Evaluation of Graphite Nanoplatelets Influence on the Compaction Properties of Asphalt Mixtures

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Introduction

In recent studies, it has been shown that the addition of small amounts of Graphite Nanoplatelets (GNPs) to asphalt binders can improve the cracking resistance of asphalt binders and mixtures at low temperatures. However, another interesting results was the significant reduction in the number of gyrations required to achieve a target air-void ratio in gyratory compaction, as shown in **Figure 1.**

Surprisingly, viscosity measurements indicated that the addition of GNPs increased the viscosity of the binder. Such discrepancies between the binder viscosity and the mixture compaction behaviour have been already observed by other authors, who pointed out the drawbacks of an experimental approach based only on the study of viscosity. Other authors have also shown that the mixture compactability does not improve linearly as the temperature increases, but, on the contrary, it gets worse above a certain level of temperature, although viscosity decreases progressively with temperature.

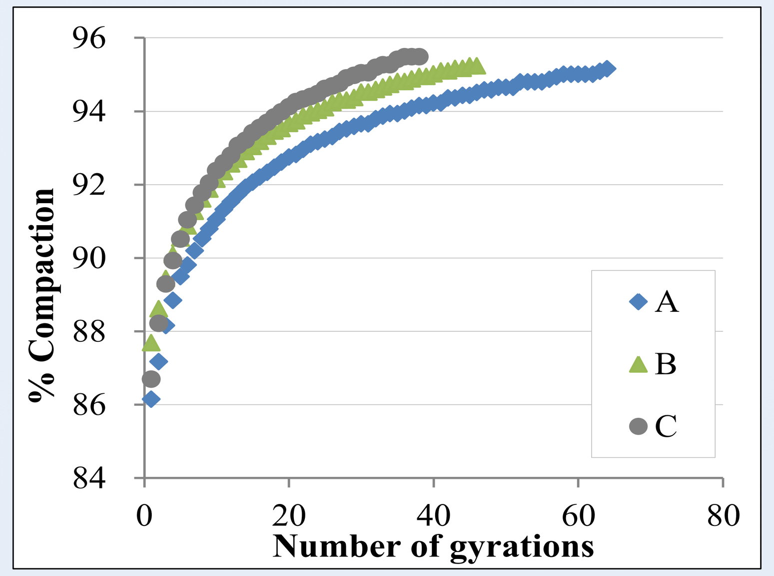

FIGURE 1. Compaction curves for mixtures with different GNP compositions; A has no GNP, B has 6% GNP, C has 8% GNP

Most likely, a different mechanism is responsible for the reduction of compaction efforts. In this research, an approach based on tribological testing is used to characterize the lubricating behaviour of asphalt binders with GNPs.

Objectives

- To examine the effect of GNP on lubrication properties of asphalt binders to investigate if the lubrication properties of GNP asphalt binders are responsible for the enhanced compactability observed in GNP modified mixture (Le et al., 2016).
- To develop a tribological testing method to evaluate the lubricating behaviour of binder between rough surfaces similar to the surfaces of natural aggregates.

Mechanism of Friction and Lubrication

In tribology, the lubrication properties of a material placed between two solids in relative motion is normally described through the Stribeck curve (**Figure 2**), which shows the evolution of the coefficient of friction μ as a function of the sliding speed. The change in the coefficient of friction values is due to the variation of the thickness of the lubricating film, as shown in **Figure 2**.

The role of nanoparticles in friction reduction has been investigated by many researchers and the mechanism involved can be described as follow: rolling effect, protective film, mending effect, and polishing effect.

For the effect of GNP on lubrication of binder, a phenomenon similar to the mending effect is expected to occur, as hypothesised in a previous study (Ingrassia, et al., 2019), and schematized in **Figure 3.**

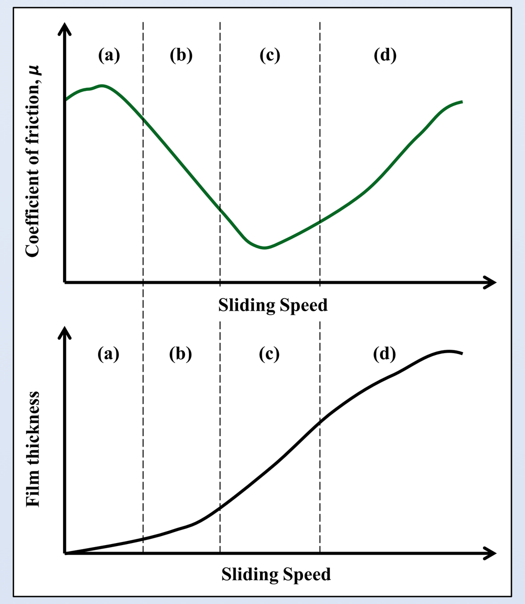

FIGURE 2. Stribeck Curve.
The Stribeck curve can be generally divided into four regions, which correspond to different regimes of lubrication:
- **the *boundary regime* (a), occurring when the lubricating film is thin and, consequently, a high μ is determined by the strong interaction between the asperities of the solids;**
- **the *mixed regime* (b), where a reduction of μ occurs, because of the increased thickness of the lubricating film, which reduces the direct contact between the solids;**
- **the *elasto-hydrodynamic regime* (c), in which the minimum μ is reached, because the thickness of the lubricating film is able to completely separate the solid surfaces;**
- **the *hydrodynamic regime* (d), where the film is so thick that there is a new increase of μ, depending on the viscous drag of the lubricant.**

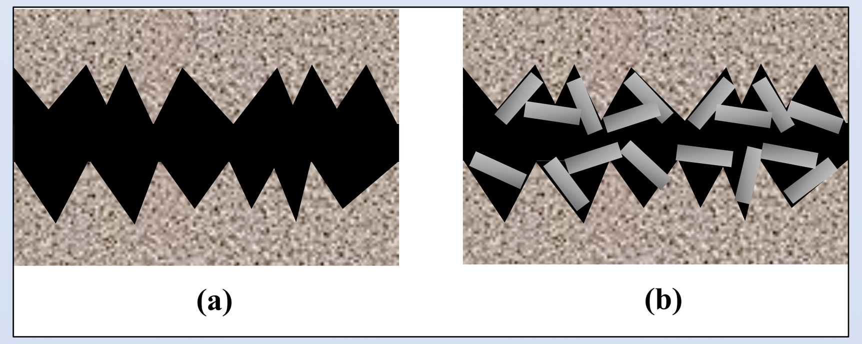

FIGURE 3. Scheme of the asphalt binder film between aggregate surfaces: (a) without GNPs; (b) with GNPs (Ingrassia, et al., 2019).
GNPs could place between the asperities of the aggregates, providing overall a reduced roughness and therefore an improved lubrication with respect to the base bitumen

Materials And Methodology

A plain PG58-28 bitumen was used as base binder. A GNP made of a synthetic graphite material with 99.66% carbon and 0.34% ash, characterized by an enhanced surface area equal to 250 m2/g, was added to the asphalt binder in two proportions: 3% and 6% by weight of the binder. Both viscosity and tribological tests were conducted to study the viscous and lubricating behavior of binder, respectively. In the tribological tests, both smooth and rough substrates were investigated.

Viscosity Tests

At Nynas, the tests were performed using the DSR cone and plate geometry, characterized by a radius of 20 mm and a slope of 2°, while in Minnesota viscosity was obtained with a Brookfield viscometer equipped with the standard SP 27 spindle. The same temperatures as tribological tests (110°C, 130°C and 150°C) were investigated. All testing performed at Nynas was done using an Anton Paar DSR equipment. **Figure 4** shows the viscosity results obtained using the cone and plate geometry. Similar results were obtained at the University of Minnesota by using a Brookfield viscometer. The increase in viscosity (with respect to the control bitumen) due to the addition of GNPs is approximately equal to 15% and 30% for the binders with 3% GNP and 6% GNP, respectively, at all testing temperatures. As expected, the viscosity values decrease with the increase in temperature. The Non-Newtonian shear-rate dependence of viscosity was only observed for the binder with 6% GNP at 110°C.

These results confirm that the improvement in the workability of GNP mixtures cannot be explained by a reduction in viscosity.

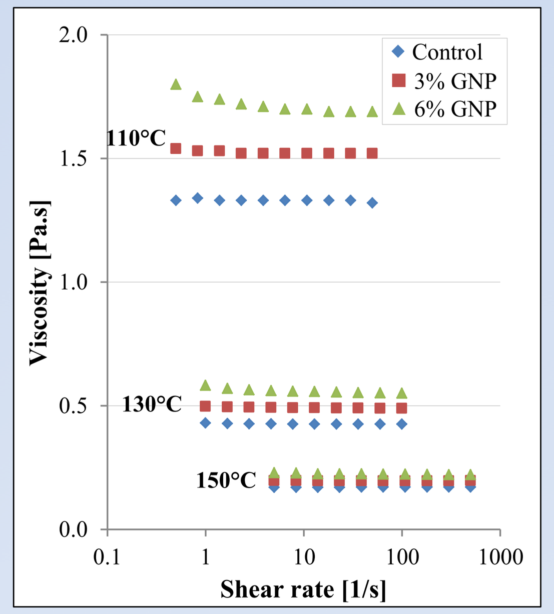

FIGURE 4 Viscosity Results (Nynas)

Tribological Tests with Smooth Surfaces

Tribological tests were performed using a ball-on-three-plates fixture mounted on a Dynamic Shear Rheometer (DSR), as shown in **Figure 5 and 6.** All tests were carried out with smooth steel ball and steel plates as substrate.

In order to simulate as much as possible the typical compaction temperatures for hot mix asphalt (HMA) and warm mix asphalt (WMA) mixtures, 110°C, 130°C and 150°C were considered as testing temperatures. During the tests, the axial force F_N was kept constant and equal to 10 N, while the rotational speed was increased in logarithmic steps from 0.1 to 1433 rpm.

The coefficient of friction μ is determined according to:

$$\mu = \frac{F_{F-TOT}}{F_{N,tribo-TOT}}$$

where F_{F-TOT} and $F_{N,tribo-TOT}$ are, respectively, the total friction force and the total normal force experienced by the specimen

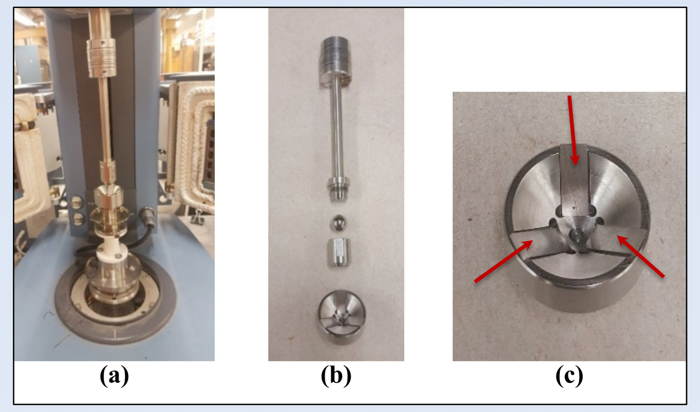

FIGURE 5 Tribological test fixture: (a) general view; (b) components of the fixture; (c) lower cup and testing plates

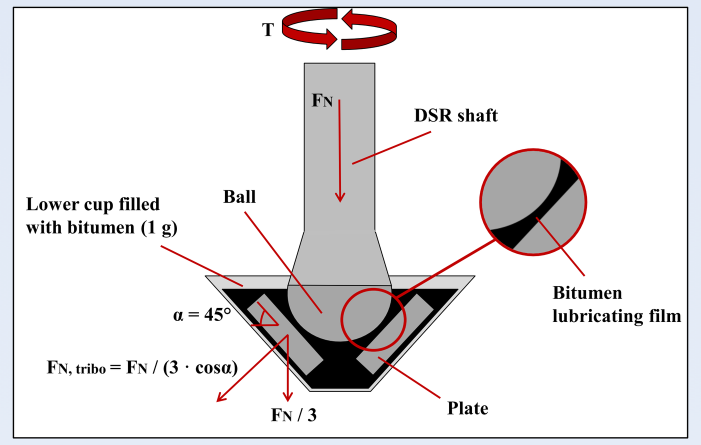

FIGURE 6 Scheme of the tribological test

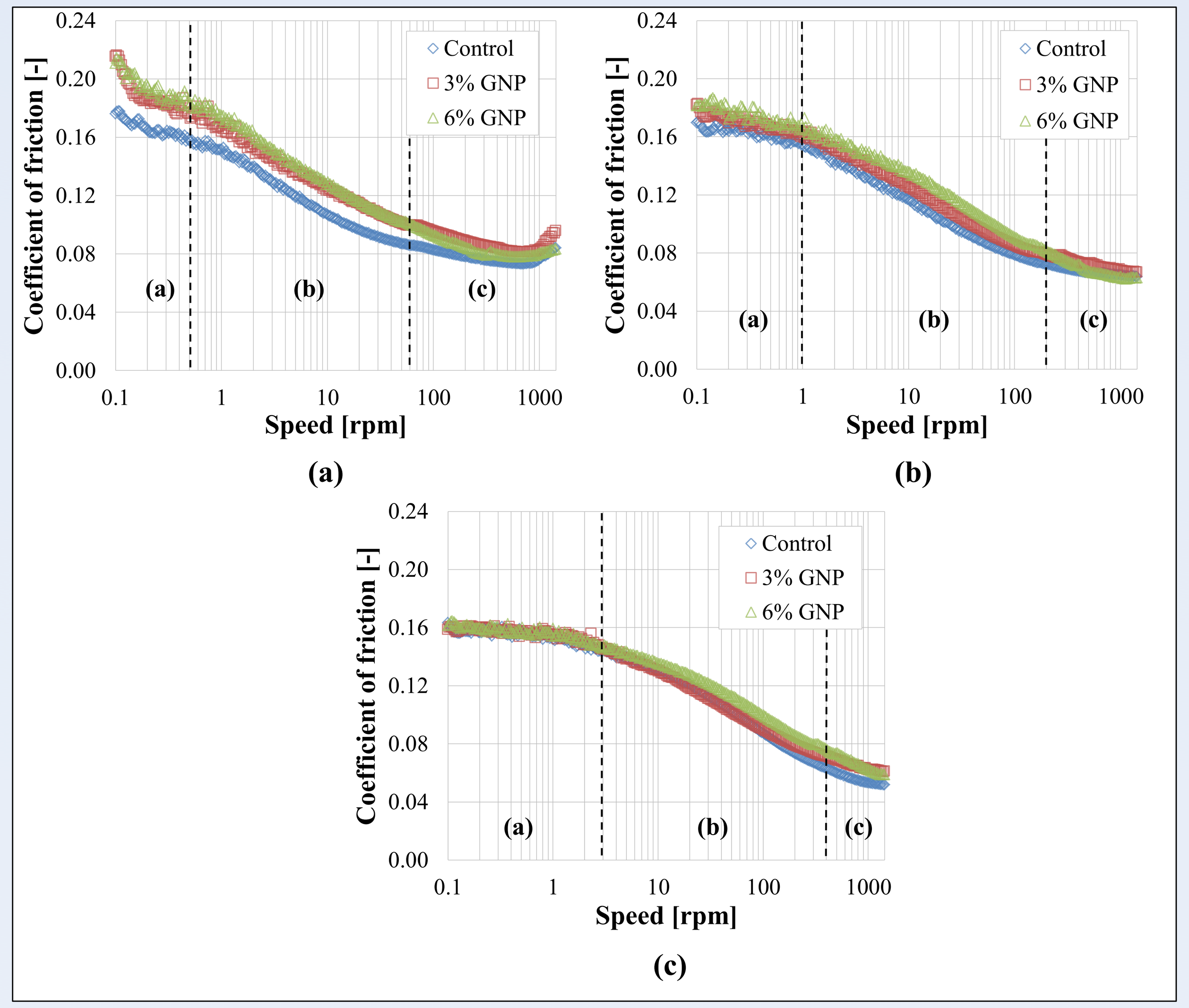

FIGURE 7 Tribological test results with smooth surfaces: (a) 110˚C; (b) 130˚C; (c) 150˚C

The tribological results obtained with smooth substrates are shown in **Figure 7**.

- A general increase in the coefficient of friction with the increase of GNP is observed for all temperatures.
- A general decrease in the coefficient of friction with the increase of temperature (or decrease of viscosity) is observed for all GNP dosages.

In summary, the possible improvement of the lubrication properties due to the addition of GNPs cannot be observed from the smooth surfaces tribological test.

Tribological Tests with Rough Surfaces

To consider the surface roughness of aggregates, the ball and of the plates were roughened to better simulate the surface of the aggregates.

The method consisted in immersing the ball and the plates in hydrochloric acid (HCl) for three days. Hydrochloric acid corroded the surfaces of the parts and made them rough and looking like an orange skin. **Figures 8(a)-(b)** present the original smooth ball and plate, whereas **Figures 8(c)-(d)** present the ball and plate after they were removed from the acid.

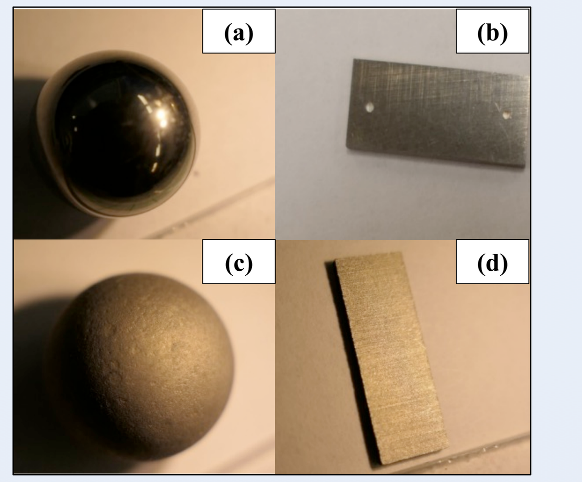

FIGURE 8 (a) smooth ball; (b) smooth plate (used); (c) rough ball; (d) rough plate

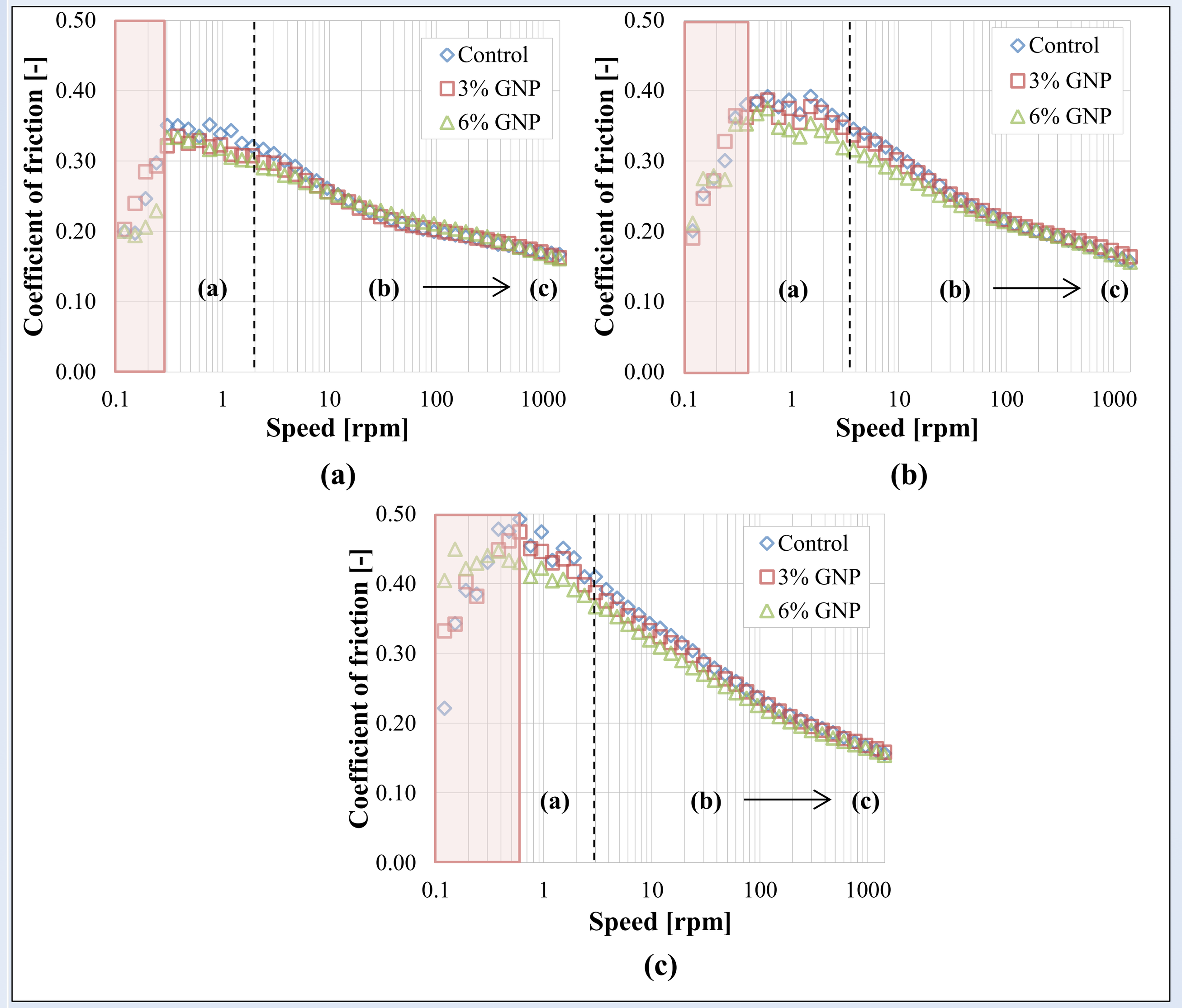

FIGURE 9 Tribological test results with rough surfaces: (a) 110˚C; (b) 130˚C; (c) 150˚C

The results of the rough surfaces tribological test are shown in **Figure 9**.

- Values of μ measured at low speeds (in the red box) should be neglected, due to the sliding of the plates in the lower cup.
- At all temperatures, the minimum of the Stribeck curve is not reached, probably because under rough surface conditions the complete separation between the solid asperities is harder to be achieved.
- contrarily to smooth substrates test results (**Figure 7**), the μ remarkably increases as the temperature increases, especially in the boundary regime (a).
- At all three temperatures, μ is reduced by the addition of GNPs in the boundary (a) and mixed (b) regimes. Conversely, μ are almost the same for all blends once the speed increases to the elasto-hydrodynamic (c) regime

In summary, the decrease of μ as the increase of GNP at slow shear rate provides a possible explanation for the reduced compaction effort due to the addition of GNP observed in previous studies (Le, et al., 2016).

Conclusions

- The viscosity of the binder increases with the quantity of GNPs. This observation confirmed that the reduced compaction efforts for GNP asphalt mixtures cannot be attributed to the reduction in the viscosity of the binder.
- From the smooth surfaces tribological tests, it was found that GNPs do not improve the lubricating behaviour of the binder in the case of smooth substrates. Conversely, the rough surfaces tribological test shows that the lubrication properties of the binder were progressively improved in the boundary and mixed regimes as the GNP amount increased.
- Since the rough substrate mirrors the actual aggregate roughness more accurately than the smooth substrate, the enhanced workability of GNP modified mixtures can be attributed to the fact that GNPs may occupy the space between the asperities of the aggregates, reducing the overall roughness and thus improving the lubrication.
- The tribological tests performed with rough substrates demonstrated that, for a given binder, friction increases significantly as the temperature increases (i.e. the viscosity decreases), especially in the boundary regime. This finding once again confirms that the viscosity is not the only parameter involved in the compaction of asphalt mixtures. Increasing temperature (decreasing viscosity) may not always be beneficial for the compaction of the asphalt mixture.

Acknowledgements

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Main references:

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