Modification of Bitumen Using Sustainable Materials for Pavement Design

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Abstract. Construction materials will be in the extinction stage in the future. Use of the waste materials to ensure environmental protection is the need of hour. For the construction of essential infrastructure like flexible pavement requires bitumen. Bitumen obtained as a petroleum refinery by-product used for construction of flexible pavement is on the verge of extinction, since natural oil petroleum is a non-renewable resource. Again low life of bitumen due to oxidation is another problem. Hence, it is important to use potential modifiers to partially substitute and enhance the life of bitumen and flexible pavements. In this paper use of numerous modifiers and their possible effects on bitumen and bituminous mix strength and toughness are described by researchers. Different modifiers such as polypropylene, Nano montmorillonite, Nano-silica, Nano-clay, Low-Density Polyethylene (LDPE), Styrene Butadiene Styrene (SBS), High-Density Poly-Ethylene (HDPE), Carbon Nano-Tubes (CNT), graphene, Graphene Oxide (GO), fly ash, cloisite, evotherm, Cecabase Rt, Nano fill, rubber waste, crumb rubber etc. were used for modification of bitumen. Binder proportions are approximately similar on the other hand percentages of modifiers used differ mostly by researchers. It is evident from the literature that the optimal proportion of modifier would only yield improved performance. However, the quality of the modifiers shows a key part, to fulfil the requirements of the ideal pavement and increased life carefully chosen based on the soil conditions and environmental circumstances.

1. Introduction
Sustainable construction materials are the necessity of the era. As a replacement for using all available natural resources of construction material like bitumen leads to focus of researchers on modification of the bitumen and evaluation of the strength and durability of modified bitumen an essential area of research. The strength and longevity improvement without environmental effects makes bitumen modification an essential element of material research. The bitumen modification would also bring a significant reduction in costs from construction point of view if partly replacement of bitumen with other enhancing the properties with addition of additives possible. Hence, the purpose of this paper is to elaborate various bitumen-modifiers evaluated by the researchers.

1.1 Need for modification of Bitumen
Yousefi [1] emphasized on the inadequacy of that pure bitumen for modern roads and growing up traffic, forcing the engineers to bitumen modification for enhancing performance throughout service life. The altered binder is steadier and demonstrates less susceptibility to permanent deformation in hot climate.
under heavy traffic, braking and accelerating forces. It counterattacks the fatigue loads and has an
excellent bond between binders and aggregates. Therefore, the material which can change the chemical
configuration of bitumen can be a modifier. Modified bitumen has more adhesion at execution
temperature than pure bitumen and lower thermal sensitivity in the range of operation temperatures and
adequate viscosity. Furthermore, its sensitivity to the loading time is poor, and its resistance to plastic
deformation, fatigue, and cryogenic cracks are high. Ultimately, the properties are perfect for efficiency
and service after aging. Section 2 of the paper describes the materials used in modification; section 3
demonstrates the literature study for different modifiers used in bitumen followed by conclusion in
section 4.

2. Materials used for Modification

2.1. Bitumen
Bitumen is described as a virtually non-volatile at ambient temperatures and softens gradually when
heated, with adhesive and water resistant properties originated by means of fractional distillation from
crude oil or available in natural asphalt lake, totally or else almost fully soluble in toluene, and very
viscous or almost hard at ambient temperatures. This is well known that the original characteristics of
bitumen, as well as the characteristics of bitumen crude oil, are highly dependent on its production and
processing method. Higher quality crude oils and proper processes of distillation will enhance the
bitumen properties. The heavier crude oil typically produces greater bitumen yields. Therefore, it is of
utmost importance to have adequate information on the bitumen characteristics from various aspects.
This knowledge proves to be more significant as other difficulties such as phase discontinuity, poor
dispersion, and instability with polymers/additives make it challenging in the manufacture and
application of bituminous materials for certain bitumen applications.

2.2. Polymers
After its use, the polymers are thrown into the motherland as waste which obstructs rainwater infiltration
owing to its non-biodegradability. Nearly 70 % of the plastic manufactured is most frequently used as
waste, including single use, for instance carrying bags, etc.
From the reports of Central Pollution Control Board, waste plastics are generated 15342 tons/day in
India and are 5.6 million tons per annum. Consequently, numerous polymer forms like Low-Density
Polyethylene (LDPE), Polypropylene, and High-Density Polyethylene (HDPE) are used as modifiers
for bitumen. The application of these modifiers safeguards the bituminous mixture along with the
underneath layers by inhibiting water percolation.

2.3. Used/waste Rubbers
Thanks to the additive and fire retardant material present in tires, tires are thrown out in the atmosphere
polluting the soil for the use of tires. Those emit methane gases when burned. Overall, fifteen million
tons of scrap tires are manufactured annually, of which India contributes one million tons. Thus, we
have to use those tires technically for our requirements without making any impact on the environment.

2.4. Clay and Nano clay
Clays are mineral deposits that occur naturally and exhibit element inconsistencies. Naturally obtainable
clay is significantly decreased to Nano size to accomplish great surface area in order to adhere to the
asphalt and bind together with the aggregates. The Nano clay alteration when applied even in small
amounts, yields multi-folds of improvement.

2.5. CNT and Graphene
Carbon nanotubes (CNTs) consist of cylinder-shaped molecules, together with single-layer rolled-up
carbon atom (graphene) sheets. This can be single-walled (SWCNT) or multi-walled (MWCNT), with
both the SWCNT having a diameter of a lesser amount of 1 nanometer (nm), and MWCNT comprising
of several dense nanotubes approaching more than 100 nm in diameter. Its dimension can exceed a few
micrometers or even millimeters. Due to the immense physical properties graphene seems to have become a new super-material. Due to its lower size, super strength with enormous mechanical, thermal, electronic, electrical, and optical properties graphene is actually the most striking nanomaterial. It has high specific surface area, chemical stability, optical transmittance, elasticity, porosity, biocompatibility, adaptable band gap, and simple chemical functionality that basically help to modify its properties. Both of these materials were dispersed with bitumen for improvement of its rheological characteristics, viscosity, softening point, penetration and resistance towards long term as well as short term aging. These two Nano modifiers help the binder to resist moisture susceptibility, which makes the pavement stronger and durable [1-36].

3. The Study of Literature

3.1. Polymers

The plastomeric and elastomeric polymer of both forms can be used in asphalt. Asphalt has many applications for paving, maintaining seals such as filling, patchwork, recycling, and slurry seal. For all of these uses, polymer evolves to blend with bitumen. Prasad K et al. [15] recommended that the binder must first be heated to 260° C–280° C after it should be added to waste plastics, or else the blend will not be accurately mixed. And if this occurs, the binder may likely catch fire before it hits its point of fire and a loss of bitumen weight can also happen. Yazdani et al. [5] added polypropylene, Styrene-Butadiene Styrene (SBS) elastomer, Nanoclay and plastomer. All the compressive strength of changed bitumen and the softening point are improved by 55%. Among the various percentages tested, it was calculated that 3 % (SBS), 5%polypropylene (PP), and 1.5 % nano-clay (NC) gave a better quality outcome in improving the properties. Vamshi [12] established that a 10 % plastic-modified bitumen stripping value was zero compared to 0.4 % virgin bitumen. There is also a 7.99 % reduction in material costs by using plastic waste on the highway. As compared with regular bitumen, modified bitumen reduces penetration.

Rajasekaran et al. [20] established thermal analysis characteristics using the thermogravimetric examination demonstrates that polymers such as polyethylene, polypropylene and polystyrene are becoming softer from 130 °C – 140 °C lacking gas growth. It is harmless when used below 150 °C with molten waste plastic. The increasing order of strength also for the modified bituminous block is polystyrene, polyethylene, and polypropylene. In their paper, Dixit et al. [6] indicated that a limit of 0.6% of plastic fiber modification was only sufficient. The stability of the Marshal increases by 5.4 % with a binder material. Rokday et al. [19] reported a 160° C mixture of the waste plastics with hot bitumen. Plastic bitumen alteration improved binder melting point. Thermoplastics, thermosets and elastomers were used as modifiers. They have also learned the bitumen does not leach the polymer. Kazmi et al. [29] has shown that 9 % of LDPE by bitumen weight is an efficient binder ratio. They received a 32.5 % increase in the stability performance relative to sleek bitumen. Void in mineral aggregates was 5 %. Up to 66.7 % of the estimated air voids. In their analysis, Menaria et al. [22] found that 8 % of waste plastic is the optimal quantity to be used to alter bitumen. They suggested plastic roads with high temperature zones around 50°C can also be laid. The binder's optimum value was 4.5 %. The dry method is observed for this kind of work.

Sahu et al. [18] observed the change in bitumen and indicated that polymer has an excellent adhesion characteristic, so there are zero stripping values in conventional bitumen for almost 72 hours against 5 % for 24 hours. For 10 % of plastics, moisture sensitivity of polymer altered bitumen covered aggregates is determined to be 0.12 %; the value for regular bitumen is 4 %. Cheshmehgol et al. [35], 2 to 14 % LDPE specimens prepared. They reported that indirect tensile strength rises by 15.2 % for unconditioned specimens, and is 23 % higher for conditioned specimens. The tensile strength ratio for unconditioned samples is 88 % and 94 % with a 10 % LDPE alteration. Singhal et al. [14] determined that overall material costs are reduced by 7.99 % when bitumen is replaced with waste plastic (6 %), and the Marshall stability value reached is 1161 kg. Once the plastic content has increased, the value of ductility and penetration decreased and the value of softening points increased.
Table 1. Percentage of modifiers used by different authors

| Percentage of modifiers | Type of modifier                  | Author                      |
|-------------------------|----------------------------------|-----------------------------|
| 1,2,3,5,10              | Crumb rubber                     | Amit Kumar Sahu             |
| 10                      | Plastic waste                    | Avula Vamshi                |
| 2,4                     | Polymer                           | Ali Jamshidi                |
| 4,6                     | Nano silica                      |                             |
| 0.1,0.5,1               | Carbon Nano tubes                | Ezio Santagata              |
| 2,4,6                   | Nano silica                      | Farhad zafari Rhi           |
| 2,4,7                   | Cloisite-15A                     | Ghaffarpour                 |
| 2,4,7                   | Nanofil-15                       |                             |
| 3,6                     | Cloisite                          | Van de ven                  |
| 6                       | Nanofil                           |                             |
| 3,6                     | Organic MMT                      | Jeroen Besamusca            |
| 2                       | Natural rubber                    | Krishnapriya                |
| 0.3, 0.4                | Evotherm                          | Manjunath                   |
| 0.3, 0.4                | Cecabase Rt                       |                             |
| 3,7,9                   | Organic MMT                      | Mahdi Lamya                 |
| 1,2,3,5,10              | Waste plastic                     | Minakshi Shingal            |
| 2,4,6,8,10,12,14        | LDPE                              | Mehdi Karimi                |
|                         |                                  | Cheshmehgol                 |
| 4,6,8                   | LDPE                              | Prakash somani              |
| 2,4,6                   | Crumb rubber                      |                             |
| 6,8,10                  | HDPE                              | Rajeshkumar                 |
| 5,7,9,11                | PE                                | Rao                         |
| 2,4,7                   | Montmorillonite                   | Saeed Ghaffarpour           |
|                         |                                   | Jahromi                     |
| 5,10,20,25              | Polyethylene (PE), Polypropylene (PP), Polystyrene (PS) | S Rajasekaran |
| 0.1,0.2,0.3,0.4,0.5,0.6,0.7,0.8,0.9 | Plastic bag fibres               | Sandhya Dixit               |
| 2,4,6,8,10,12           | Polyethylene Teraphthalate, LDPE, HDPE | Shiva Prasad               |
| 10,20,30,40             | Waste plastic                     | Shwetha Rokdey              |
| 7                       | Nanofil-15                        | S. Ghaffarpour Jahromi      |
|                         | Cloisite-15A                      |                             |
| 3,6,9,12                | Fly ash                            | Saad Issa sarsam            |
| 1,2,3,4                 | Silica fumes                      |                             |
| 1,2,3,4,5,6,7,8,9       | Waste polyethylene, waste crumb rubber | Yuqiao Yang                |
| 6,8,10,12,14,           | PE,PP,PS                          | Yash Menaria                |
| 2, 4                    | Nano montmorillonite              | Zhanping You                |

3.2. Rubbers
Rubber is collected from unwanted tires which are not usable in cars. As we go for the use of waste material, as our material costs are raising and we are paving the way also for environment cleaning. The adjustment can be applied to both crumb rubber and natural rubber. This can be put in to bitumen in powdered form. Rubber adsorbs the bitumen's aromatic substance, and therefore covers one another.
Kumar et al. [37] adjusted bitumen by crumb rubber of 10 % and found an optimal HDPE content of 8 % for both unmodified and modified bitumen, resulting in an improved Marshall Stability value. The 6 % optimum binder content is decided in their work. Krishnapriya [27] researched the effectiveness of modified mixes made from Natural Rubber. She determined that the optimal binder content was 5%. Fatigue life raised the resilient modulus by 0.9 sec for the rest period. Natural modified bitumen in rubber has outstanding resistance to rutting. Yang et al. [6] found that 8 % of the polyethylene additive was good in penetration associated to the similar percentage of the polyethylene and rubber crumbs compound waste. The softening point was set at 8% for waste polyethylene but the ductility is lower. The crumbs of rubber waste have low temperature resiliency. Somani et al. [21] saw an optimal 6% bitumen content as they reached a 945 kg Marshall Stability value. The stability value falls above 6 percent of the binder. So, the alteration of 8 per cent LDPE and 12 % crumb rubber resulted in a Marshall stability value of 1250 kg and a flow value of 3.86 mm. Manjunath et al. [23] decided to add 0.3 % and 0.4 % of the at a temperature of 130 °C, the great stability is achieved and the stability values improved to 24 % and 22 % for Evotherm, and cecabase grade 2 respectively.

3.3. Nano clay
Besamusca [10] studied two Nano clays of organic montmorillonite (MMT). He stated that the modifiers are influenced by bitumen’s aging characteristics, which may be related to changes in viscous properties due to modifiers being included. Viscosity is reliant on the shear rate, and must be considered highly reliant on temperature. Using Nanofill-15 and Cloisite-15A, Jahromi et al. [32] altered bitumen by 0.2 %, 0.4 %, and 0.7 % more dynamic creep efficiency as compared to unmodified bitumen as showing modified bitumen. Tests related to Nanofill-15A, such as resilient modulus test, dynamic creep test, indirect tensile strength (ITS) test, and fatigue resistance test performed between MMT and cloisite-15A shows good performance. Jahromi et al. [31] analyzed samples at 5°, 25° and 40 °C at a loading frequency of 0.5 Hz. Pulse times were 500 ms and retrieval time was 1500 ms. Modification is accomplished using 2 to 7 % Nano clay. The stiffness of both Cloisite-15A and Nanofill-15 ranged from 8 % to 40 % and 3 % to 18 % respectively, depending on the test temperature and the Nano clay content. Creep tests performed at 40 °C and 60 °C for altered mixes revealed a strong pulse. Van de Ven et al. [11] evaluated the impact of addition of Nano clay modifiers in new and aged bitumen by means of Penetration, softening point and dynamic shear rheometer; cloisite Nano clay enhanced resistance to rutting, ITS, and stiffness for 40/60 bitumen. In both short and long-term aging, Nano fill additive improved resistance to aging for 70/100, bitumen. Jahromi et al. [33] reported that the fatigue-resistant parameter improved by incorporating cloisite in 7% compared with unmodified bitumen 1.2 to 1.4 times in unexploited condition. Modification of Nano fill reduces fatigue life but aging shows the same fatigue life. He also observed that improved bitumen properties depend on the form of clay pre-treatment, Nano clay, and polymer variety.

You et al. [4] used bitumen by weight between 2 % and 4 %. Rotational viscosity grew because of 41 % and 112 % on average. Complex dynamic shear modulus improved by 66 % with the addition of 2 % Nano clay A and 125 % with the addition of 4 % Nano clay A. Similarly, Nano clay B increases 184 % and 196 % of the dynamic shear modulus. Using both the Nano clays (NC) the risk of strain failure can be diminished in the tensile strength test. Yang et al. [7] noticed that both organophilic as well as sodium MMT can enhance resistance towards aging and these alterations have a higher side considering the bitumen quality at low temperatures, where snow susceptibility is present. Mahdi et al. [24] took 3 %, 7 %, and 9 % of organic MMT NC (N3 and N4) adding into bitumen. All N3 and N4 have X-Ray equivalent diffraction properties of 2.28, and 3.8 respectively. Complex shear modulus has evolved in dynamic shear rheometer (DSR) and reduced phase angle for modified binders as opposed to original ones. Thus altered binders have improved shear deformation resistance. Jamshidi et al. [30] recorded a major change in asphalt rheology with different kinds and amount of Nano-materials (2 % and 4 %). Centered on a rutting factor gradient analysis, the critical temperature for the NI44 P2 binder found to be 64 °C. Silica is well recognized for adhesion and the exceptional surface area of nanosilica material and stiffness modulus diminishes temperature sensitivity. Silica fumes improve the softening point and
viscosity when fly ash raises the softening point and reduces viscosity. Rhi et al. [2] observed from their inspection that Nano silica penetration declined after 6% addition, softening point improved and ductility lowered. This may be that more binder is absorbed from a high surface area. Between the several proportions, 2%, 4%, and 6%, it was the 6% nanosilica that soon led the binder not to age.

3.4. CNT and Graphene Oxide (GO)
Earlier work has shown that Nano modifiers provide a beneficial means of improving asphalt material properties. Most of the studies present have concentrated on the mechanical properties of modified asphalt at moderate temperatures. Xiao et al. [8] proposed that carbon nanoparticles may be useful in improving the rheological characteristics of bituminous mixes in the short and long term after aging treatments. Amirkhanian et al. [36] suggested the use of a moderately large proportion of Nano-particles (> 1 per cent) to enhance permanent deformation resistance at elevated temperatures in their work on unaged binders. For some phase angle value of 1 percent CNT binder, Santagata et al. [17] found an unspecified 0.5% mix phase indicating a higher Complex Shear Modulus. Earlier in the Rolling Thin Film Oven Experiment (RTFO), there were fewer differences between the aged and the elderly, but then the Pressure aging vessel came back when a 1% mixture was stiffer than 0.5%. Khattak et al. [28] tried to modify neat bituminous binders with carbon Nano-fibers to a degree that was affected by the mixing technique used to disperse Nano-fibers in the binder. Few studies have investigated the mechanical properties at low temperatures, which are critical in harsh winter conditions for asphalt paving. Some of the most significant challenges to applying advanced Nano-materials to the asphalt industry are the relatively high cost of materials.

Wu et al. [9] showed that GO application would help to boost the resistance of bitumen to ultraviolet along with thermos-oxidative aging because of preventing the aging cycle. Yu et al. [3] inspected the microscopic study of the relationship among the GO and asphalt binder using various methods. This experiment dictated far enough that the addition of GO does not alter gel-soluble arrangement to gel-type arrangement and keeps the asphalt binder step of original maltene solubility. Results of the investigation conveyed by Liu et al. [25] specified that GO application does not alter the glass transfer temperature of the asphalt binder; though, such addition may upsurge the amount of crosslinking to bitumen. They also explored the usefulness of GO’s modified bitumen for pavement applications. In addition to three other additives, Sasobit, Waste Cooking Oil (WCO), and Sasobit + WCO, they followed their tests by applying 0.05% of GO to warm mix asphalt binders (WMABs). After the study, it was established that the GO substantially increased the viscosity, permanent deformation resistance and high temperature elasticity of unmodified bitumen (PG 64-22). Li et al [26] examined the boundary among the GO and bitumen, and the effect of adding GO to the composite output based on asphalt. During the mixing process they said CO₂ gas was released; there was no chemical reaction among the GO and bitumen. The CO₂ was precipitated by the degradation of the GO. It was found that the penetration and ductility of the binder was subsequently diminished by mixing with GO but the viscosity and softening point of the material was increased. After analysis, it also determined that the GO can improve the anti-rutting efficiency at both high and low temperatures, with thermal stability. Table 1 shows the details of work done by various researchers/authors.

4. Conclusions
The following conclusions were drawn from the works of searchers on bitumen modification using waste polymer and rubber.
- Modifiers help in improving the water-resistant property of the binder.
- The main aim of using modifiers is that the pavement bitumen should be less susceptible to the temperature at the operational condition and that workability need preservation at construction condition and the modifiers do this magically.
- The cost of an altered binder in comparison with the conventional binder is reduced.
- Compared to neat binders, modified binders possess improved stability and longevity.
- Modifiers enhance the thermal stability on bitumen alteration.
Bitumen is an organic substance that gets older as it comes into contact with UV light and oxygen on the other hand it gains resistance to aging through altering by means of Nano clay.

Predicting permanent deformation associated with complex shear modulus and phase angle exhibits enhancement for Cloisite characteristics but with Nano fill very less change occurs.

Mixing the modifiers in specific work rate or rotation for a definite time plays a vital part in quality improvement.

Most researchers have focused on pre- and post-modification, viscosity test, softening point, penetration, and Marshall Stability test that yield a quick result for the properties.

Short-term rutting, long-term fatigue, and water resistance overall times are regions to be explored and therefore innovative research is being conducted in these areas only by few researchers, it needs to be focused even more.

Further, the modification must be examined in the field even subsequently all laboratory tests have been carried out. As live traffic and environmental situations represents the real-time challenges for at least a minimum width and length.

The proportion of the application of modifiers must not be moreover high because it reduces efficiency.

Suitable application of modifiers grounded on neat bitumen with adequate amount transforms the road pavement by means of improved strength, longevity, and budget reduction.

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