CHAPTER 4

MATERIAL CHARACTERIZATION

4.1 Preamble

This section concerns regarding the details of collection, preparation, and characterization of various materials (aggregates, fillers, and bitumen) used in the preparation of bituminous mastic and mixes. The physical characteristics of aggregates and bitumen were determined as per the MoRTH (2013) guidelines. Special emphasis was given in determining physical, morphological and chemical characteristics of fillers extracted from waste materials so as to investigate their correlation with the performance of bituminous mastics and mixes. At first, characterization of fillers was done using tests that have been included in the Indian standards on filler for bituminous mixes (MoRTH, 2013). Additional tests that have been identified as reliable performance indicators in various studies (Chandra and Choudhary, 2013; Kandhal et al., 1998) have also been included. Physical characterization parameters were assessed using specific gravity test, plasticity index test, particle size distribution test, porosity tests (Rigden void test and German filler test). Harmful clay contents in all fillers were determined using Methylene blue value test. Morphological and mineralogical analyses were performed using a Scanning Electron Microscope (SEM), X-Ray Diffraction (XRD) and X-Ray Fluorescence (XRF) techniques. In addition to these, the affinity of materials towards bitumen was assessed using pH value and hydrophilic coefficient tests.

4.2 Collection and Preparation of Materials

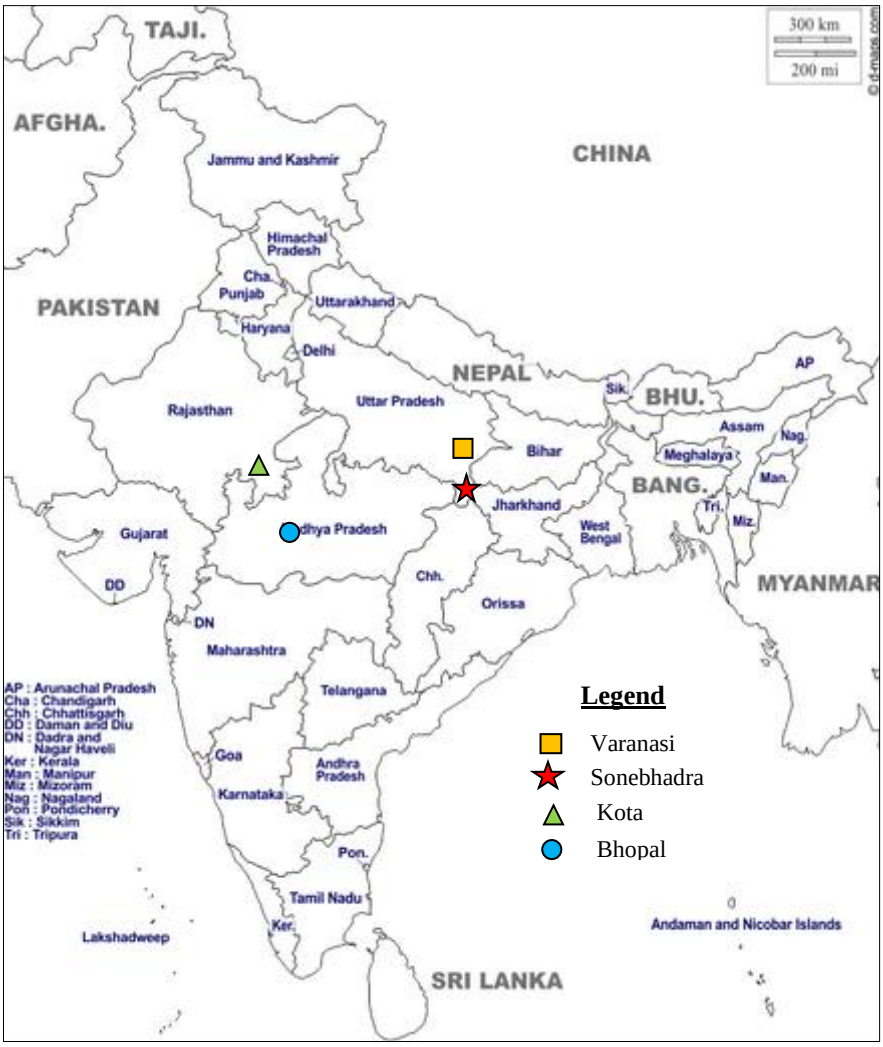


Plate 4.1 Map of India showing various sources of materials used in this study

4.2.1 Aggregates

Aggregates can be considered as the building blocks of bituminous mixes as they are responsible for providing adequate strength and durability to the mixes during its life-time against external factors such as traffic and other agents of weathering. To ensure the optimum performance of bituminous mixes the aggregates should possess the satisfactory engineering properties. This study utilized cleaned and crushed aggregates of dolomite origin for designing of the bituminous mixes. The aggregates were collected from a stone crusher located at Dalla quarry in the Sonebhadra district

(24°41′23″N, 83°3′55″E) of the Indian state Uttar Pradesh (Plate 4.1). The aggregates were washed and sieved through the different sieves and then stored in separate stacks. These aggregates were mixed in a specific proportion to obtain a specific gradation for mix design which is discussed in subsequent chapters.

4.2.2 Bitumen

Bitumen is a petroleum-based hydrocarbon product which is used as the binder to bind aggregate together in the bituminous mix and prevent their displacement/seperation against the external stress caused by the imposed traffic. Bitumen fills the voids between the aggregates and imparts impermeability to the mix. It also acts as a protective layer over the aggregates in the mix and ensures the durability of mixes against moisture and aging. In this study, VG 30 (Viscosity Grade 30) bitumen is used which is grossly equivalent to 50/70 penetration grade bitumen. It was collected locally in Varanasi (25°19'0.1" N, 83°0'37.5" E) from the regional office of Public Works Department (Plate 4.1). The characteristics of the bitumen were determined as per relevant specifications in the subsequent sections.

4.2.3 Fillers

In this study, stone dust of dolomite origin is used as the conventional filler. It was obtained from the stone crusher in the Sonebhadra district along with the aggregates. The Kota stone was collected directly from the dumping ground of a dimensional limestone industry located in the Indraprasth industrial area of Kota city (25°10'57.1" N, 75°50'20.7" E) in the state of Rajasthan (Plate 4.1 and 4.2) in India. The glass powder has been collected from the dumping ground of the Kochar glass factory located in Govindpura industrial area of Bhopal city (23°15'16.9" N, 77°24'10.4" E) in

the state of Madhya Pradesh (Plate 4.1 and 4.3). Apart from these three fillers, composite fillers were used which were prepared by mixing glass powder and hydrated lime in different proportions. Hydrated lime is produced by slaking or hydrating quick lime in water to form a hydroxide (Kakade et al., 2017; Lee 2007). Quick lime was purchased from the local market in Varanasi, which was mixed with the sufficient quantity of water to form hydrated lime (Kakade, 2015). All fillers were sieved through 0.075 mm sieve and stored in a sealed container.

Plate 4.2 Collection of Kota Stone dust in Kota

Plate 4.3 Dumping of Glass Powder near the glass factory in Bhopal

4.3 Characterization of Various Materials

4.3.1 Aggregates

Various tests were performed to determine the engineering properties of the aggregates used in the study. Details of the following tests are stated below and results are collectively stated in Table 4.1

4.3.1.1 Specific Gravity and Water Absorption

Specific gravity is one of the most crucial properties of the aggregates since it plays a critical role in determining the volumetric properties of the mix. It is also considered as an indicator to assess the strength or quality of the aggregates. The aggregates having higher specific gravity are considered stronger than those with lower specific gravity. The aggregates having higher water absorption signifies their higher porosity. The aggregates having high porosity usually considered as unsuitable unless they are found to be acceptable as per strength, hardness, and impact test criterion. These tests were performed as per IS: 2386 (Part III) (1963). The maximum permissible water absorption prescribed by MoRTH (2013) guidelines is 2%.

4.3.1.2 Aggregate Crushing Value Test

Aggregates used in road construction should have satisfactory resistance to crushing under the rollers during their construction and under the application of heavy wheel loads during their service life. This resistance to crushing of aggregates under the influence of gradually applied compressive load was determined using “Aggregate Crushing Value” analysis as per the procedure stated in IS: 2386 (Part IV) (1963) guidelines.

4.3.1.3 Aggregate Impact Value Test

Aggregates used in road construction are subjected to the impact of pounding action during the compaction by heavy rollers and also due to movement of heavy wheel loads of traffic. These impacts might break the aggregates into smaller pieces and hence aggregates should be sufficiently tough to resist fracture under impact load. Aggregate impact value determines the relative measure of the resistance of aggregates to impact and was determined as per the test procedure given in IS: 2386 (Part IV) (1963). The maximum permissible Aggregate Impact Value for the aggregate used in bituminous concrete mixes as prescribed by MoRTH (2013) guidelines is 18%.

4.3.1.4 Los Angeles Abrasion Test

Aggregates used in the surfacing course of the pavements are subjected to the wearing action on their top surfaces due to the movements of the traffic. Hence aggregates should be hard enough to resist the abrasion caused by the moving traffic. Los Angeles abrasion value measures the resistance to abrasion and was determined as per the test procedure given in IS: 2386 (Part IV) (1963). The maximum permissible Los Angeles Value for the aggregate used in bituminous concrete mixes as prescribed by MoRTH (2013) guidelines is 25%.

4.3.1.5 Shape Test

The presence of flaky and elongated aggregates are undesirable for the construction of base and surface courses since they have an inherent weakness with the possibilities of breaking down during compaction and the service life of the pavement. Use of angular aggregates can ensure better interlocking between them. Combined flakiness

and elongation index was used to calculate the amount of flaky and elongated particles in the aggregates as per IS: 2386 (Part I) (1963) specification. The maximum permissible combined flakiness and elongation value for the aggregate used in bituminous concrete mixes as prescribed by MoRTH (2013) guidelines is 30%.

4.3.1.6 Soundness Test

Soundness test is used to determine the resistance of aggregates to the weathering action. This test was performed as per IS: 2386 (part V) (1963) in which aggregates were immersed in the saturated solution of sodium or magnesium sulphate for 16 hours at 27^{o}C to cause accelerated weathering. Subsequently, the aggregates were take out and washed and then dried in the oven. The weight loss of aggregates occurred during this wet and dry cycle was noted and procedure was repeated for 4 more cycles. The maximum permissible weight loss after 5 wet and dry cycles should not exceed 12 and 18% for sodium and magnesium sulphate solution respectively.

4.3.1.7 Stripping Test

Stripping is determined as the detachment of bitumen coating from the aggregates in the presence of water. The displacement of bitumen from the aggregates is due to their tendency to show higher affinity towards water than binders which is influenced by the physico-chemical forces acting on the bitumen aggregate system. To ensure the suitability of the coarse aggregates for bituminous pavement construction, they should show higher affinity towards binder in the presence of water i.e. they should have minimum stripping or maximum retained coating. The testing of the stripping value is done as per the IS 6241(1971) specification. The maximum permissible stripping prescribed by MoRTH (2013) guidelines is 5%.

Table 4.1 Properties of aggregates used in this study

Parameter	Test Specification	Results	Requirements as per the MoRTH (2013) specifications
Bulk specific gravity of coarse aggregates of NMAS 13.2 mm	IS 2386 (Part III) (1963)	2.795	Not mentioned
Apparent specific gravity of coarse aggregates of NMAS 13.2 mm		2.820	Not mentioned
Water Absorption of coarse aggregate (%) of NMAS 13.2 mm		0.374	Maximum 2%
Bulk specific gravity of fine aggregates of NMAS 2.36 mm		2.702	Not mentioned
Apparent specific gravity of fine aggregates of NMAS 2.36 mm		2.747	Not mentioned
Aggregate crushing value (%)	IS 2386 (Part IV) (1963)	15.4	Not mentioned
Aggregate impact value (%)		11.1	Maximum 18%
Los Angeles abrasion value (%)		13.4	Maximum 25%
Combined flakiness and elongation index (%)	IS 2386 (Part I) (1963)	21.3	Maximum 30%
Retained bitumen coating on coarse aggregates (%)	IS 6241 (1971)	99	Minimum 95%
Soundness (Na_2SO_4) (%)	IS 2386 (Part V) (1963)	2	Maximum 12%

4.3.2 Bitumen

Various tests were performed to determine the engineering properties of the bitumen used in the study. Bitumen was tested in un-aged condition as well as after being short-term aged was done according to IS 73 (2013). Details of tests are mentioned in the subsequent sections below and results are shown in Table 4.2.

4.3.2.1 Specific Gravity

Specific gravity is used to conduct weight-volume conversion to determine the volumetric properties of the mix. The specific gravity of the bitumen was determined at 25°C as per IS 1202 (1978) specification using the specific gravity bottle.

Plate 4.4 Penetration testing apparatus

4.3.2.2 Penetration Test

The penetration test is an indirect test to determine the consistency of paving grade bitumen at 25°C. The older grading system of bitumen prescribed in IS 73 (1992) specification was based on penetration values. The penetration value was determined as per IS 1203 (1978) specification and referred as the distance in one-tenth of mm, a standard needle penetrates in a bitumen sample under a load of 100 g for a standard loading time of 5 seconds (Plate 4.4). The harder the bitumen, the lower will be its penetration value and vice versa. The permissible range for the binder used in bituminous concrete mixes as prescribed by IS 73 (2013) guidelines is 50-70 dmm.

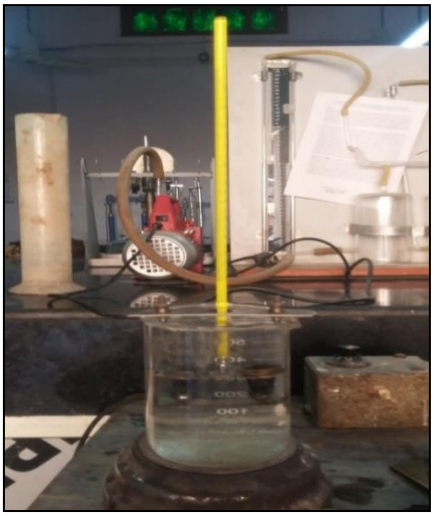


Plate 4.5 Softening point test of bitumen using Ring and Ball apparatus

4.3.2.3 Softening Point Test

Bitumen is a thermoplastic material that changes its state from the solid to liquid with the increase in temperature. Hence bitumen should have sufficient fluidity before it is used in the aggregate mixes. The softening point is the temperature at which bitumen attains a particular degree of softening and is usually determined by ring and ball test as per IS 1205 (1978). In this analysis, a steel ball (3.5 g in weight) is placed on a bitumen sample contained in a brass ring. The sample is placed in a water or glycerine bath. The temperature of the bath was raised at 5°C per minute until the ball travelled a distance of 2.5 cm (Plate 4.5). This temperature is considered as the softening point of the bitumen. Water is used for the bitumen having softening point of 80°C or below. While, glycerine is used for softening points greater than 80°C. The minimum softening point of 47°C is prescribed in IS 73 (2013) specification for VG 30 bitumen.

4.3.2.4 Absolute and Kinematic Viscosities

Viscosity is defined as the ratio between the applied shear stress and shear strain rate at standard temperature. Viscosity test measures the resistance to flow of bitumen. The most commonly used viscosity test on bitumen is the absolute viscosity test which was done as per IS 1206 (Part II) (1978) specification using Cannon Manning viscometer at 60°C (Plate 4.6). It determines the viscosity of bitumen at maximum operating temperature which pavement experiences in most parts of India, and is reported in Poise. The viscosity of the bitumen at a higher temperature (135°C) is termed as kinematic viscosity and was determined using Cannon Fenske viscometer as per IS 1206 (Part III) (1978). It is reported in Stokes (St) or centistokes (cSt). To determine the viscosities in both cases, the time required for the bitumen to cross two pre-marked points on the viscometers at the standard temperatures (60°C and 135°C) was calculated. This time is then multiplied by the calibration factor of the standard

viscometer to obtain the values of the viscosities. The absolute viscosity of VG 30 should be within the prescribed range of 2400-3600, while it should not have kinematic viscosity lower than 350 cST as per IS 73 (2013). The ratio of the viscosity of short-term aged and un-aged bitumen at 60°C should not exceed 4 (IS 73, 2013).

Plate 4.6 Determination of absolute viscosity using Cannon Manning viscometer in a viscometer bath

4.3.2.5 Ductility Test

Bitumen should possess suitable ductility to form a thin film around aggregates to improve physical interlocking between them. The bitumen that doesn't possess suitable ductility forms crack which resulted in the formation of pervious surface. The ductility of bitumen is expressed as the maximum distance up to which a standard briquette of bitumen can be stretched before failure. As per IS 73 (2013), the ductility of bitumen should be determined after subjecting it to short- term ageing in a thin film oven. VG 30 bitumen should have a minimum ductility of 40 cm as per IS 73 (2013).

4.3.2.6 Flash Point Test

Bitumen tends to emit volatile vapours above a certain temperature which can momentarily cause a flash. To avoid any hazardous condition at the field it is necessary to determine this critical temperature at which flash can occur in the bitumen. This temperature is known as the flash point temperature and field engineers

need to restrict the mixing and compaction temperatures well below this temperature to avoid any fire hazard. The flash point of the bitumen is determined using Pensky Martin closed cup apparatus as per IS 1209 (1978) specification. The minimum flash point of 220°C is prescribed in IS 73 (2013) specification for VG 30 bitumen.

4.3.2.7 Solubility Test

The solubility test determines the purity of the bitumen by means of its solubility in carbon disulphide or trichloroethylene. This test was performed as per the IS 1216 (1978) specification according to which 2 g bitumen sample was mixed with 100 ml of trichloroethylene. The solution was then filtered through a filtering mat in a filtering crucible and the materials retained on the filter was dried and weighed to calculate the percentage mass dissolved in the trichloroethylene. Good quality VG 30 bitumen should have minimum solubility of 99% as per IS 73 (2013) specification.

Table 4.2 Properties of bitumen used in this study

Parameter	Test specification	Results	Requirements as per IS 73 (2013)
Tests on neat bitumen			
Specific Gravity	IS 1202 (1978)	0.999	-
Penetration at 25°C, 100g, 5 s (dmm)	IS 1203 (1978)	62	50-70
Softening point (ring and ball test) (°C)	IS 1205 (1978)	51.5	Min 47
Absolute Viscosity at 60°C (poise)	IS 1206 (Part II) (1978)	2972	2400-3600
Kinematic Viscosity at 135°C (cSt)	IS 1206 (Part III) (1978)	380	Min 350
Flash Point (Pensky Martens closed cup) (°C)	IS 1209 (1978)	245	Min 220
Solubility in trichloroethylene (%)	IS 1216 (1978)	99.9	Min 99
Tests on residue after thin film oven test			
Viscosity ratio at 60°C	IS 1206 (Part II) (1978)	2.4	Max 4.0
Ductility at 25°C (cm)	IS 1208 (1978)	65	Min 40

4.3.3 Fillers

Various tests were performed to determine the physical, morphological and chemical properties of the fillers used. The details and relevance of the tests are stated below.

Plate 4.7 Determination of specific gravity in pycnometer

4.3.3.1 Specific Gravity

As mentioned before specific gravity is used in making weight-volume conversions, in calculation of fractional voids of fillers as well as in calculations of particle size distribution in hydrometer analysis. The specific gravity was determined using pycnometer as per ASTM D854-14 (2014) specification (Plate 4.7). Here specific gravity of fillers refers to their apparent specific gravity which only includes the volume of aggregate particles in the calculation without including the volume of any capillary or pore filled with water after soaking for 24 hours. Specific gravities of all fillers are stated in Table 4.3. Stone dust has the highest specific gravity followed by Kota stone dust, glass powder, and hydrated lime. Indian paving guidelines has prescribed the weight batching in the mix production process. Therefore, filler with lower specific gravity occupies higher volume in bituminous mixes for the same weight and vice versa. Some studies have observed influence of specific gravity of filler on the OBC of bituminous mixes (Korayem et al., 2018; West and James, 2006).

Table 4.3 Specific gravities of fillers

Material	Specific gravity
Glass powder	2.369
Kota stone dust	2.650
Hydrated lime	2.363
Stone dust	2.698

4.3.3.2 Plasticity Index

The plasticity index (PI) of the filler is defined as the difference between its liquid and plastic limits. The plastic limit is the minimum water content at which filler changes its state from semi-solid to plastic (Plate 4.8). The liquid limit is defined as the minimum water content at which filler tends to flow under the application of a very small shearing force. PI is an indirect indicator of determining active clay content in the material, which can expand in the presence of water and forms a barrier to adhesion between filler and bitumen, thus weakening the mix. However, some studies reported no correlation between the PI of filler and field performance of bituminous mixes (Kandhal et al., 1998). Thread rolling method (Plate 4.9) for the determination of plastic limit is also subjected to criticism by several experts due to its crudeness (Choudhary, 2008; Sivakumar et al., 2009). However, Indian MoRTH (2013) specification still use plasticity Index criteria for the choice of the appropriate filler and the maximum PI values of the fillers is limited to 4 (except for cement and hydrated lime). Plasticity indices of various materials were determined as per IS 2720 (Part 5) (1985) specification. Glass powder and hydrated lime were found to be non-plastic in nature. Stone dust and Kota stone dust were collected from open areas and have depicted plasticity indices of 2.3 and 3.1 respectively. All fillers satisfied the requirement of Indian specifications.

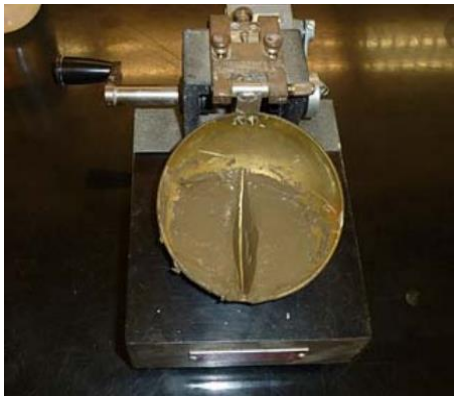


Plate 4.8 Determination of liquid limit

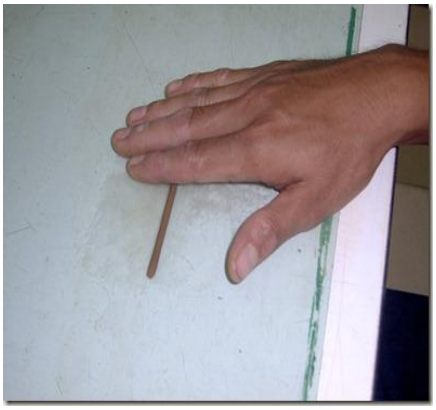


Plate 4.9 Determination of plastic limit

Table 4.4 Plasticity Index and MBV of fillers

Material	Methylene Blue Value (mg/g)	Plasticity Index
Glass powder	1.25	Non-Plastic
Kota stone dust	3.75	3.1
Stone dust	3.25	Non-Plastic
Hydrated lime	0.25	2.3

4.3.3.3 Methylene Blue Value Test

The quantity of harmful fines of smectite (montmorillonite) groups, iron hydroxides and organic matter present in filler samples is determined by calculating the Methylene Blue Value (MBV) of fillers. It was emphasized earlier that the harmful clay in filler can expand in the presence of water and forms a barrier to adhesion between filler and bitumen, thus weakening the mix. Studies have suggested that higher Methylene blue value (MBV) of filler is responsible for inferior performance of bituminous mix against rutting and moisture susceptibility (Antunes et al., 2016; Chandra and Choudhary, 2013; Kandhal et al., 1998; Kuity et al., 2014; Sharma et al., 2010). Clay minerals are crystalline hydrated alumino-silicates which have charged surface and ability to exchange cations. Due to their cation exchange capacity, clay minerals absorb Methylene blue ($C_{16}H_{18}N_3S^+$) solution in an aqueous solution. The quantity of Methylene blue absorbed by the filler is directly proportional to the quantity of harmful fines present in the filler. In comparison to PI analysis, the MBV is a far reliable test and also recommended in European standard EN 933-9 (1999).

Plate 4.10 Performing MBV test in laboratory

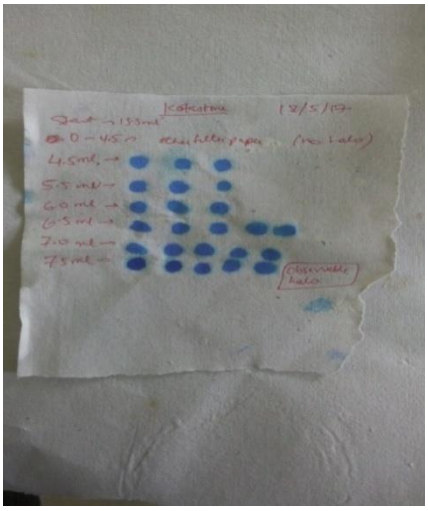


Plate 4.11 MBV analysis of Kota stone dust

In this study, the titration based spot test is prescribed by International Slurry Seal Association, is used to calculate the active fines in the fillers (ISSA 1989, Kandhal et al., 1998). The fundamental principle of this test is to add a different amount of standard aqueous solution of the methylene blue dye to the solution of filler and water until the adsorption of the dye ceases. For this 10 g of filler is dispersed in 30 g of distilled water in a beaker to form filler suspension. The methylene blue dye solution was also prepared by mixing 1 g of methylene blue (MB) with enough distilled water in such a manner that 1 ml of resulting solution contains 5 mg of methylene blue. This MB solution should be filled in the burette and then systematically titrated stepwise in 0.5 ml into the continually stirred filler suspension (Plate 4.10). After each 0.5 ml addition of MB solution to filler suspension, stirring should be done for one minute. After that, a small drop from the suspension was removed with a glass rod and placed on a filter paper. The successive additions of MB solution were done until the endpoint is reached. The endpoint was indicated by the formation of permanent light blue coloration or "halo" around the ring of clear water (Plate 4.11).

There is no specification of MBV for filler in India; however, in Portuguese specification (EP, 2009), maximum permissible limit for MBV is specified as 10g/kg. Kota stone (3.75g/kg) has the highest MBV followed by stone dust (3.25g/kg), glass powder (1.25g/kg), and hydrated lime (0.25g/kg) respectively. All fillers have low MBV which signifies the absence of high active clay minerals in their composition.

4.3.3.4 Void Content

Void contents in fillers (also known as Rigden Voids) compacted to the maximum density is used to analyze the porosity of the fillers. The fillers with higher porosity tend to stiffen the mastic and influence several factors such as OBC, rutting resistance and workability of the bituminous mixes. Hence fillers with excessively high void contents are not preferred in bituminous mixes. The void content in the fillers is usually determined using the Rigden voids apparatus as per the procedure specified in BS EN 1097-4 (2008) specification (Plate 4.12). This test method is based on assumption that the maximum compacted density of fillers can be obtained by compacting the dry fines in a specific manner. To achieve this maximum density 10 g of filler sample is placed into the Rigden voids apparatus and an impact force is given to achieve maximum compaction and form compacted filler bed. After that volume of the compacted filler is calculated to obtain maximum compacted density. The void content can be calculated according to the equation below.

$$Void\ Content\ (\%) = 100 \times \left(1 - \frac{G_{fb}}{G_s}\right) \quad [4.1]$$

$$G_{fb} = \left(\frac{W}{A \times d}\right) \quad [4.2]$$

Where

W = weight of filler taken in analysis

A = cross section area of the Rigden void apparatus (cm^2)

d = depth of the compacted filler bed (cm)

G_{fb}= maximum compacted density of filler bed (cm^2)

G_s= Apparent density of filler (cm^2)

Plate 4.12 Standard Rigden void apparatus

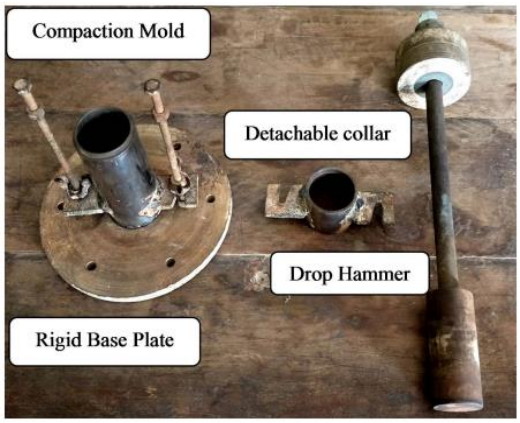


Plate 4.13 Mini compactor apparatus used in the study

In this study, a specially fabricated mini compactor apparatus working on similar principle of the Rigden voids apparatus, was used for determination of the maximum compacted density of fillers (Plate 4.13). Assembly of this equipment was inspired by mini compactor used in geotechnical applications for calculation of unit weight of the soil samples (Gupta and Prasad, 2018; Sridharan and Sivapullaiah, 2005). It consists of a cylindrical mould (diameter 38.1 mm and height 100 mm), detachable collar, a drop hammer (1 kg weight), and a rigid base plate. The dry filler was placed in five layers and each layer is compacted with 25 blows to ensure maximum compaction. After the compaction, the collar was carefully removed and excessive filler was carefully trimmed using a knife. Weight of the sample and equipment (mould and base plate) was then recorded. After deducting the weight of empty equipment, the weight of filler was calculated and divided with the volume of the mould to determine the maximum compacted density (Equation 4.2). The apparent specific density was obtained by multiplying the apparent specific gravity of filler (refer to section 4.3.3.1) with the unit weight of water. Finally, the void content of fillers was calculated using Equation 4.1.

From Table 4.5, it is observed that hydrated lime has the highest void content followed by glass powder. The Kota stone dust displayed the least amount of voids while stone dust has marginally higher void content.

Table 4.5 Void content and German filler values of the fillers

Material	Void content (%)	German filler value (g)
Glass powder	45.73	75
Kota stone dust	32.17	97
Stone dust	36.03	85
Hydrated lime	54.12	35

4.3.3.5 German Filler Test

The German filler test is a simple and inexpensive test which indirectly measures the Rigden voids or porosity of the fillers. The German filler test is a measure of the amount of dried filler required to absorb 15 g of hydraulic oil until it loses its cohesion. In this test, 15 g of hydraulic oil is placed in a small bowl to which 30 g of filler is added to it and mixed. An attempt is made to form a ball from filler mixture. If a ball is formed and holds together, 5 g of filler is added to the mix and process is repeated. The process is repeated until the mixture loses cohesion, which indicated that all the hydraulic oil present in the mix is fixed in the voids of fillers (Plate 4.14). The total amount of filler added up to this point is reported as the German filler value.

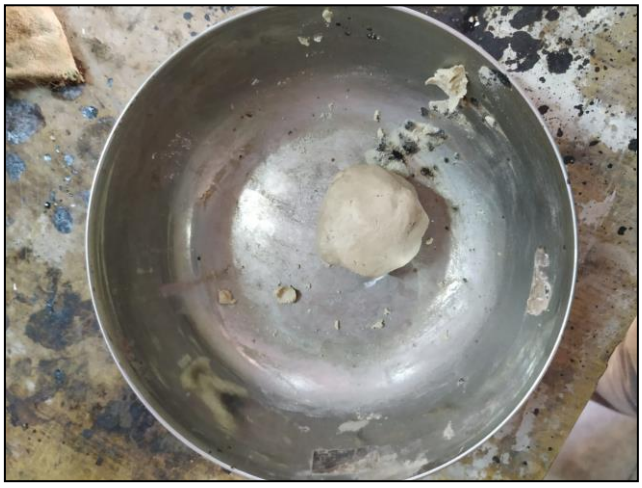


Plate 4.14 German filler test of Kota stone dust

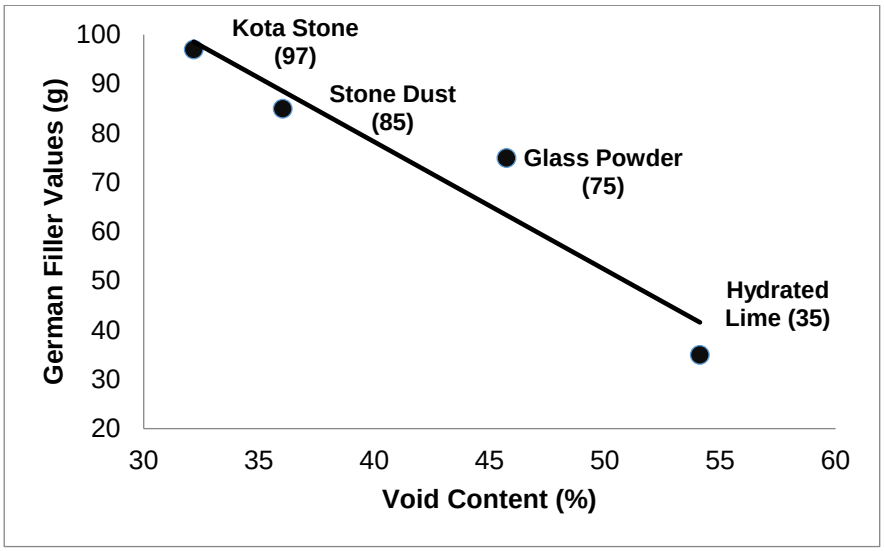


Figure 4.1 Relationship between void content and German filler test values

Some studies have suggested that German filler value test can be used as an alternative test for the relative assessment of filler's porosity (Choudhary, 2008; Kandhal et al., 1998). Lower the German filler test value, higher is the voids in filler and vice versa. Table 4.5 displayed the German filler values of all fillers in the study. Kota stone dust (97g) has the highest German filler value followed by stone dust (85g), glass powder (75g) and hydrated lime (35g). It implies that Kota stone dust and hydrated lime are the least and most porous fillers respectively. Interestingly, a linear equation is found to exist between void content and German filler values as shown in Equation 4.3. It suggested that German filler test can be used alternatively for the relative assessment of porosity of the fillers.

$$German\ filler\ value = -2.595 \times void\ content + 182 \quad (R^2 = 0.910) \qquad [4.3]$$

4.3.3.6 Particle Size Distribution Test

The particle size of fillers has a significant influence over the performance of bituminous mastics and mixes. Various studies has observed the influence of particle size and gradation of fillers on the rutting, fatigue, moisture susceptibility, and optimum binder content of bituminous mixes and mastics (Huang et al., 2007; Kandhal et al., 1998; Modarres and Bengar; 2017; Xu et al., 2019). Particle size distributions of fillers other than hydrated lime were determined using hydrometer analysis as per the ASTM D422-63 (2007) guideline. Particle size analysis of hydrated lime was done using Malvern Mastersizer 3000 apparatus in Advanced Materials and Processes Research Institute (AMPRI) Bhopal (Plate 4.15) owing to its fine size and reactivity with water.

Plate 4.15 Malvern Mastersizer

Figure 4.2 represent the particle size distribution curve of all materials. From these curves, materials differentiating parameters such as fineness modulus (FM), D_{10}, D_{30}, D_{50}, and D_{60} (particle size corresponds to 10, 30, 50, and 60% passing), coefficient of uniformity (C_u), and coefficient of curvature (C_c) were obtained. Fineness modulus was calculated to determine fineness of each material as per Equation 4.4.

$$FM = \frac{P_{75} + P_{50} + P_{30} + P_{20} + P_{10} + P_5 + P_3 + P_1}{100} \quad [4.4]$$

Where FM = Fineness modulus and P_x is the cumulative percentage of filler retained on sieve size "x" µm by mass.

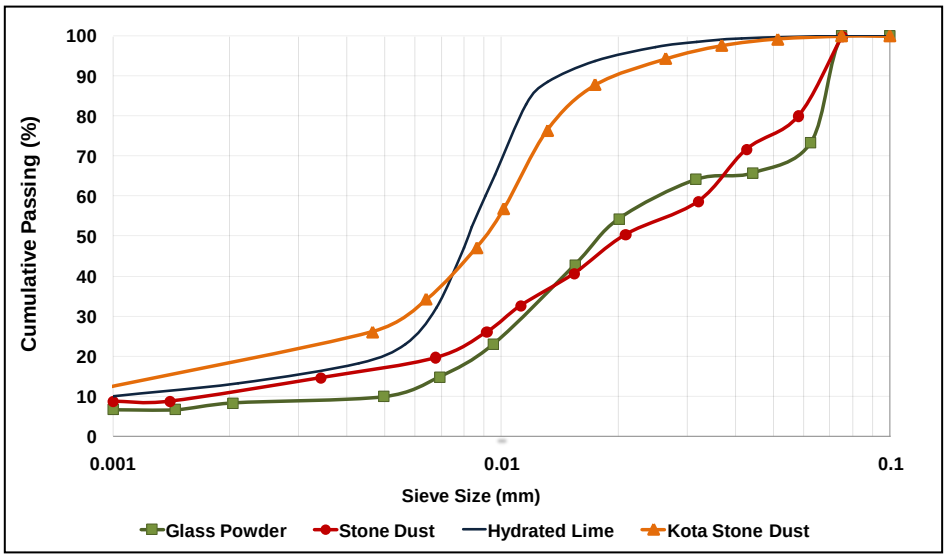


Figure 4.2 Particle size distributions of various materials

Particle size distribution parameters of the fillers are stated in Table 4.6.Stone dust was found to be coarsest material (FM = 5.38), followed by glass powder (4.66), Kota stone dust (3.03), and hydrated lime (2.93). It must be noted that the activity of any filler is associated with its fineness and finer filler typically has higher surface activity. D_{50} values of fillers also followed a similar trend and stone dust (21µm) and hydrated lime (8 µm) has the highest and lowest D_{50} values respectively. Coefficient of uniformity is a measurement of range of particles present in sample and is given as:

$$C_u = \frac{D_{60}}{D_{10}} \quad [4.5]$$

The greater the value of C_u, wider is the range of particle sizes present in the sample. This influences the Rigden void in the filler and affects the cost and performance of mix against various pavement distresses. Stone dust was found to have the highest C_u value (17.89) and glass powder was reported to have the lowest value (4.92). This was

due to the presence of a relatively high percentage of uniformly sized particles in glass powder. Granular material with a higher percentage of uniformly sized particles is difficult to compact properly in field conditions. Coefficient of curvature (C_c) indicates the shape of the gradation curve and is given as:

$$C_c = \frac{(D_{30})^2}{D_{60} \times D_{10}} \quad [4.5]$$

Hydrated lime and glass powder ash have the highest (4.988) and lowest (1.152) coefficient of curvature values respectively.

Table 4.6 Particle size distribution parameters of studied materials

Type of Filler	Fineness Modulus (FM)	D_{50} (µm)	Coefficient of Uniformity (C_u)	Coefficient of Curvature (C_c)
Kota stone	3.03	9	12.71	4.135
Glass powder	4.66	19	4.92	1.152
Stone dust	5.38	21	17.89	1.813
Hydrated lime	2.93	8	9.01	4.988

4.3.3.7 Particle Shape and Surface Texture

Shape and surface texture of filler particles has a significant influence on stiffening of bitumen mastics as well as on OBC, moisture resistance, rutting resistance and compatibility of bituminous mixes (Pasandin et al., 2016; Melotti et al., 2013; Tayebali et al., 1998; Zulkati et al., 2012). A morphological study of various fillers was performed using a scanning electron microscope (SEM) analysis as per ASTM E986-04 (2010) procedure (Plate 4.16). The photomicrographs of fillers were taken at different resolution levels to make a qualitative evaluation of geometric characteristics of various filler particles.

Plate 4.16 Set up for SEM at the central instrument facility, IIT (BHU) Varanasi

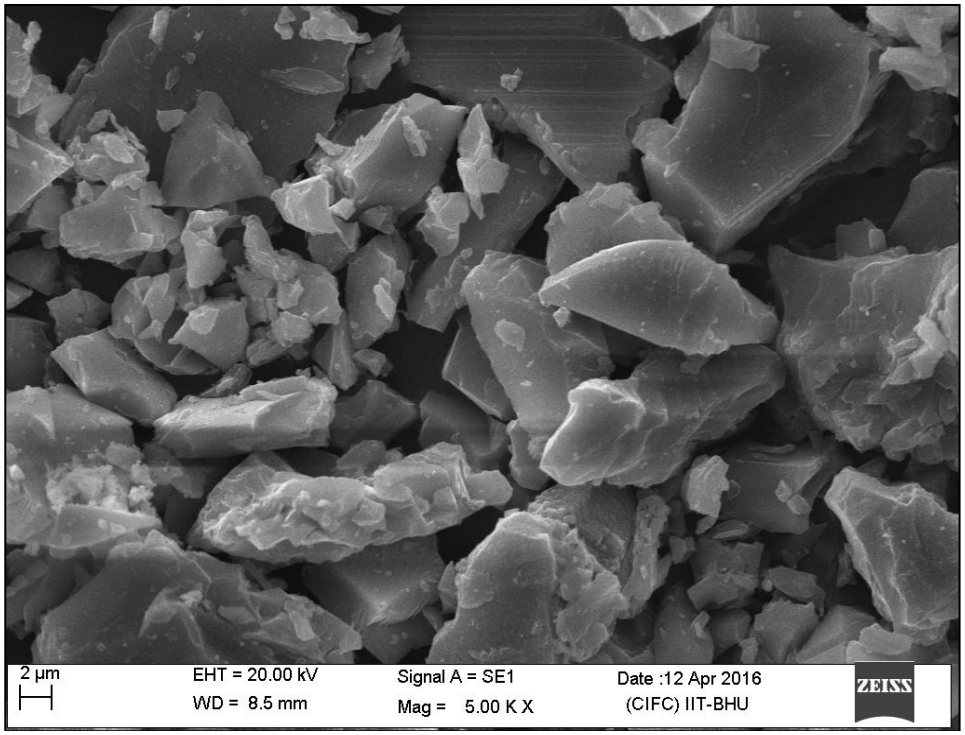


Plate 4.17 Photomicrograph of glass powder at 5000X magnification

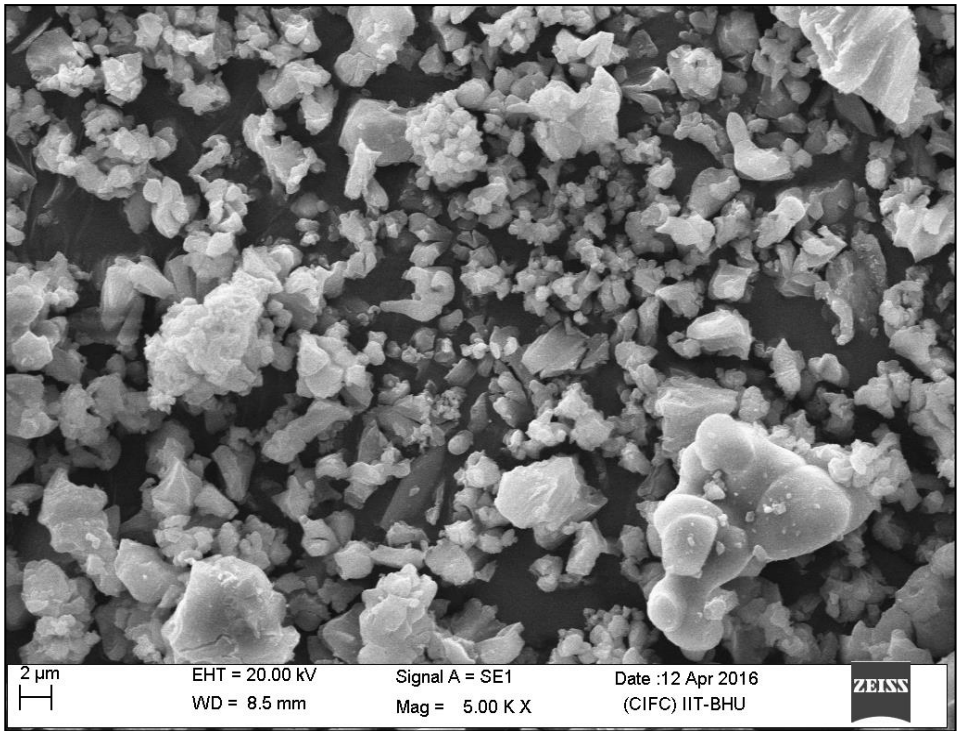


Plate 4.18 Photomicrograph of hydrated lime at 5000X magnification

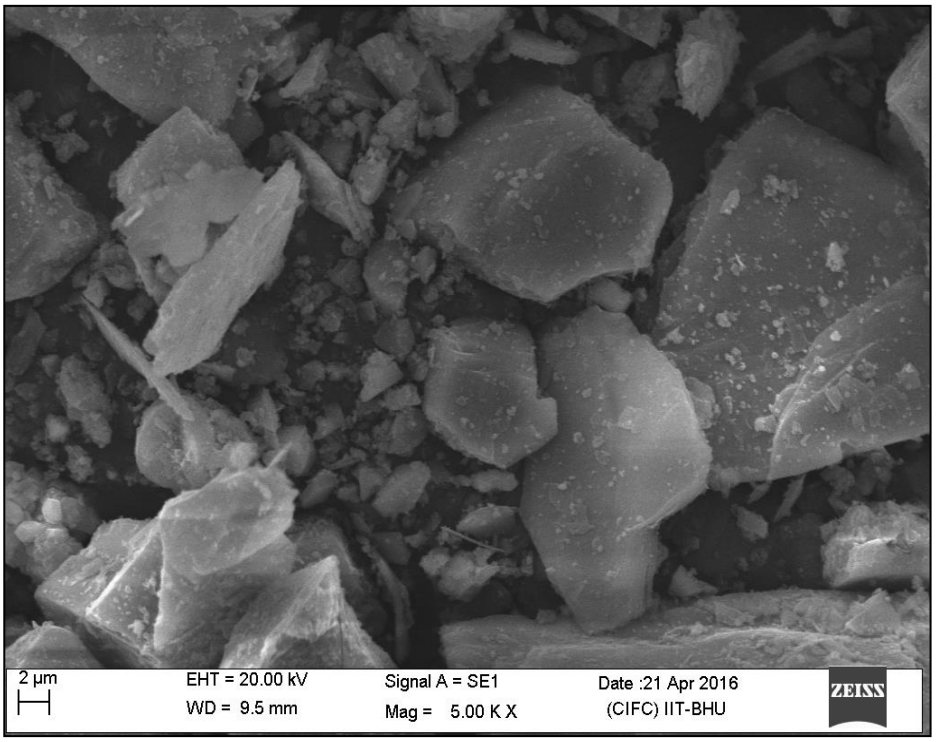


Plate 4.19 Photomicrograph of stone dust at 5000X magnification

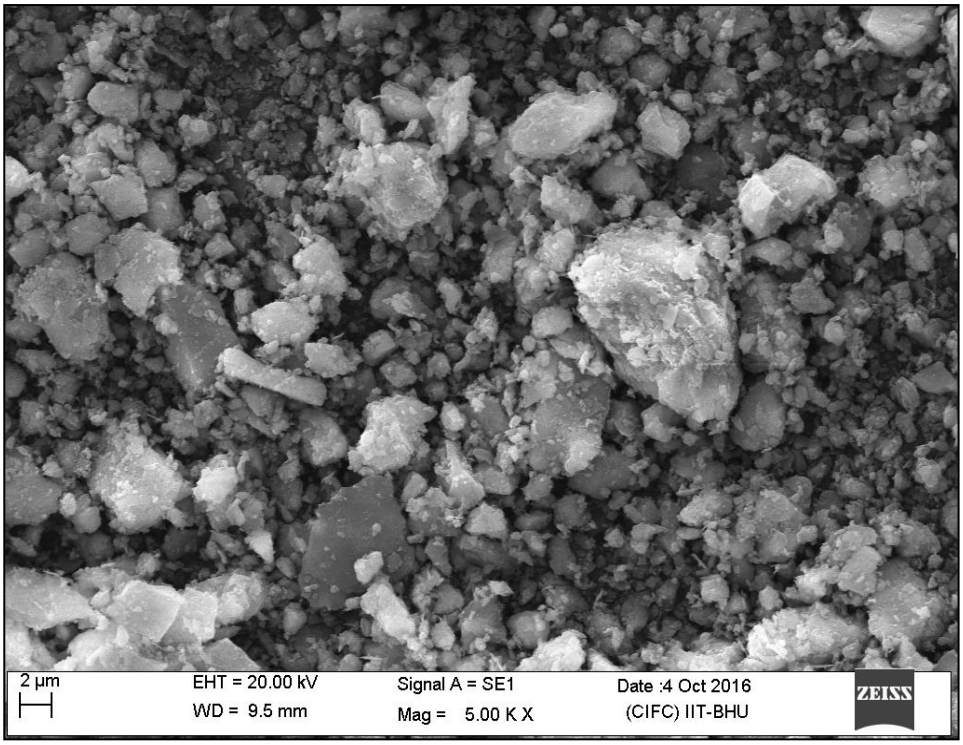


Plate 4.20 Photomicrograph of Kota stone dust at 5000X magnification

Photomicrographs of all fillers at the magnification level of 5000X are displayed in Plates 4.17-4.20. The conventional stone dust looks to have well-graded angular particles with texture lies between smooth to medium rough. The particles of waste glass powder are also angular but seem to be more uniformly graded than that of stone dust. The texture of stone dust looks to be rougher than that of glass powder which may result in higher bitumen absorption on filler surface. Both hydrated lime and Kota stone dust has small particles with varying shape varies in between subrounded to granulous. In the case of hydrated lime, there is an agglomeration of smaller particles, showing a slightly smooth texture. Kota stone dust also has smaller particle but it has the roughest texture amongst all four fillers.

4.3.3.8 Mineralogical Composition

The mineralogy of mineral fillers not only affect their interaction with bitumen but also affect the interaction of bitumen mastic with the aggregates (Bagampadde 2004;

Chandra and Choudhary 2013; Lesueur et al., 2013; Loorents and Said, 2009; Pasandin et al., 2016; Zejiao et al., 2017). This subsequently influences the performance of bituminous mixes, especially against moisture sensitivity. The mineralogical composition of the fillers used in this study was determined using X-ray diffraction (XRD) analysis. XRD analysis is a rapid analytical technique used to determine the primary mineralogical components of various fillers. It is based on the constructive interference of X-rays and the crystalline test specimen. In this analysis, the X-ray beam generated from the cathode ray tube of constant wavelength has been directed on the sample and the diffraction pattern of that beam is recorded. By using Bragg's law, the distances between the planes of the atoms that constitute the specimen can be calculated. The Braggs law is:

$$n\lambda = 2dsin\theta \qquad [4.7]$$

Where, the integer "n" is the order of the diffracted beam, "λ" is the wavelength of the incident beam of X-ray, "d" is the d-spacing (distance between the adjacent plane of atoms), and "θ" is the incident angle of the X-ray beam. The procedure is repeated for different incident angles and a spectrum between the diffraction and angle between the incident and diffraction beam (2θ) is recorded. The particular set of d-spacings captured in an X-ray scan provides a specific fingerprint of the mineral present in the filler (Choudhary, 2008). When this pattern is properly compared with the standard reference pattern, it can help to identify the mineral present in the sample. In this study, the XRD analysis was performed at 1.5406 Å wavelengths with Cu Kα radiation via Rigaku benchtop XRD device (Plate 4.21). The data range for analysis was taken between 10º to 70º with a step size of 0.02. X-ray diffractograms

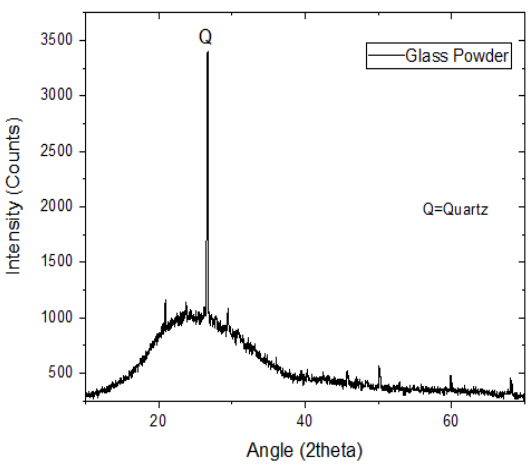
(plots between intensity (counts) and 2θ values) of the fillers were drawn and the prominent lines in the observed pattern were indexed using JCPDS software.

Plate 4.21 XRD facility at central instrument facility, IIT (BHU) Varanasi

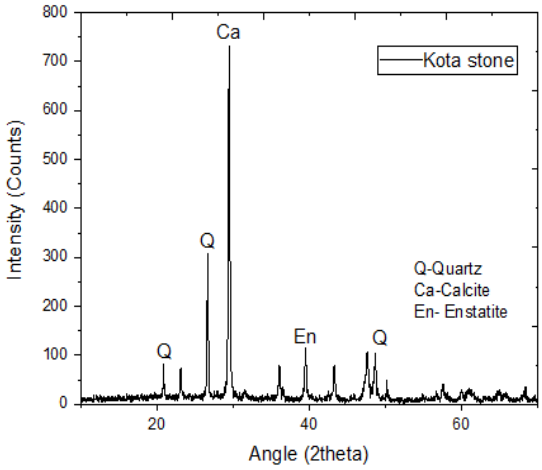
The X-ray diffractograms of all fillers are stated in Figure 4.3, along with identified primary phases. Minerals that are marginally or completely water-soluble are responsible for poor stripping performance of the filler. Conventional stone dust filler primarily constituted dolomite in its mineralogical composition which is a calcium-based water-insoluble mineral that promotes bitumen aggregate adhesion (Bagampadde et al., 2004; Pasandin and Perez, 2015; Pasandin et al., 2016). Similarly, Kota stone also have calcite in its composition, which is another calcium-based water-insoluble mineral that forms strong bonds with bitumen and associated with good adhesion and strong bitumen-aggregate bonds (Bagampadde et al., 2004; Choudhary 2008; Pasandin and Perez, 2015; Pasandin et al., 2016; Xu et al., 2019). Silica in the form of quartz was also found in the composition of all fillers in varying quantities. It was the only mineral found in glass powder and its effect over the stripping potential is highly contradictory. Some researchers believe that it is associated with poor bitumen adhesion (Kuity et al., 2014; Bagampadde, 2004). Other primary minerals along with their empirical formulae are stated in Table 4.7.

Table 4.7 Primary minerals in studied fillers

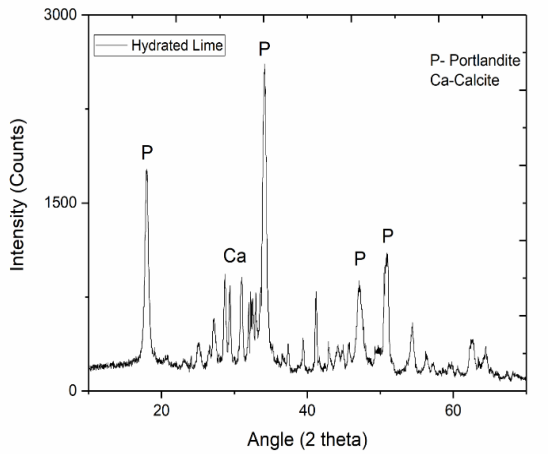
Type of Filler	Mineralogical composition determined from XRD	Empirical formula
Glass powder	Quartz	SiO_2
Hydrated lime	Portlandite	$Ca(OH)_2$
	Calcite	$CaCO_3$
Kota stone	Enstatite	$Mg_2Si_2O_6$
	Quartz	SiO_2
	Calcite	$CaCO_3$
Stone dust	Dolomite	$CaMg(CO_3)_2$
	Quartz	SiO_2
	Ertixite	$Na_2Si_4O_9$



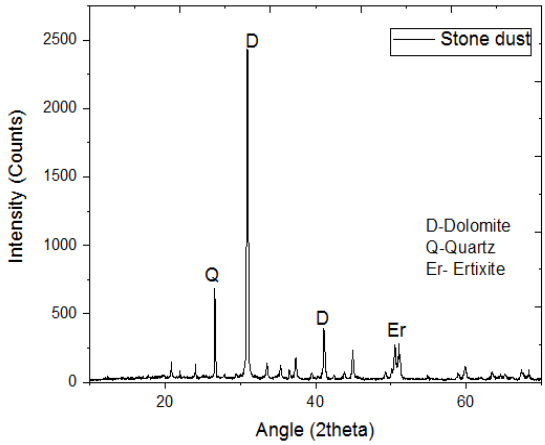
Glass powder



Kota stone



Hydrated lime



Stone dust

Figure 4.3 XRD diffractograms of various materials

4.3.3.9 Chemical Composition

The chemical nature of aggregates and filler influence their interaction with bitumen which subsequently affects the bonding strength and the moisture sensitivity of the mix. The aggregates and fillers can be categorized in acidic and alkaline categories based on their SiO_2 and CaO content. An aggregate can be termed as acidic if its silica (SiO_2) content is higher than 65%, basic when its SiO_2 content is less than 52%, and neutral when its SiO_2 content fall between 52 and 65% (Rice 1958). Antunes et al., (2016) has compared the degree of acidity of various fillers by simply comparing the ratio of SiO_2 and CaO contents in them. The siliceous aggregates have high amount of silica and exhibit weak adhesion with bitumen due to formation of hydrogen bonds between them, while calcareous aggregates have strong electrostatic adhesion with bitumen even in presence of water (Majidadeh, 1968; Stuart, 1990; Bagampadde, 2004).The chemical composition of the fillers was determined using Optim X, X-Ray Fluorescence analyzer with an X-ray tube equipped with Rh anode with a maximum power of 50 kV as the excitation source (Plate 4.22). The chemical composition of various fillers is stated in the Table 4.8.

Plate 4.22 Optim X, X-Ray Fluorescence analyzer

Table 4.8 Comparison of chemical composition of various fillers

Composition	Stone dust	Glass powder	Kota stone	Hydrated lime
SiO_2	14.53	72.88	22.92	1.97
CaO	43.25	9.62	40.32	73.12
Al_2O_3	0.63	1.87	2.53	0.38
Fe_2O_3	0.29	0.40	0.78	0.24
MgO	7.62	1.24	0.69	0.51
Na_2O	0.49	12.31	0.81	0.02
K_2O	0.19	0.43	0.58	0.01
SO_3	0.00	0.03	0.01	0.76
Loss on Ignition	32.72	1.914	31.29	22.93
Ratio of SiO_2/CaO	0.336	7.46	0.568	0.027
Ratio of CaO/SiO_2	2.98	0.134	1.76	37.12

Out of the four fillers, glass powder waste alone comes in the category of acidic filler due to predominance of silica in its composition. Other fillers were identified as alkaline due to their low silica content. Hydrated lime displayed the most alkaline nature due to its highest CaO content (73.12%) followed by stone dust (43.25%) and Kota stone (40.32%). As SiO_2 and CaO contents in filler have detrimental and beneficial effect on moisture resistance of mixes, their ratios (SiO_2/CaO and CaO/SiO_2) could be used to differentiate between the fillers (Majidadeh, 1968; Bagampadde, 2004).

4.3.3.10 Hydrophilic Coefficient and pH Value

The adhesion between the bitumen and fillers depends upon the affinity of filler towards bitumen. Contact of certain materials with water shows higher affinity towards water as compared to bitumen making them hydrophilic. Amount of absorption of bitumen on the surface of hydrophilic materials at their dry state is reported to be much lower than that of hydrophobic materials. Since hydrophilic fillers don't interact well with bitumen, leading to the formation of bituminous mixes

with low strength, low impermeability, and low heat resistance, and hence they are not preferred in the preparation of bituminous mix (Geber and Gomze, 2010). The method to identify hydrophilic fillers is known as hydrophilic coefficient test (Gezencvej, 1985). The hydrophilic coefficient is the ratio of the volumes after the sedimentation of equal volumes of filler in water and kerosene for 72 hours. Set up for hydrophilic coefficient test is shown in Plate 4.23. Hydrophilic fillers have higher volume in water than in kerosene; this led to a higher value of the hydrophilic coefficient. The hydrophobic filler should have a hydrophilic coefficient value lower than 1, which signifies its higher affinity towards bitumen than with water. Any good filler should have a hydrophilic coefficient value lies in the range of 0.7 and 0.85 (Gezencvej, 1985).

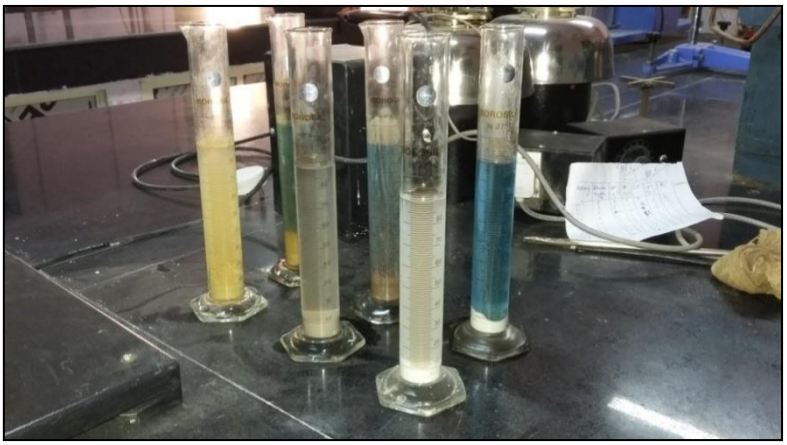


Plate 4.23 Hydrophilic coefficient test of various fillers

Plate 4.24 Set up for pH tests of various fillers

Hydrophilic coefficient values of all materials are shown in Table 5.8. The measured hydrophilic coefficient of all wastes along with stone dust was found to be less than and equal to 1, indicating their better affinity to bitumen than with water. All fillers have hydrophilic coefficient lying in optimum range.

Table 4.9 Hydrophilic coefficients and pH values of studied materials

Filler	Hydrophilic coefficient	pH value
Glass powder	0.81	8.52
Kota stone	0.80	10.22
Stone dust	0.77	12.57
Hydrated lime	0.75	12.74

pH values of all fillers were also tested and shown in Table 4.9. The solution of each filler was prepared by mixing filler and de-ionized water at a ratio of 1:9 by weight and then set for two hours before testing (Plate 4.24). Alkaline materials form a stronger bond with bitumen due to its acidic nature, hence offering superior resistance to stripping. pH values for all materials were found to be greater than 7, which displays their alkaline nature. Hence strong aggregate-bitumen bond was expected in the mixes. Hydrated lime and glass powder had the highest and lowest pH value respectively. This might be due to the presence of a high amount of Portlandite in hydrated lime and predominance of silica in glass powder.

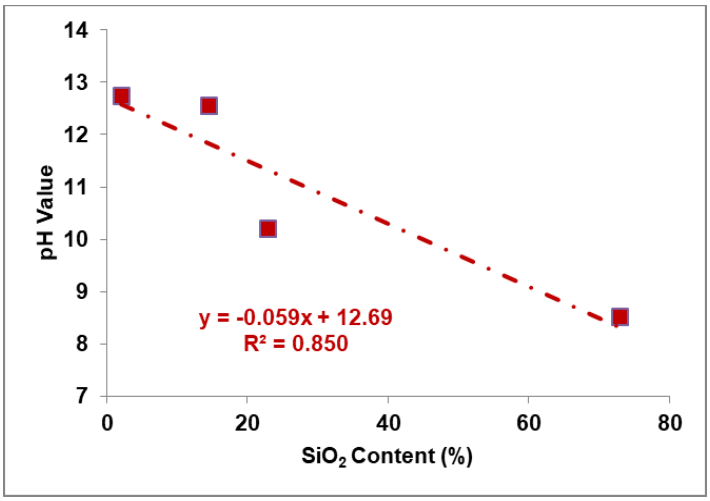


Figure 4.4 (a) Relationship between pH value and SiO_2 content in fillers

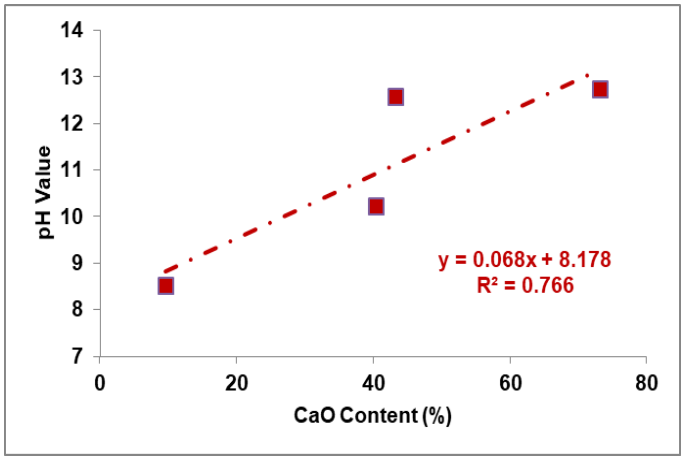


Figure 4.4 (b) Relationship between pH value and CaO content in fillers

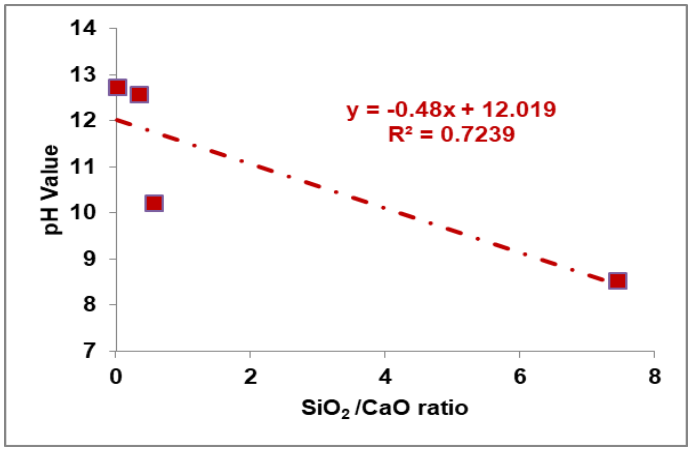


Figure 4.4 (c) Relationship between pH value and SiO_2/CaO ratio

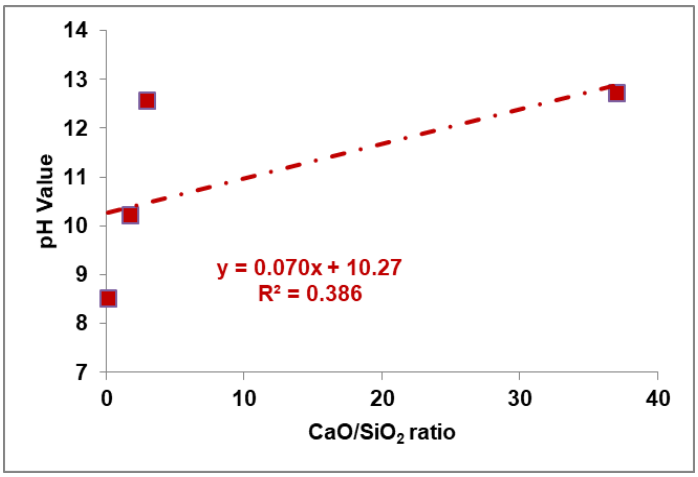


Figure 4.4 (d) Relationship between pH value and CaO/SiO_2 ratio

Figure 4.4 Relationships between pH value and oxide contents

The good relationship between the pH values of the fillers and the parameters determined from XRF tests like SiO_2 content, CaO content, and the ratio of SiO_2 and CaO (SiO_2/CaO ratio) is observed (Figures 4(a), 4(b), and 4(c)). These parameters could be helpful in predicting the interaction between the filler and bitumen. However, interestingly the ratio of (CaO/SiO_2 ratio) doesn't give any good correlation with the pH values of the fillers (Figure 4(d)). The current specification lacks a test method for chemical characterization of the fillers. Hence based on the available data it can be said that pH value test can be adopted as a simple and economical alternative of the much expensive and complicated XRF analysis which could provide details regarding the SiO_2 and CaO contents in the filler. However, a future study should also be conducted by analyzing a larger set of fillers having varying chemical compositions.

4.4 Summary

This section discussed the characterization properties of various ingredients of the bituminous mixes (aggregates, bitumen, and filler). Aggregates and bitumen were found to satisfy the criteria prescribed by the Indian specifications (MoRTH, 2013) and are expected to deliver satisfactory performance as the part of bituminous mastics and mixes. Emphasis was given to determine the physical, morphological, and chemical characteristics of the fillers. All materials fulfilled the criteria of particle size and plasticity index specified in the MoRTH specification. Glass powder and hydrated lime displayed the non-plastic nature, while the plasticity index of stone dust and Kota stone dust are within the prescribed limits. This signifies the presence of a low volume of active clay contents in them. Apart from the plasticity analysis, the methylene blue value test was also conducted which confirmed the presence of a low

amount of active clay content in the fillers. A similar trend was observed between the MBV and plasticity index of fillers. The analysis of void content of fillers was done based on the principle of Rigden void apparatus. New test equipment (mini compactor) was used to determine the porosity of the fillers. It was observed that the hydrated lime has the highest volume of voids followed by glass powder, stone dust, and Kota stone. Hence hydrated lime and glass powder may display the higher tendency to stiffening in mastics and mixes. The porosity of fillers was also determined using German filler test which is a simple and inexpensive test method. A good correlation was found between both methods and hence both tests can be used in a complementary manner. Hydrated lime and Kota stone dust were found to be the finest fillers followed by glass powder and stone dust. Hence they may display a higher tendency to act as bitumen extender, which might result in the formation of mixes with lower OBC. Photomicrographs of various fillers showed that hydrated lime and Kota stone has fine granular particles with rough texture, while glass powder and stone dust consist of angular particles with a relatively smooth texture. Hydrated lime, Kota stone, and stone dust displayed alkaline and hydrophobic nature and have a predominance of calcium-based bitumen adhesion promoter minerals such as portlandite, calcite, and dolomite as determined from pH, hydrophobic coefficient, XRD, and XRF analysis. Hence they are expected to form strong bonding with bitumen even in the presence of water. Although glass powder also displays hydrophobic nature, it has a predominance of silica in its composition which may adversely affect its bonding with bitumen. Good correlation was found between SiO_2 content, CaO content, and SiO_2/CaO ratio determined from XRF and the pH value of the fillers which suggested that both tests can be used in a complementary manner. An attempt will be made to relate aforesaid characteristics of fillers with the performance of bituminous mastics and mixes in the subsequent chapters.