% of highways in the world covered with asphalt pavement1 and annual demand for the asphalt binder on the order of 100 million tons,2 the potential gains from recycling toward sustainability are tremendous. Economics favor recycling, with significant savings from asphalt binder and aggregate reuse, as well as the costs associated with hauling new materials to a construction site and carrying wastes away for disposal. It has been estimated that the overall cost can be reduced by half or more using 100% reclaimed asphalt pavement (RAP).3 The asphalt binder represents a large fraction of these potential savings. A trend since 2000 of rapidly increasing asphalt binder cost coupled with price volatility and a broader awareness of the need for sustainability has created a growing desire to increase the usage of RAP by state departments of transportation.

Changes in the properties of the aged asphalt binder due to volatilization and oxidation have been cited as the major impediment to recycling of binder.4 Rejuvenators act to improve the rheological properties of the RAP binder and thereby make it a renewable resource. With rejuvenators, high percentages of RAP up to 80%5 and even 100%3,6–8 have been shown to be possible.

Recently, there has been much interest in alternative rejuvenators and particularly waste materials like used engine oil or cooking oils.8 Their performance indicates great promise. For instance, Ji et al.5 found that a waste vegetable oil (WVO) outperformed a commercial rejuvenator overall. Waste cooking oil (WCO), including animal and/or vegetable fats, is itself a considerable environmental problem. In the United States alone, the WCO daily production has been estimated to be 100 million gallons or about 380 000 tons,9 much of which goes to landfills. When poured down a drain, greases and oils can clog pipes and contaminate waterways. Thus, their use as a rejuvenator not only enables the recycling of RAP but also helps to address environmental problems with WCOs.

Much of the research with WCOs as a rejuvenator has been on the amount added to 100% RAP binder and its effect on the rheological properties of the restored binder. Ji et al.5 found that a waste vegetable oil (WVO) outperformed a commercial rejuvenator overall. Waste cooking oil (WCO), including animal and/or vegetable fats, is itself a considerable environmental problem. In the United States alone, the WCO daily production has been estimated to be 100 million gallons or about 380 000 tons, much of which goes to landfills. When poured down a drain, greases and oils can clog pipes and contaminate waterways. Thus, their use as a rejuvenator not only enables the recycling of RAP but also helps to address environmental problems with WCOs.

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coauthors carried out rheological testing on a simulated RAP binder and found that 4–6% WCO yielded rutting resistances similar to the virgin binder. This group also found that several asphalt binders recovered from RAP retained a high-temperature Superpave grade at 10% WCO. Improved rheology was observed by Zaumanis et al. when using 12% WVO with the reclaimed binder. Tests included penetration and softening point for rutting, linear amplitude sweep for fatigue, and kinematic viscosity for workability.

A recent review notes the importance of dispersion of a virgin binder, recovered RAP binder, and rejuvenating agent, and yet research is lacking on the rheological properties of an asphalt binder with all three components when a WCO is used. Instead, investigators have examined either virgin binders or RAP binder alone with WCOs. Since prior work with WCOs was carried out with 100% RAP binders, the effect of increasing the amount of RAP has not been determined. Thus, the focus of this study was to investigate the rheological properties of reconstituted binders composed of a simulated RAP binder at varying amount, WVO, and virgin binder under different ageing conditions.

Additionally, approaches to studying the effects of WCO with the RAP binder have been highly variable. The amount and type of WCO used as a modifier or rejuvenator have varied. Waste oils have come from residences and/or restaurants often with ill-defined components, lengths of service, and applied temperature. With that source of variability taken into consideration, some investigators have employed fresh vegetable oils of known composition. Researchers have conducted tests with the recovered binder from RAP or with the simulated RAP binder prepared by ageing the virgin binder. In addition to a need for work with a ternary system, the WCO literature with the RAP binder is deficient in reports where the final blend was aged and the rheology further characterized. Therefore, in this study, efforts were made to evaluate the properties of a ternary system with 40 and 60% simulated RAP binders, a 64-22 virgin binder, and WCO under different ageing conditions. Also, the effect of time of use of a WVO was examined, which heretofore has not been described.

## RESULTS AND DISCUSSION

The complex and highly variable composition of asphalt derives from the nature of the petroleum feedstocks and the different processes utilized for its production. Mechanical testing helps to ascertain the suitability of a particular asphalt along with any additives for the creation and application of the mix and performance of the pavement under load and weathering conditions. Additives like antistripping agents and warm mix products are usually employed at a minimal level to keep costs down, while the use of more WVO would better address the waste disposal problem. The RAP binder and WVO tend to counterbalance each other in a ternary system, including the virgin binder. As such, results on how the amount of RAP binder influences the mechanical properties are of interest.

Rotational Viscosity (RV). Rheological properties from dynamic shear rheometer (DSR) tests serve as an indicator of rutting and cracking performance of the binder, when used in an asphalt mix, whereas viscosity reflects the coatability of asphalt binder on aggregate at the hot mix asphalt (HMA) plant. A lower viscosity enables the effective coating of aggregate and permits lower processing temperature to reduce energy requirements and emission of volatile organics. As shown in Figure 1, the viscosity measurements displayed considerable dependence on composition and a typical trend in viscosity with temperature. Addition of the RAP binder to the virgin binder increases the viscosity, as expected. Results were consistent with 60% RAP samples being more viscous than the corresponding 40% blends. At 135 °C, for example, the viscosities of the 40 and 60% RAP binder blends were found to be 962 and 1337 mPa·s, respectively, compared to that of 546 mPa·s for the virgin binder. However, the values with RAP–virgin binder blends decreased markedly with the addition of WVO. The viscosity of the 40% RAP binder blend reduced to 333 mPa·s with 1 day used WVO and to 429 mPa·s with 11 day used WVO. Similarly, the viscosities of the 60% RAP binder blend became 425 and 445 mPa·s for 1 and 11 day used WVO, respectively. Thus, the addition of WVO reduced the viscosity of RAP binder blends (40 or 60%) to values below that of the virgin binder. Similar trends were observed at higher temperatures.

Rolling thin-film oven (RTFO) ageing mimicked the effects of HMA processing on virgin binder properties. The viscosity of all binder blends increased on short-term ageing although the magnitude of the change was greatly reduced with the used vegetable oil (Figure 2). At 135 °C, the viscosity of the PG 64-22 virgin binder was 945 mPa·s for the virgin binder. However, the values with RAP–virgin binder blends decreased markedly with the addition of WVO. The viscosity of the 40% RAP binder blend reduced to 333 mPa·s with 1 day used WVO and to 429 mPa·s with 11 day used WVO. Similarly, the viscosities of the 60% RAP binder blend became 425 and 445 mPa·s for 1 and 11 day used WVO, respectively. Thus, the addition of WVO reduced the viscosity of RAP binder blends (40 or 60%) to values below that of the virgin binder. Similar trends were observed at higher temperatures.

![Figure 1](https://dx.doi.org/10.1021/acsomega.0c00377)  
**Figure 1.** Viscosities of unaged virgin binder and combined virgin—RAP binders with and without WVO (n = 3; standard deviation was 7.7 Pa·s or less). The addition of WVO reduced the viscosity of the blends. Curves for 40% RAP-1 day used oil and 60% RAP-1 day used oil are marked by the curve for the 60% RAP-11 day used oil.

![Figure 2](https://dx.doi.org/10.1021/acsomega.0c00377)  
**Figure 2.** Viscosities of the virgin binder and combined virgin—RAP binders with and without WVO after RTFO (n = 3; standard deviation was 7.2 Pa·s or less). Curves for 40 and 60% RAP with 11 day used oil cover the respective 1 day oil results.
blend to 562 mPa·s (1 day used WVO) and 541 mPa·s (11 day used WVO). For the RTFO-aged 60% RAP binder blend, the corresponding results for 1 and 11 day used WVO were 700 and 675 mPa·s, respectively. Different periods of use of the WVO, such as 1 or 11 day used oil, seemed to make little difference in the results. These findings suggest that mixing and compaction temperatures could be lowered to save energy and reduce emissions of volatile organics, as evident in Table 1. The asphalt mixes containing RAP can be mixed and compacted at similar temperatures to those for control mixes when blended with 10% WVO.

Table 1. Mixing and Compaction of Unaged Binder Blends Containing RAP and WVO

| binder blend type | mixing temperature (°C) | compaction temperature (°C) |
|------------------|--------------------------|-----------------------------|
| virgin binder    | 156                      | 145                         |
| 40% RAP          | 172                      | 160                         |
| 60% RAP          | 180                      | 165                         |
| 40% RAP + 1 day oil| 154                      | 140                         |
| 40% RAP + 11 day oil | 159                      | 147                         |
| 60% RAP + 1 day oil | 164                      | 151                         |
| 60% RAP + 11 day oil | 157                      | 145                         |

*Viscosity = 170 mPa·s. **Viscosity = 280 mPa·s.

A relatively small amount of WVO significantly affects the viscosity. The mechanism by which WVO acts is an interesting question meriting discussion. These findings for the viscosity of aged and unaged binder blends containing the RAP binder may offer some insight. A rejuvenator acts to restore the mechanical properties of aged asphalt, but it appears here that it also mitigates the effects of short-term ageing.

The main mechanisms of ageing are considered to be loss of volatiles, oxidation, and steric hardening with oxidation.28 As evidence of oxidation, other researchers using Fourier transform infrared (FTIR) spectroscopy have found the formation of sulfoxides and ketones with a shift in composition to more asphaltenes and less resins and aromatics,29,30 which have been reported to correspond to increased stiffness.28,30 Their FTIR results are consistent with oxidation as a factor that fits nicely with antioxidants being shown to lessen the effects of short-term and long-term ageing of a binder.31

Vegetable oils themselves are prone to oxidation.32 The polyunsaturated fatty acids in WVO are highly susceptible to oxidation with the formation of aldehydes, ketones, epoxyoxides, and hydroxy compounds with trans, trans-2,4,decadienal, a major decomposition product of heated oxidized linoleate. As such, the unsaturated fatty acids could serve as sacrificial species to mitigate the effects of ageing. However, in a small companion study, we used an X-ray microanalysis technique, wavelength-dispersive spectroscopy (WDS), to examine the sample composition of carbon, sulfur, and oxygen before and after ageing (Table 2). Results did not support a protective effect of WVO against oxidation during ageing as large differences in viscosity did not correspond to oxygen content. Moreover, while protection by unsaturated fatty acids seems plausible, Zaumanis et al. found that triglycerides with mostly lauric and myristic acid moieties increased PG temperature after ageing (i.e., did not protect against ageing).33

The fact that the WVO in this study reduces the viscosity of a binder with a high fraction of RAP binder (Figure 1) without correspondence to the oxidation of the binder blends suggests that another mechanism is at play. Further examination of the molecular structure helps to provide a possible explanation of how WVO can lower the viscosity of both aged and unaged binder blends. The asphalt binder is a complex mixture sometimes described as maltenes and asphaltenes; recently, asphalt binder fractions in the order of increasing polarity have been denoted as saturates, aromatics, resins, and asphaltenes. Li et al. and Greenfield proposed a number of model compounds representative of similar groupings (saturates, naphthenic aromatics, polar aromatics, asphaltenes).34 We note that the representative molecules are almost entirely large multiringed structures. Such molecules and their clusters will be quite rigid, with limited conformational degrees of freedom and limited ability to bend and rotate under stress. They will be effective in transferring momentum across lines of shear. Therefore, they will be more viscous since viscosity is a measure of the ability of a fluid to transmit momentum by shear forces. In contrast, the glycerides in the fats are much more flexible molecules that can readily bend and rotate under application of a shear force to undermine the transmission of force. Their presence reduces the direct interactions of rigid molecules with one another and with their aggregates to lower viscosity. Oxidation during RTFO ageing tends to increase the concentration of polar species, particularly asphaltenes, with greater molecular interaction and association of polar components35 that can facilitate transfer of momentum over longer distances. The consideration of molecular flexibility of WVO in affecting viscosity is similar to a recent description of bio- oils as containing highly “mobile” species that have the potential to soften binder blends.35

Reduced viscosity might be thought of as a result of a “molecular lubricant” between the stiff components of the binder. Xu et al. carried out molecular dynamics simulations of their own selection of model compounds to investigate diffusion of a rejuvenator into the binder with RAP species.36 Interestingly, radial distribution functions show that, with the rejuvenator, saturates interposed themselves between asphaltenes where they were absent otherwise. This further supports the theory of molecular lubricants.

**Dynamic Shear Rheometer.** The rutting parameter, G*/sin δ, from the DSR test is of interest as the addition of oil is expected to soften the binder. The rutting parameters of unaged virgin binder and unaged binder blends containing RAP binder or WVO compositions are shown in Figure 3. As expected, the virgin PG 64-22 binder satisfied the minimum requirement of 1 kPa at 64 °C and the addition of RAP binder increased the stiffness significantly. For instance, at 64 °C, the G*/sin δ of the unaged virgin PG 64-22 binder was 1.5 kPa, while the unaged binder blends containing 40 and 60% RAP binders exhibited much higher G*/sin δ of 6.0 and 10.3 kPa,
respectively. As a result, the high-temperature Superpave grade of binder blends containing 40 and 60% RAP binders was found to be PG 70; see Figure 4. The effect of increasing RAP with WVO was much more profound on the rutting resistance parameter than on viscosity. Incorporation of the 1 and 11 day used WVO in the 40% RAP binder blend lowered the high-temperature performance grade to PG 58, while the 60% RAP binder blend with 1 and 11 day used WVO had a high-temperature PG of PG 64, with a value for $G^*/\sin \delta$ less than that of virgin binder. Thus, increasing the amount of RAP binder improved the rheological properties of the RAP binder blend for the level of WVO used. Mirhosseini et al. found similar trends for 10−30% RAP binder in a PG 64-22 binder rejuvenated with unused date seed oil. After RTFO ageing, the rutting parameter was found to increase for all of the binder blends (Figure 5). Similar to unaged binder blends, the addition of 10% WVO to the 60% RAP binder blend was found to exhibit a similar high-temperature PG as that of the control binder, i.e., PG 64. At 64 °C, the rutting parameter for the virgin binder increased to 4.1 kPa with the addition of 60% RAP binder, which reduced to values of 2.6 and 2.3 kPa when blended with 1 and 11 day used WVO, respectively. The results indicate higher susceptibility to rutting for asphalt mixes containing the 40% RAP binder blend with 10% WVO than that of the control mixes.

The ageing of the WVO was found to have little influence on the value obtained for the rutting parameter. Results were observed to be similar for the 1 or 11 day used oil for both 40 and 60% RAP binder blends. Appearance of the oils was also similar (Figure 6). Both samples were transparent, with the 11 day oil slightly darker. Little prior work on the age of the WCO has been reported. Zhang et al. examined a rejuvenated binder with vegetable oil aged for 2, 4, 6, 8, 10, 12, 14, and 16 h. According to Zhang, the performance of the vegetable oil as a rejuvenator deteriorated with time of its use. Azahar et al. collected WCO samples from a restaurant during the months of April, August, and December to represent “different frequent duration times WCO was used”. However, the actual length of time in service was not specified in their article, which presented an acid value instead and emphasized modification of the WCO by transesterification to improve the rheological properties.

Intermediate performance temperatures of the binder blends were determined by examining the fatigue parameter ($G^*/\sin \delta$) after long-term ageing by PAV (Figure 7). According to Superpave specification, the value of $G^*/\sin \delta$ should be equal to or less than 5000 kPa at an intermediate temperature. The intermediate temperatures for the virgin binder, 40% RAP binder blend, and 60% RAP binder blend were found to be 12.6, 16.8, and 17.6 °C, respectively. The effect of adding 10% WVO on the fatigue parameter of 40 and 60% RAP binder blends was found to be significant. At 25 °C, the $G^*/\sin \delta$ value for the virgin PG 64-22 binder was found to be 2110 kPa, which increased to 2770 kPa with the addition of 40% RAP binder. However, with the addition of 10% of 1 and 11 day used WVO, the $G^*/\sin \delta$ value reduced to 395 and 369 kPa, respectively. Similarly, the $G^*/\sin \delta$ value for the 60% RAP binder blend was found to reduce from 2966 to 432 kPa and
The WVO lowers the viscosity of the RAP-modified binder blends significantly, which was lower or equivalent to the viscosity of the virgin binder. A lower viscosity enables the effective coating of the aggregate, permits lower mixing and compaction temperatures at the HMA plant and during compaction on site, and reduces fuel requirements and emission of volatile organics. Therefore, it can be said that using WVO helps make the asphalt mix production an environmentally sustainable green technology.

However, this study indicates that the WVO mitigates the impact of using RAP on the rutting resistance of the virgin binder. The rutting resistance of the RAP-modified binder blends was equivalent and, in some cases, slightly lower to the rutting resistance of the virgin binder when used with WVO. Therefore, a high percentage of RAP can be used in the WVO-modified binder blend that helps retain the high-temperature performance grade of the virgin binder without forfeiting the intermediate- and low-temperature performance grades.

Different periods of use of the WVO, such as 1 or 11 day used oil, seem to make little difference on the resistance of RAP-modified binder blends to rutting, fatigue cracking, and low-temperature cracking. A molecular lubrication theory provides a basis for computational modeling to improve the selection of WVO components.

Recommendations. Nearly all RAP is reused in some fashion, though much of it goes to purposes other than pavement. While 100% recycle appears possible, it can be expected that transitioning to higher RAP compositions will occur stepwise as evaluations in the field proceed and adjustments to design and implementation occur. This will create more opportunities for the development of WVO as a rejuvenator, which has been deemed feasible as a rejuvenator. Further studies, however, should be conducted using the virgin binder with RAP binder to determine whether the results of this work apply broadly to other asphalt binder grades and sources of WCO.

A number of challenges exist before widespread use of waste cooking oils as rejuvenators becomes feasible. Current HMA processing equipment may require modification to handle larger amounts of RAP. For widespread use, it will be necessary to establish a collection network and the infrastructure for handling and storage of WCOs. Some work has already been done on the gauging quality of waste cooking oil. Additional research is needed to develop standards of testing to ensure suitable sources of WVO with some consistency. Design guidelines will help to ensure Superpave rutting resistance, and other criteria are met when using WVO with RAP binder blends. Moreover, field tests must be run to see how well WVO functions in actual pavements with respect to moisture-induced damage. While there are challenges, the potential benefits for sustainability with WCOs mean addressing the above should be worthwhile.

## MATERIALS AND METHODS

**Recycled Asphalt Pavement (RAP) Binder.** The simulated RAP binder used in this research was prepared by ageing a manufacturer-specified PG 64-22 virgin binder using an RTFO (AASHTO T 240) followed by PAV (AASHTO R 30). This material serves as a substitute for the RAP binder that would be produced from the recycling of asphalt pavement. The benefit to using a simulated RAP binder is that the ageing methods are controlled, so samples can be prepared in a more consistent manner in an effort to lower the
large variability that can occur from natural ageing, which could skew results.

**Waste Vegetable Oil (WVO).** Mel-fry essential oil, which is composed of cotton seed and canola oil with antioxidants, was collected from fryers at the University of Oklahoma’s Couch Restaurant that operate at 350 °F (176.7 °C) for usually 10–12 h per day. This vegetable oil is a combination of cotton seed (54% linoleic acid, 26% palmitic and stearic saturated acids, and 19% oleic acid) and canola oil (62% oleic acid, 18.6% linoleic acid, 9% α-linolenic acid, and 7.4 saturated acids). Oil samples were collected daily from the same fryer for 14 days. Oil samples used in this study were 1 and 11 day used oil. As the oil level of the bath got low, more oil was added throughout its total life span. Thus, the oil collected was not always 100% oil that started at the beginning of collection; however, the effects of this were considered negligible, as the oil level did not drop dramatically day to day. Thus, the majority of the oil collected was indeed used for the amount of time indicated.

**Sample Preparation.** Virgin PG 64-22 binder blends containing 0, 40, and 60% (by weight of the virgin binder) simulated RAP binder were prepared. For each of the RAP composition, both 1 day used and 11 day used WVO (10% by weight of the virgin asphalt binder) blends were prepared (Figure 8). For blending purposes, different protocols have been used by researchers. For example, Zhang and Yu, Baldino et al., Rani et al., and Ghabchi et al. used HSM for blending asphalt binder and additives at a rotational speed that varied from 500 to 5000 rotations per minute (rpm). Accordingly, in this study, a high shear mixer was used to prepare the asphalt binder and additive blends at a rotational speed of 1000 rpm for 45 min at 145 °C. A higher temperature such as 160 °C was used when blended the virgin PG 64-22 binder with the RAP binder because of its high stiffness.

Each of these blends was aged in the laboratory with methods that accelerate the oxidation of binder samples. Short-term ageing to simulate ageing from the mixing and paving process was achieved by performing the RTFO ageing of samples at 325 °F (163 °C) with 4 L/min air flow for 85 min (AASHTO T 240-13). Long-term ageing to simulate approximately 5–7 years of oxidation was achieved by conducting the PAV-ageing at 100 °C with a compressed air of 305 psi for 20 h (AASHTO R 28-12). The binder was heated and poured into molds to get a smooth surface and then cooled at room temperature. WDS was performed in the Samuel Roberts Noble Microscopy Laboratory at the University of Oklahoma, where samples were sputter-coated and then loaded into a JEOL JSM-840 SEM equipped with a Kevek X-ray analyzer with IXRF software. The system provides quantitative analysis of low-atomic-weight elements on the sample surface.

**Dynamic Shear Rheometer (DSR) Test.** Rheometric properties were tested using DSR following the AASHTO T 315-12 test method at 61, 64, and 67 °C temperatures. For some samples, it was necessary to make measurements at lower temperatures to determine the high-temperature performance grade (PG). The rutting parameter ($G^*/\sin \delta$) was calculated from the experimental values of the complex modulus ($G^*$) and the phase angle ($\delta$). Also, DSR tests on PAV-aged binder samples were performed at intermediate temperatures of 22, 25, and 28 °C to determine the fatigue parameter ($G^* \sin \delta$). This method of testing allows the evaluation of the performance of the rejuvenated RAP samples as a result of using WVO.

**Bending Beam Rheometer (BBR) Test.** The BBR tests on PAV-aged asphalt binders were conducted by following the AASHTO T 313-12 test method. The test was conducted at two different temperatures, namely, −21 and −24 °C for the WVO-modified binders and −9 and −12 °C for the virgin binder with and without RAP binder. Creep relaxation ($m_{60}$) and stiffness ($S_{60}$) values, measured at 60 s after load application, were used to find the low-temperature PG of the binder blends.

## AUTHORS INFORMATION

### Corresponding Author

Edgar A. O’Rear – School of Chemical, Biological and Materials Engineering, The University of Oklahoma, Norman, Oklahoma 73019, United States; orcid.org/0000-0001-9565-3343; Email: eorear@ou.edu

### Authors

Connor R. Dugan – School of Chemical, Biological and Materials Engineering, The University of Oklahoma, Norman, Oklahoma 73019, United States

Chris R. Sumter – School of Chemical, Biological and Materials Engineering, The University of Oklahoma, Norman, Oklahoma 73019, United States

Shivani Rani – School of Civil Engineering and Environmental Science, The University of Oklahoma, Norman, Oklahoma 73019, United States

S. Ashik Ali – School of Civil Engineering and Environmental Science, The University of Oklahoma, Norman, Oklahoma 73019, United States

Musharraf Zaman – School of Civil Engineering and Environmental Science and Southern Plains Transportation Center, The University of Oklahoma, Norman, Oklahoma 73019, United States

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.0c00377
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