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TO STUDY THE EFFECT OF RECYCLED POLYETHYLENE ON PHYSICAL AND RHEOLOGICAL PROPERTIES OF POLYMER MODIFIED BITUMEN

Dr. Lokesh Kumar,
Associate Professor,Department of Chemistry,
J. V. Jain College, Saharanpur -247001

ABSTRACT

The purpose of this research was to investigate the influence that recycled polyethylene has on the rheological and physical characteristics of polymer-modified bitumen. In order to investigate the impact that polyethylene content has on bitumen binder, several different kinds of tests, including rheological, mechanical, thermal, and surface morphological examinations, were carried out. The rheological characteristics of the composite binder were investigated with the use of a dynamic shear rheometer. According to the findings of thermogravimetric analysis, the thermal stability of polyethylene modified binder is significantly enhanced in comparison to that of unmodified binder. The complex shear modulus, storage modulus, and loss modulus of the bitumen binder have all been enhanced as a result of the gradual rise in the polymer contents. The accumulation of polymeric waste as a result of day-to-day applications of polymer materials like building and packaging is one of the main contributors to environmental issues, and recycling polymers is one solution to this problem. Due to the fact that the majority of polymer materials are derived from oil and gas, the recycling of waste polymeric materials contributes to the conservation of natural resources. This article discusses the recent developments in the recycling of polymeric waste from traditional polymers and their systems (blends and composites), including polyethylene (PE), polypropylene (PP), and polystyrene (PS). Additionally, the paper introduces the concepts of mechanical and chemical recycling. In addition, the influence that mechanical recycling has on the attributes of recycled materials, such as their mechanical, thermal, rheological, and processing properties, is examined in this work. These properties are highlighted.

Keywords- *polymer, recycling, polyethylene.*

INTRODUCTION

The amount of discarded plastic has been steadily increasing over the course of the past few decades, and traditional methods of waste disposal, such as burying it in landfills or burning it, have a detrimental impact on the surrounding ecosystem. The utilization of this waste plastic in the production of polymer-modified bitumen (PMB) not only lowers the financial cost of producing PMB, but also the ecological cost of trash disposal. This is accomplished by finding new uses for the excessive amounts of waste plastic. In addition to this, the creep resistance of PmB is often increased, which results in the bituminous felt having a longer lifespan.

Utilizing Differential Sscanning Calorimetry (DSC), dynamic shear rheometry (DSR), and fluorescence microscopy, Fawcett, McNally, and colleagues conducted in-depth research on the performance of virgin polypropylene-based polyolefins and polyethylene. Their findings were published in the journal Polymer. They discovered that PmB had better high-temperature capabilities, as well as swelling in the polymer-rich phase and a restricted miscibility of the polymer-rich phase in the bitumen. In addition, they found that the miscibility of the polymer-rich phase in the bitumen was limited. In addition, differential thermal analysis, differential scanning rheology, and fluorescence microscopy have shown that numerous recycled polymers can be utilised successfully in engineering applications. However, the compatibility of polymers is determined by the composition of the basic bitumen that lies beneath them. This composition shifts based on the source of the crude oil as well as the processing settings that are utilised during the refining process. Additionally, the process of recycling may alter the polymers through breakdown as well as contamination. In the work that is being presented, the applicability of six recycled polyolefinic polymers to modify bitumen is being studied. The bitumen that is being used in the study has a different chemical composition than the bitumen that was used in Fawcett et. al. study's. This is being done to account for the two factors that have been mentioned. In light of the fact that roofing applications typically call for especially high proportions of polymer and that the utilization of recycled polymers would provide the greatest benefit, the purpose of this paper is to evaluate whether or not these recycled polymers are suitable for use in roofing applications.

Polyethylene (PE) is a type of plastomer that has garnered attention for its potential use in the modification of bitumen. The use of PE in asphalt not only makes the material more robust but also makes it more resistant to rutting when the temperature is high. In addition, PE possesses the chemical features of being extremely strong and exhibiting “a longer breakdown period when compared to other commercial plastics”. This is because PE is made from polyethylene. Low-density polyethylene (LDPE), linear low-density polyethylene (LLDPE), and high-density polyethylene are the types of polyethylene that are utilised in PMB production the most commonly (HDPE). Carbon atoms are bonded to long chains of PE to form LDPE, which has a density ranging from 0.91 to 0.94 g/cm3. On the other hand, HDPE has fewer and shorter-chain branching due to its density of 0.94 grammes per cubic centimetre. L-LDPE is a slightly modified form of LDPE. It has the same density as LDPE (0.918 g/cm3), but its molecular chain linearity is more comparable to that of HDPE, which has shorter branching than LDPE. Branching is a significant structural characteristic in polymers; “long-chain branching demonstrates improved melting qualities and processability, whereas short-chain branching improves mechanical and thermal properties”.

“Bitumen modification with recycled linear low-density polyethylene (RLLDPE) is a technique that was created to encourage recycling and reuse of plastic trash. This was done in an effort to lessen the negative effects that road construction has on the environment. The processing conditions have a significant impact on the qualities of recycled polyethylene. For example, a high processing temperature and residence time will diminish the material's mechanical properties, particularly its elongation at break, and will also hasten the material's degradation. It should also be pointed out that recycled polyethylene has surface contaminants and other components scattered throughout the polymeric matrix. Additionally, recycled polyethylene has components that are derived from degradation, such as oxidative degradation, which occurs as a result of their exposure to the elements”.

Ethylene-vinyl acetate copolymer, often known as EVA copolymer, is another type of plastomer that is frequently used in the PMB sector. EVA copolymer is made up "of two different segments: ethylene and vinyl acetate. Both segments are complementary to one another in the sense that the ethylene segment, which is non-polar but crystalline, contrasts with the vinyl-acetate section, which is polar but non-crystalline. The presence of acetate groups in EVA contributes to the material's lack of crystallinity when compared to that of LDPE. Despite this, EVA is a semi-crystalline copolymer, and as such, it improves the physical, chemical, and morphological qualities of PMB. It does this by generating a rigid, robust, and three-dimensional network that is resistant to the stresses that are present in asphalt binder".

"In addition to the groups of plastomer and elastomer, various different types of polymers are now being manufactured from recycled materials". In today's world, there is an ever-increasing focus on the incorporation of recycled materials into asphalt production processes. Crumb rubber (CR) is one of them, and it has a long and illustrious history. CR is manufactured from recycled and processed automobile tyres. At both high and low temperatures, it has been demonstrated that CR can significantly improve the resistance of bitumen to fatigue, rutting, and thermal cracking. During the wet process, CR is mixed into the hot bitumen; following this step, the rubber absorbs aromatic oils from the bitumen, which causes it to swell. The modification of the bitumen causes it to become more rigid as a direct consequence of the loss of aromatic oils in the bitumen. Despite the fact that "CR, is recognized as a low-cost (i.e. in comparison to the costs of other non-recycled polymers) and environmental friendly option for modifying bitumen, CR demonstrates a low resistance to heat. This is a disadvantage because it reduces the pavement service-life period and, potentially, the end-of-life recyclability of the material. Because of this, the combination of CR with a variety of different kinds of polymers has been investigated, and the results have revealed that the properties of the bitumen have significantly been improved".

Plastics find their way into practically every element of contemporary life. The most common kind of plastic is called polyethylene or PE for short. Every year, production facilities all around the world turn out millions of metric tonnes of polyethylene, the majority of which is destined for use in packaging. PE is long-lasting and deteriorates at a far slower rate than other polymers. Plastics don't biodegrade very quickly, therefore they stick around for a very long time in the environment, where they contribute to pollution and pose harm to the local wildlife population. In addition, considerable quantities of plastics are not disposed of in the appropriate manner and are, rather, left in the natural environment. Recycling used materials and promoting eco-friendly industries are becoming increasingly popular around the world. As a result of this, the transformation of PE bags, which are utilised in day-to-day living, from a pollutant into a useful material, such as a bitumen modifier, would be very effective. Melting waste plastics in an attempt to reprocess them into usable materials has been shown to be ineffective due to the fact that the process results in thermo-mechanical decreases and losses in the characteristics of the polymers.

Low density polyethylenes, abbreviated as LDPE, and high density polyethylene, abbreviated as HDPE, are the two primary subtypes of PE. LDPE has a density between 0.91 and 0.94, and is made up of carbon atoms that are bonded to a long chain of polyethylene. In contrast, the HDPE does not contain any branching (density more than 0.94). HDPE is more expensive and more difficult to manufacture than LDPE, despite the fact that it is far more durable than LDPE. On the other hand, high-density polyethylene (HDPE) is produced through polymerization carried out at pressures and temperatures that are significantly lower than those required for the production of low-density polyethylene (LDPE). This study focuses on the usage of thermoplastics, specifically high density polyethylene (HDPE), polypropylene (PP), and linear low density polyethylene (LLDPE), as modifying polymers for base bitumen(penetration 80 dmm). All of the polymer concentrations were maintained at or below 5 percent by weight of the bitumen throughout the process. When bitumen is mixed with polymer, a multiphase system is created. Within this system is a phase that is abundant in asphaltenes that are not absorbed by the polymer. This phase increases the viscosity of the mixture by creating a more complicated internal structure.

The variations in molecular weight and polarity of basic bitumen and polymer have a significant impact on the compatibility of the mixture, regardless of whether the mixing process is carried out mechanically or chemically.

Even though polyolefin does not greatly increase the volume of the mix, it does raise the viscosity. However, even this slight volumetric shift occurred caused of the structural alteration in the PMB blend. Polyethylene, which is a type of plastomer, provides the binder with stiffness and decreases the amount of deformation that occurs under stress. When this was done, the effect was amplified since the concentration of polyethylene in the base bitumen was maintained at less than one percent by weight. The Marshall Stability, resilient modulus, water susceptibility, and fatigue life of the modified binder are all improved when the concentration of LLDPE is increased up to 2.5 percent. When thermoplastic is combined with bitumen, even at room temperature, the result is a rise in viscosity, which leads to an increase in stiffness at the operating temperature. However, thermoplastics do not demonstrate any substantial elastic behaviour.

Under conditions of stress and deformation, the shape of polyethylene is profoundly altered because sliding of chain segments with regard to entanglements takes place at the nodes. The addition of bitumen results in an increase in the blend's viscosity, which can be detected as an increase in the softening point and a decrease in the penetration values. This leads to an improvement in the material's resistance to deformation. PMB blend demonstrates a two-phase morphology as well. Depending on the polymer type, concentration, base bitumen composition, and aromaticity (lower molecular weight polar naphthenic), the system may either have an asphalt-rich phase with a homogeneous dispersion of polymer spheres or a continuous polymer-rich phase with a dispersion of asphaltene globules in it. Both of these morphologies are possible. When a polymer and base bitumen that is compatible with it are combined, the polymer strands get inflated as a result of the polymer strands absorbing a portion of the base bitumen's low molecular weight oil fraction. When the polymer-rich phase transforms into the continuous phase because of the considerably higher fraction of swollen polymer, the swollen strands join to one another at nodes to produce a network that is three-dimensional. This network has a considerable impact on the mechanical characteristics of the binders, which, in turn, has an effect on the bituminous binder blends. Because bitumen and polymer have different densities and viscosities at 25°C and 160°C, the rheology of multiphase systems is very complicated because of the instability of the mix. This ultimately has an effect on morphology, but there is no theory that explains why polymers are effective modifiers.

OBJECTIVE OF STUDY

1. Study on characteristics of the polymer modified bitumen.

2. Study on effect of recycled polyethylene on physical and rheological properties polymer modified bitumen.

RESEARCH METHODOLOGY

When choosing a polymer for a particular application, having a solid understanding of both its physical and rheological qualities is absolutely necessary in order to achieve the highest possible level of performance. It is possible to anticipate the physical and rheological performance of a polymer under specific conditions using a variety of different testing methodologies. In this article, two different experimental testing procedures are anticipated to result in superior combinations of recycled HDPE and LLDPE composite materials.

Melt Index Measurement as per ASTM Standard D-1238

The melt flow rate is a measurement of "the viscosity of the polyethylene resin when it is in its molten state. This measurement is used to estimate how easily plastic products may flow. It is a parameter that is related to the average molecular weight of the resin chains of polymer that are extruded through an aperture of a standard size under defined conditions of pressure and temperature over the course of ten minutes".

$$\text{Flow rate} = (426 \times L \times d)/t$$

(OR)

$$\text{Volume rate} = 426 \times L/t$$

Where,

L = Length of calibrated piston travel in cm,

D = Density of resin at test temperature in g/cm3 , and

T = Time of piston travel for length in sec.

Rheometers are the tools that are necessary to carry out an accurate rheological characterization. The ratio of melt flow rate to viscosity, measured in shear, is inversely proportional. Table 1 displays the results of melt index tests conducted on recycled mixed HDPE and LLDPE polymer composites. The results of these MFI tests have been presented in figure 2.

Density Measurement as per ASTM Standard D-792

The physical tests currently being conducted on the polymer materials include density measurements. The percentage of crystals that are contained within a mass of polyethylene can be determined by looking at its density. Crystals are denser than amorphous regions because "of the layering and close packing of polyethylene molecules" which results in crystals. Amorphous regions are characterised by a more tangled and disorganised arrangement of molecules. The density of a substance is described by the equation and is a measurement of the "compactness" of matter found inside the substance.

Grams and millilitres or centimetres cubed serve as the standard units of measurement for volume and mass, respectively, in the metric system. As a result, the unit for density is either grammes per millilitre (g/ml) or grammes per cubic centimetre (g/cc). At a temperature of 23°C, a density gradient column containing aqueous methanol solutions was used to quantify the densities of the films. The density is determined with the assistance of a Mettler ME-40290 that has been mounted on a Mettler AE 240 scale. Table 2 displays the results of density tests conducted at various temperatures on recycled mixed HDPE and LLDPE polymer composites. These findings from the density test are depicted in figure 3.

Density g/cc, $\rho_1 = (A/B) \times \rho_s$

Where,

ρ1 = Density of the Sample,

ρs = Density of methanol (0.86 g/cm3 at 24 °C),

A= Weight of the sample in air, and

B = weight of the sample in methanol.

Fig. 1 Displacement method test for measuring the Density

RESULT AND DISCUSSIONS

Melt Flow Rate Tests of Thermoplastics by Extrusion Method

When the flow rate of the melt is lower, the length of the molecules and the molecular weight are both increased. Because of the higher molecular weights, it is harder to push the resin through the conventional opening. Therefore, the outcomes of using resins with a higher viscosity, which can be determined by a lower melt flow rate. At a temperature of 190°C, the experiment is carried out with pressure delivered by a standard load caused by a weight of 2.16 kilogrammes. The melt flow rate that results from the experiment is referred to as the melt index (MI), and its values can be found in table 1.

Melt flow index measurement table 1 reveals that it was discovered that the Melt flow index (MFI) values of recycled combined HDPE and LLDPE polymer composites are higher in different percentages (by weight) at different temperatures such as 80, 83, and 86°C when compared to the material (i.e. HDPE and LLDPE at 80 and 20 percent (by weight) at 83°C. This was discovered by observing that the MFI values of recycled combined HDPE and LLDPE.

The Environmental Stress Crack Resistance (ESCR) of resins with this type of distribution is good, and they also have a good impact resistance and good process ability. Molecular weight distribution (MWD) of polymer is contingent on the particular manufacturing technique that was utilised in the production of the polyethylene resin in question. When it comes to polymers that have "the same density and average molecular weight, the melt flow rates of these substances are generally unaffected by MWD. Therefore, resins with the same density and melt index (MI) might have highly varied molecular weight distributions without changing their overall molecular weight".

Table 1 presents the results of the MFI tests conducted on recycled mixed HDPE - LDPE composites

Matl. No	Temp. in °C	Recycled Polymer materials in wt%		MFI in g/10 min
		HDPE	LLDPE	
1	80	75	25	0.50
2		80	20	0.48
3		85	15	0.50
4	83	75	25	0.49
5		80	20	0.30
6		85	15	0.48
7	86	75	25	0.50
8		80	20	0.49
9		85	15	0.48

"As a consequence of this, it is possible for resins to have highly varied molecular weight distributions despite having the same density and melt index (MI)".

It is not possible to determine the range of chain lengths present within the molecules based on the average molecular weight as determined by the MI. If the molecular weight distributions (MWD) of the polyethylene polymers are not the same, then the polymers may have highly different properties despite having the same MI and the same density. A polymer that has a MWD that is narrow will crystallise more quickly and with better uniformity than one that has a wider MWD. This will result in less warpage and greater fidelity to the geometry that was intended. There is a possibility that a polymer with a broad MWD will have improved resistance to stress cracking, impact resistance, and simplicity of processing.

When the extrusion process has been going on for 10 minutes, the materials have achieved the greater viscosity and the lower melt index value. A lower MI (a higher average molecular weight) is predictive of greater tensile strength, toughness, greater stress crack resistance, and the greater energy required at any extrusion temperature to extrude polyethylene resins. This is because a higher average molecular weight corresponds to a lower MI.

It was shown that recycled polymer composites made of HDPE and LLDPE combined had a lower melt index at 83°C with combinations of 80 and 20 percent (by weight) respectively.

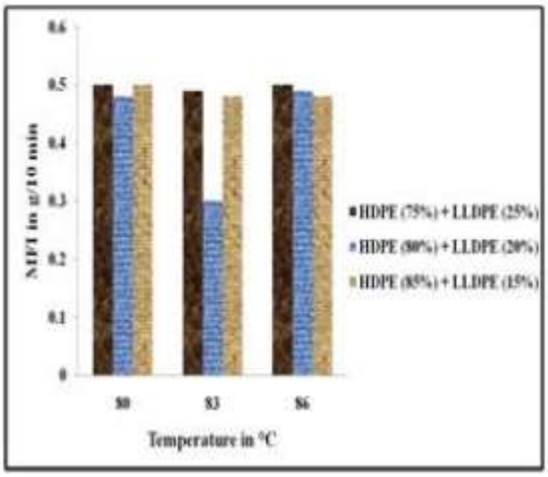

Figure 2 shows the results of the MFI tests conducted on recycled mixed HDPE and LLDPE, expressed as a weight percent.

Density Test of PMC by Displacement Method

Table 2 Density test results for Recycled combined HDPE-LLDPE composites

Matl. No	Temp. in °C	Recycled Polymer materials in wt%		Density at 23 °C in g/cm³
		HDPE		
1	80	75	0.90	0.50
2		80	0.89	0.48
3		85	0.85	0.50
4	83	75	0.90	0.49
5		80	0.95	0.30
6		85	0.92	0.48
7	86	75	0.89	0.50
8		80	0.89	0.49
9		85	0.90	0.48

According to the density measurement table 2, it was discovered that the density of recycled combined HDPE and LLDPE polymer composites changed at various temperatures, such as 80°C, 83°C, and 86°C, depending on the combination that was being used.

It has been found that recycled mixed HDPE and LLDPE polymer composites will have a higher density at 83°C with the combinations of 80 and 20 percent (by weight). This is because of the higher molecular interaction that exists between these two resin components.

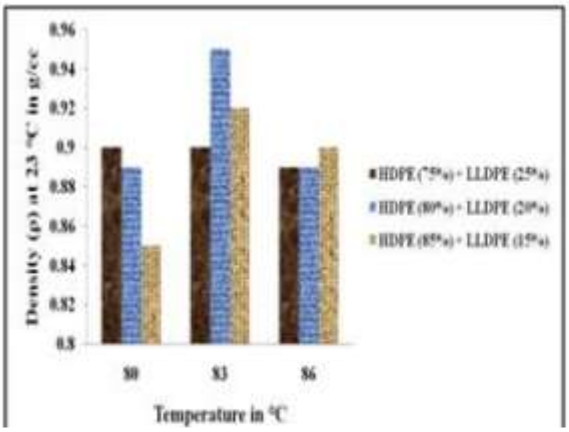

Fig. 3 Density tests result for recycled combined HDPELLDPE by wt%

CONCLUSION

It is possible to draw the conclusion from the prior conversation regarding the recycling process of polymeric waste that mechanical recycling is the recycling method that is most favoured and utilised when compared to the chemical recycling method in which the waste is subjected to complex chemical treatments. This conclusion can be reached as a result of the fact that mechanical recycling was discussed. “In the presence of suitable compatibilizers, the incorporation of minor amounts of virgin polymers with waste from the same or other polymers can result in good properties compared to those of the waste. For instance, the blending of Polyactic acid (PLA) with PET(polyethylene terephthalate) waste resulted in a good thermal stability of the fabricated blend that was almost similar to that of PET waste. These findings are based on the results of mechanical recycling. Additionally, in the case of PC waste, the incorporation ofacrylonitrile-butadiene-styrene (ABS) in conjunction with compatibilizers led to an improvement in the mechanical properties in comparison to those of the trash. In conclusion, the utilisation of the blending approach for the mechanical recycling of waste forms of polymer materials in the presence of suitable compatibilizers should be given more interest in the future. The incorporation of PE into the asphalt binder resulted in a considerable increase in the flexibility of the asphalt binder. This might be proven by the decrease in the phase angle value (), which takes place as follows: The value of the rutting parameter, G* /sin, increased as the percentage of PET in the asphalt binder increased. However, the value of G* /sin for the PE-modified asphalt binder at all of the different percentages of PE complied with the Superpave specifications at high temperatures. As a direct consequence of this, the asphalt binder's ability to withstand rutting saw a large increase.”

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