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# Bauxite Residue: A viable filler for asphalt mix

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Research Paper

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## Bauxite Residue: A viable filler for asphalt mix

Bauxite residue or red mud is used as surrogate filler for stone dust in asphalt concrete mixes. Asphalt mixes with both fillers were designed and strength, volumetric properties, and performance against various distresses (rutting, cracking, long-term aging, and moisture susceptibility) were assessed. Red mud mixes displayed higher Marshall stability and resistances against rutting and cracking due to mineralogy and fineness of red mud. Alkaline and hydrophobic nature of red mud produced satisfactory moisture resistant mix. However, red mud mixes had higher optimum binder content and lower long-term aging resistance which was attributed to porous nature of red mud.

### Key words:

waste utilization, filler, red mud, asphalt concrete, sustainability

Prethodno priopćenje

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## Otpadni boksit - održivo punilo za asfaltne mješavine

Otpadni boksit ili crveni mulj koristi se u mješavinama od asfaltbetona kao zamjensko punilo umjesto kamenog brašna. Projektirane su asfaltne mješavine s oba punila i ocijenjena je njihova čvrstoća, volumetrijska svojstva i ponašanje u raznim okolnostima (pojava kolotruga, pojava pukotina, dugotrajno starenje i osjetljivost na vlagu). Mješavine s crvenim muljem odlikuju se višim vrijednostima stabilnosti prema Marshallu i otpornije su na pojavu kolotruga i pojavu pukotina zbog mineralogije i finoće crvenog mulja. Alkalna i hidrofobna svojstva crvenog mulja omogućila su postizanje zadovoljavajuće otpornosti mješavine na vlagu. Međutim, mješavine s crvenim muljem imale su viši optimalni udio veziva i manju otpornost na dugoročno starenje, što se pripisuje poroznosti crvenog mulja.

### Ključne riječi:

korištenje otpada, punilo, crveni mulj, asfaltbeton, održivost

### 1. Introduction

Asphalt concrete mix is one of the most frequently used surface courses in the global flexible pavement network. These mixes primarily consist of aggregates (coarse and fine) along with filler and asphalt binder. Aggregates of various sizes form a skeleton to resist traffic load imposed upon them, and bitumen acts as binder, and leads to adhesion between these aggregates. Filler can be termed as the finest portion of aggregate which passes through a particular sieve (0.075 mm in United States, and India, and 0.063 mm in Europe) [1].

Filler particles play a dual role in asphalt mixes. Coarser filler particles primarily act as the inert material that fills interstices between larger aggregates in mixes and improves density and impermeability of mix. On the other hand, filler particles that are finer than the bitumen film have an active role in the modification of viscosity and consistency of the bitumen-filler mastic [2]. This behaviour influences asphalt mix performance with regard to various pavement distresses such as rutting, fatigue, low-temperature cracking, aging and moisture susceptibility [2]. Poor stiffness within the mastic can also cause drain-down during transportation of mix whereas highly stiff mixes are poorly workable and are difficult to compact [3]. Influence of filler over performance of asphalt mastic and mixes depends upon various physical (particle shape, size, texture, size distribution, porosity, etc.) and chemical (mineralogy, clay content, etc.) properties of fillers, as well as on their physicochemical interaction with the asphalt binder [2-4]. Hence the selection of optimum quality and quantity of filler is vital for the design of good hot mix asphalt. Due to environmental concerns and inflation of the cost of virgin materials, researchers have recently been attempting to utilize various wastes and recyclable materials as fillers in asphalt mixes. Waste materials such as coal fly ash, copper tailings, granite dust, marble dust, glass waste, brick dust, and sewage sludge ash, have been satisfactorily used as filler in various asphalt mixes [5-9].

Bauxite residue also popularly known as “red mud” is a solid by-product generated during production of alumina by caustic leaching of bauxite during the Bayer process. It is a mixture composed of minerals originally present in the parent bauxite and that formed during the Bayer cycle. It is disposed of in the form of the slurry, possessing alkaline nature and high ionic strength. Global production of red mud exceeds 145 million tons annually, out of which four million tons is produced in India alone [10]. Its disposal in ponds and landfills could cause contamination of groundwater due to leaching of heavy metals. Various studies have been conducted over the past decades in order to successfully utilize a significant amount of red mud in relevant applications [10, 11]. However, there is a lack of detailed studies which consider utilizing red mud in asphalt mixes, especially as filler. A recent study compared the rutting resistance of asphalt mixes having red mud and conventional stone dust as fillers [12]. Asphalt mix consisting of red mud displayed superior resistance against permanent deformation, because of relatively smaller specific surface area of red mud, which ultimately reduced the required amount of bitumen in mix and restricted its thermal susceptibility. Other than that, finer grains of red mud (finer than 20 µm) were also found to alter bitumen properties, which also assisted in improving rutting resistance of the mixes [12]. Despite such substantial research, no significant amount of red mud is actually being utilized anywhere in the world. This study investigates the utilization of red mud as mineral filler in asphalt concrete, which is one of the most frequently used material for surface wearing courses in India. This study is an initial effort at characterizing and exploring the suitability of red mud as mineral filler in asphalt mixes employing Marshall mix design. Performance characteristics of mixes containing both fillers are evaluated by examining their fundamental characteristics and by performing various laboratory investigations, involving a minimum of three specimens in each test to meet the reproducibility criteria in standard testing.

**Table 1. Properties of Aggregates and VG-30 Asphalt**

Material	Characteristics	Results	Specification used
Aggregates	Bulk specific gravity of coarse aggregate [g/cm <sup>3</sup> ]	2.736	ASTM C127-15 [13]
	Apparent specific gravity of coarse aggregate [g/cm <sup>3</sup> ]	2.810	ASTM C127-15 [13]
	Bulk specific gravity of fine aggregate [g/cm <sup>3</sup> ]	2.694	ASTM C128-15 [14]
	Apparent specific gravity of fine aggregate [g/cm <sup>3</sup> ]	2.739	ASTM C128-15 [14]
	Aggregate impact value [%]	13.4	IS:2386 (Part IV) [15]
	Los Angeles abrasion value [%]	14.7	IS:2386 (Part IV) [15]
	Combined flakiness and elongation index	23.8	IS: 2386 (Part I) [16]
Asphalt	Absolute viscosity at 60°C, (poise)	2692	IS: 73 [17]
	Penetration at 25 °C, 100 g, 5 s, (0.1 mm)	62	
	Softening point, (Ring & Ball Apparatus) [°C]	51.5	
	Ductility at 27 °C (pull of 5 cm/minute) [cm]	>100	
	Specific gravity	0.999	

Table 2. Adopted gradation of Asphalt Concrete (Grading II) mix [1]

Sieve sizes [mm]	19	13.2	9.5	4.75	2.36	1.18	0.6	0.3	0.15	0.075
Lower-upper limits [%]	100	90-100	70-88	53-71	42-58	34-48	26-38	18-28	12-20	4-10
Adopted gradation [%]	100	95	79	62	51	41	32	23	14	7

## 2. Materials and experimental investigations

### 2.1. Materials

#### 2.1.1. Aggregates

Crushed aggregates of dolomite origin were collected from Dalla Quarry situated in Sonbhadra District ( $24^{\circ}41'23''N$ ,  $83^{\circ}3'55''E$ ), Uttar Pradesh, India and used in this investigation. Aggregates were sieved and washed over their respective sieve sizes to remove fines attached to them. Physical properties of aggregates are given in Table 1. Asphalt concrete (Grading II) mix, which is one of the most popular wearing course, was designed as per Indian specifications [1]. The chosen gradation is specified in Table 2.

#### 2.1.2. Asphalt

The VG 30 (Viscosity Grade 30) asphalt of local origin was used in this study. It is used in lieu of 60/70 penetration grade asphalt and is the most popular asphalt in India. Various properties evaluated as per IS: 73 [6], are given in Table 1.

#### 2.1.3. Filler

Conventional stone dust of dolomite origin, collected from the Dalla Quarry, Sonbhadra District ( $24^{\circ}41'23''N$ ,  $83^{\circ}3'55''E$ ), was utilized as control filler in this study. On the other hand, red mud used in this study was collected from the dumping ground of Hindustan Aluminium Corporation (Hindalco) plant of Renukoot city ( $24.2^{\circ}N$ ,  $83.03^{\circ}E$ ), Uttar Pradesh, India. Oven dried filler with only the fraction that passes through 0.075 mm sieve was used in this analysis.

Physical characterization parameters such as specific gravity and particle size distribution were assessed using the specific gravity test [18] and particle size analysis [19], respectively. German filler test [20] was conducted to determine the intergranular porosity or fractional void content in both fillers. It is a simple test in which filler is added continuously in small dosages to the 15g of hydraulic oil, up to the point the filler oil mix losses its cohesion. The total amount of filler added to oil before the filler oil mix become cohesionless is termed as its German filler value. The porous filler has a lower German filler value and vice

versa. The harmful clay content in both fillers was determined using the methylene blue value test (MBV) [21]. In this analysis, a standard aqueous solution of Methylene Blue (MB) dye, is added in the filler-water suspension incessantly, until the adsorption of the dye ceases. The MBV states the quantity of MB that is needed to cover the total surface area of the clay in the filler. Hence, MBV is proportional to the amount of harmful clay present in filler. Morphological and mineralogical analyses were performed using the scanning electron microscope (SEM) and X-ray diffraction (XRD) techniques. Elemental compositions of materials were determined using the energy dispersive X-ray (EDX) technique. Apart from these, the affinity of materials towards asphalt was assessed using pH value and hydrophilic coefficient [22] tests. pH values of fillers are determined by testing pH of filler-water solution prepared by mixing filler and de-ionized water at 1:9 ratio by weight. The filler having alkaline nature displays greater affinity towards asphalt. The Hydrophilic coefficient is obtained by calculating the ratio of volumes after sedimentation of equal volumes of filler in water and paraffin for 72 hours. The filler having the hydrophilic coefficient of less than 1 has higher affinity towards asphalt than towards water. Various results are stated in Table 3, and in Figures 1-3.

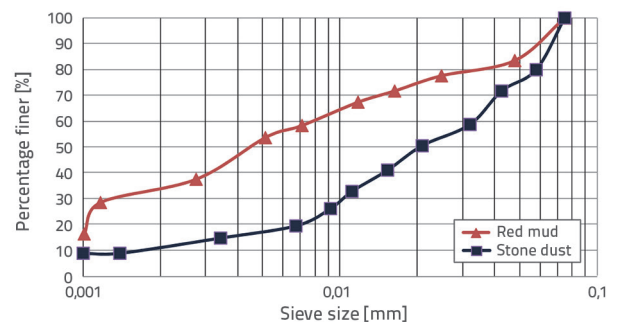


Figure 1. Particle size distribution of both fillers

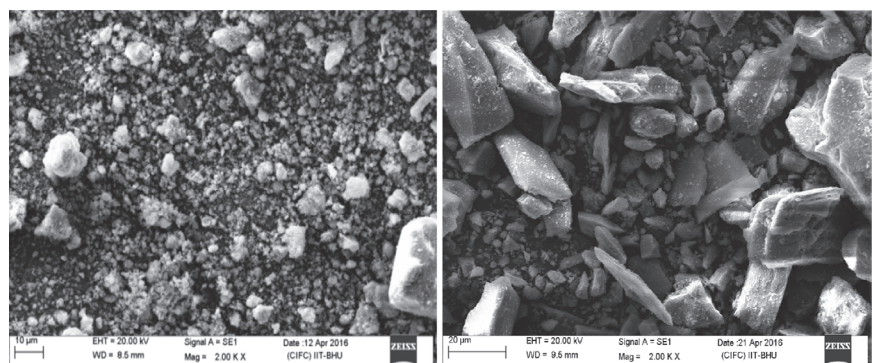


Figure 2. SEM images of Red mud (left) and Stone dust (right)

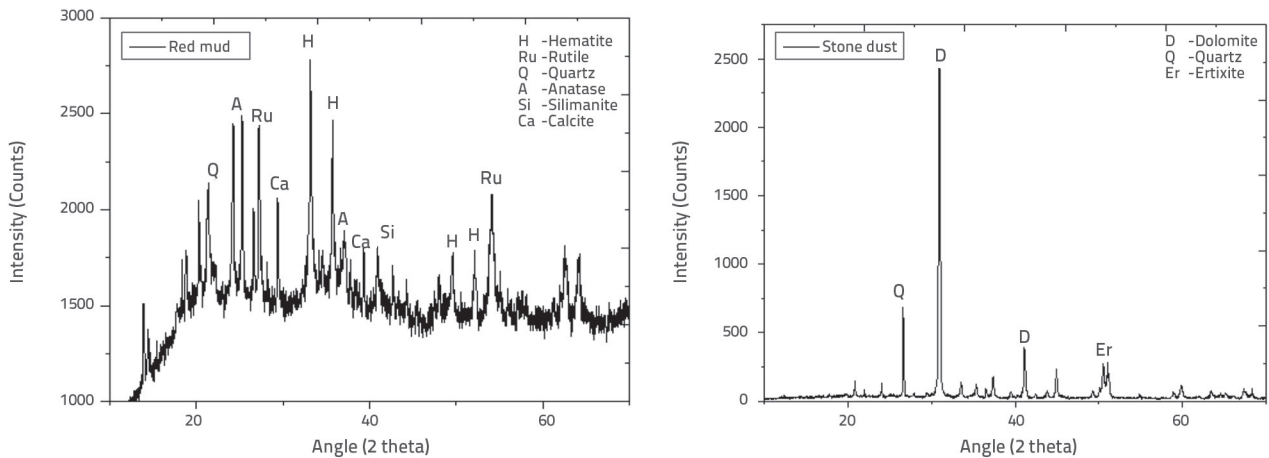


Figure 3. XRD images of Red mud (left) and Stone dust (right)

Table 3. Characterization Properties of Filler Particles

Filler characteristics	Red mud	Stone Dust	Inferences
Specific gravity	3.12	2.70	Red mud has higher specific gravity due to predominance of Aluminium and Iron in its composition.
Methylene blue value [m/g]	2.875	3.250	Red mud has lower harmful clay content per unit weight of the material.
German filler value [g]	50	85	Red mud has higher porosity/fractional voids per unit weight.
Fineness modulus	3.03	5.38	Red mud can be considered as finer filler than stone dust.
Coefficient of Uniformity (Cu)	12.37	17.89	Stone dust has relatively well-graded particles.
Particle shape and texture (SEM)	Small onglomerated particles with a highly rough texture	Angular particles with smooth to rough texture	Small particles and rough texture of red mud may lead to higher bitumen absorption.
Primary elemental composition (EDX)	Iron, Sodium, Carbon, Aluminium, Silicon, Titanium, Oxygen, Calcium	Calcium, Magnesium, Silicon, Oxygen, Carbon, Sodium, Iron, Aluminium	Red mud primarily consists of Iron, Silica, and Aluminium in form of their oxides. It also consists of oxides of Titanium in small amounts and various other heavy metal oxides in trace amounts
Primary mineralogical composition (XRD)	Hematite (Fe <sub>2</sub> O <sub>3</sub> ), Rutile (TiO <sub>2</sub> ), Quartz (SiO <sub>2</sub> ), Anatase (TiO <sub>2</sub> ), Sillimanite (Al <sub>2</sub> SiO <sub>5</sub> ), Calcite (CaCO <sub>3</sub> )	Dolomite (CaMg(-CO <sub>3</sub> ) <sub>2</sub> ), Quartz (SiO <sub>2</sub> ), Ertixite (Na <sub>2</sub> Si <sub>4</sub> O <sub>9</sub> )	Stone dust contains dolomite in its composition, which is calcium based insoluble mineral having good bitumen adhesion. Similarly, red mud also has calcite in its composition, which is responsible for good asphalt/aggregate adhesion. No expansive clay mineral was found in both materials. Thus satisfactory stripping resistance is expected from both.
Hydrophilic coefficient	0.85	0.77	Both materials displayed hydrophobic nature.
pH value	9.98	12.57	Both materials are alkaline in nature and display good affinity towards asphalt.

## 2.2. Design and testing of asphalt concrete mix

### 2.2.1. Design of asphalt concrete mix

The Marshall method of asphalt mix design is recommended in Indian conditions for determining an optimum asphalt content (OAC) of the mix [1]. The mix design procedure as specified in MS-2 [23] was used to determine the OAC of the mix. As per this procedure, samples of asphalt concrete mixes were prepared at specified gradation (Table 2) and at five different asphalt contents (4.5 %, 5 %, 5.5 %, 6.0 %, and 6.5 %). The aggregates were heated to 160°-170°C for 24 hours before mix preparation. Then, asphalt was heated to 135°-140°C before mixing with aggregates. The mixes were compacted using a standard Marshall hammer with 75 blows for each side. Mixing and compaction temperatures of both mixes were determined as per guidelines specified in MS-2 specification [23]. For each filler, a total of 15 samples (3 for each asphalt content) were tested to determine the Marshall stability, flow, percentage of air voids (VA), voids in mineral aggregates (VMA), and voids filled with asphalt (VFA). Asphalt content with 4 % VA is determined, and other aforesaid properties were compared to their respective specification values. At OAC, the apparent film thickness (AFT) was also calculated as per procedure devised in NCHRP Report 567 [24]. AFT can be calculated using Equation (1):

$$AFT = \frac{1000VBE}{S_s P_s G_{mb}} \quad (1)$$

Where AFT is Apparent film thickness (microns); VBE is Effective binder content (% by total mix volume);  $S_s$  is Aggregate specific surface ( $m^2/kg$ );  $P_s$  is Aggregate content, (% by total mix weight);  $G_{mb}$  is bulk specific gravity of mix

### 2.2.2. Marshall Quotient (MQ)

Marshall Quotient (MQ) is the ratio of Marshall stability (kN) to flow (mm) at OAC and can act as an indicator to estimate permanent deformation (rutting) of asphalt mixes. Higher MQ displays a material's higher resistance to permanent deformation and shear stresses, and hence to rutting.

### 2.2.3. Indirect tensile strength (ITS)

The tensile strength of compacted asphalt mixes is associated with pavement's resistance against cracking distresses. It was determined as per ASTM D 6931 [25]. A higher indirect tensile strength (ITS) corresponds to a superior resistance to fatigue and low temperature cracking. For each filler, three Marshall specimens were cast at their respective OAC and

were divided in two groups. Each specimen was loaded along the diametric plane at a constant compressive loading rate (51mm/min) through two opposite loading strips up to failure. The peak load was noted and ITS values were calculated as per equation stated below

$$ITS = \frac{2000P}{\pi DT} \quad (2)$$

where ITS is the indirect tensile strength in kPa; P is the peak load (N); D is the diameter of the sample (mm), and T is the thickness of the sample (mm).

### 2.2.4. Retained Marshall stability (RMS)

The moisture susceptibility of both mixes was determined by means of the retained Marshall stability (RMS). For each filler, six Marshall specimens were cast at their respective OAC and were divided into two groups. The first group of specimens (unconditioned specimens) was conditioned by placing them in water at 60°C for 30 minutes. These specimens were loaded to failure at a constant compression rate of 51 mm/min using curved steel loading plates. The second group of specimens (conditioned specimens) was immersed in a water bath at 60 °C for 24 hours. After that specimens were failed as per the above-specified loading arrangement. The RMS was determined using the mean stability of each group as per equation below

$$RMS = \frac{MS_{cond}}{MS_{uncond}} \cdot 100 \quad (3)$$

where  $MS_{uncond}$  is the average Marshall Stability for unconditioned specimens (kN); and  $MS_{cond}$  is the average Marshall stability for conditioned specimens (kN).

### 2.2.5. Adhesion test

Loss of adhesion between asphalt-aggregate interfaces is one of the primary mechanisms responsible for moisture damage. The adhesion mechanism could be sub-divided into two parts, active adhesion, and passive adhesion.

Active adhesion: It can be termed as the ability of asphalt to completely coat aggregates during the hot mix asphalt mixing operations. Mixing times of both mixes were determined in order to define the influence of filler on active adhesion. The asphalt, aggregates, and fillers were heated separately at 170°C and were then mixed manually. The total time (in seconds) that elapsed between the moment of addition of asphalt and the moment of achieving 100 % coating was measured [26].

Table 4. Average Marshall and volumetric parameters of mixes at OAC

Mix type	OAC [%]	Marshall stability [kN]	Flow [mm]	Bulk specific gravity [g/cm <sup>3</sup> ]	VMA [%]	VFA [%]	AFT [μ]
Stone dust	5.19	14.36	3.2	2.462	14.79	72.95	5.25
Red mud	5.32	17.38	3.5	2.499	14.83	73.02	5.20
Requirement	-	9 (min)	2-4	-	14 (min)	65-75	-

Passive adhesion: It is defined as the ability of asphalt to stick on the aggregate surface under the presence of external factors such as road traffic and moisture [27]. The boiling water test was conducted as per ASTM D 3625 [28] to analyse the effect of filler on passive adhesion. A loose sample weighing approximately 250 g was cooled to between 85°C and the boiling point of water. The sample was then placed into a container with the boiling distilled water for ten minutes. The asphalt that appeared on the water surface was skimmed off, and the sample was cooled to room temperature. The sample was then removed from water and placed on a white paper towel. The degree of asphalt coating was then visually observed by three persons. Inadequate passive adhesion may accelerate various pavement distresses such as cracking, premature rutting, ravelling, formation of potholes, and bleeding.

### 2.2.6. Long-term aging

The long-term aging procedure specified in Strategy Highway Research Program (SHRP) A-003A is adopted to simulate age hardening effects after ten years [29]. For each filler, six Marshall specimens were cast at their respective OAC, and were divided into two groups. Average Marshall stabilities of the samples belonging to the first group (unconditioned samples) were determined as per ASTM D 1559 [30]. All samples in the second group were conditioned in an oven at 85°C for 5 days. After conditioning, samples were tested as per ASTM D1559 [30] to determine Marshall Stability values after aging at 60°C. The Mean Marshall Stability Ratio (MMSR), which is the ratio between average Marshall stability after aging and before aging was calculated to analyse the effect of filler on age hardening.

## 3. Discussion of test results

### 3.1. Marshall and volumetric analysis

The Marshall and volumetric test results determined at OAC are given in Table 4. It can be observed from Figure 4.a that OAC of the red mud mix (5.32 %) is higher than that of the stone dust mix (5.19 %). This may be attributed to higher porosity

of red mud, as evident from its lower German filler value (50 gm) compared to stone dust (85 gm). Because of their high porosity, red mud mixes displayed slightly higher VMA at OAC. This justifies their higher requirements for asphalt to reach 4 % air voids, which led to their higher OAC as related to stone dust mixes.

Red mud mixes have higher bulk specific gravity than stone dust mixes, which may be due to higher specific gravity of red mud. Red mud mixes have 21.03 % higher stability than that of stone dust mixes at their respective OAC's. This higher stability of red mud mixes is due to their chemical composition, as RM mixes primarily consist of iron and aluminium in form of their oxides. Overall, both mixes fulfil the required Marshall and volumetric criteria specified in Indian specifications [1]. The AFT of red mud mix (5.20 μ) was found to be slightly lower than that of the stone dust mix (5.25 μ). This is due to higher asphalt absorption by red mud due to its higher porosity, which reduced the effective binder content of red mud mix as compared to the conventional stone dust mix.

### 3.2. Resistance to permanent deformation

The Marshall Quotient (MQ) value of the red mud mix (4.97 kN/mm) was found to be slightly higher than that of the stone dust mix (4.49 kN/mm) (Figure 4.a). It is evident that lower AFT of a mix may lead to higher rutting resistance [24]. Red mud mixes have a lower apparent film thickness than the stone dust mix, which is why they have a higher MQ value than a conventional mix. Higher stiffness of red mud mixes may also be attributed to the fineness of red mud and its tendency to distribute uniformly in the mix [31].

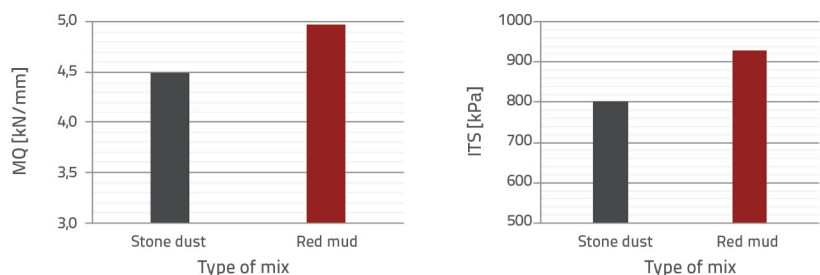


Figure 4. Performance of mixes with regard to: a) rutting resistance (left); b) cracking resistance (right)

Table 5. Average Marshall and volumetric parameters of mixes at OAC

Mix type	Active adhesion (mixing time in seconds)	Passive adhesion (asphalt coverage in %)
Stone Dust	98	90
Red mud	108	85

### 3.3. Resistance to cracking

Red mud mixes have superior ITS (930 kPa) than conventional stone dust mixes (802 kPa). Red mud is relatively finer than stone dust as evident from its lower fineness modulus values. Finer fillers usually form asphalt mixes with enhanced stiffness and ITS, due to their tendency of uniform distribution and formation of integrated structure in the mix [31]. This may be a responsible factor for superior ITS value of red mud mixes (Figure 4.b).

### 3.4. Resistance to moisture permeation

Both mixes satisfied the Indian criteria for minimum 75 % of retained stability [1]. However, stone dust mixes (90.76 %) were found to have slightly higher retained stability compared to red mud mixes (86.48 %) (Figure 5.a). This may be due to higher percentage of dolomite in mineral composition of stone dust and higher AFT of mixes. Mixes prepared with red mud also show good resistance to moisture permeation. This may be attributed to higher content of bitumen encapsulating aggregate particles, which provided good protection against deterioration by moisture. It may also be due to the lower harmful clay content in red mud and/or presence of calcite in mineralogical composition of red mud, which enhances its moisture resistant behaviour.

### 3.5. Adhesion analysis

Similar to the retained Marshall stability test values, stone dust mixes are found to have higher active and passive adhesion values. Stone dust has higher active adhesion since mixes prepared with it take less time to achieve complete coating compared to red mud mixes (Table 5). In the case of the boiling water test, retained asphalt coverage is similar in

both cases (Table 5). The superior active adhesion of stone dust mix is evident since stone dust has a higher affinity towards asphalt than red mud, as observed from its lower hydrophilic coefficient and higher pH value.

### 3.6. Resistance against long term aging

The effect of long-term aging on the performance of stone dust and red mud mixes is expressed in terms of MMSR, as shown in Figure 5.b. Red mud mixes (89.28 %) have lower MMSR than stone dust mixes (97.65 %). It was previously observed that AFT of red mud mixes was lower than that of conventional mixes. Lower AFT of mixes causes accelerated hardening of constituent asphalt binder [32]. Hence lower AFT of red mud mixes may lead to the lowering of asphalt-aggregate adhesion, which caused deterioration in Marshall stability of the conditioned sample.

## 4. Conclusion

The suitability of red mud as mineral filler (in lieu of stone dust) in asphalt concrete mixes was analysed in this study. Red mud displayed all positive filler traits. Marshall and volumetric properties of asphalt concrete mixes prepared with red mud satisfied all criteria specified in Indian specifications. In fact, red mud mixes displayed superior Marshall stability than conventional mixes, which may be attributed to the presence of oxides of aluminium and iron in its composition. However, red mud is relatively porous in nature and so it absorbed a higher amount of bitumen, which subsequently increased OAC of its mix. Mixes containing red mud also displayed superior Marshall quotient as well as ITS values due to greater fineness of red mud. Red mud mixes had marginally inferior moisture resistance compared to stone dust mixes. However, red mud mixes fulfilled the minimum retained stability requirements, but also displayed reasonably good active and passive adhesion values. This may be attributed to highly alkaline and hydrophobic nature of red mud, which ensured good asphalt cement-filler adhesion.

Apart from this, the presence of adhesion promoters like calcite, and absence of active clay minerals, also ensured its satisfactory performance

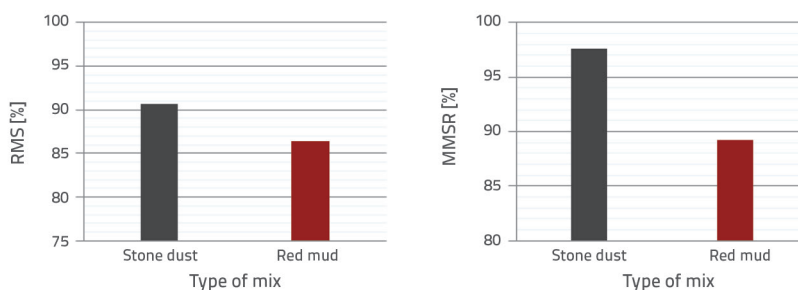


Figure 5. Performance of mixes with regard to: a) moisture susceptibility (left); b) long-term aging (right)

with regard to moisture. Red mud mixes displayed relatively lower resistance to long-term aging, which was due to greater oxidation of asphalt cement due to lower AFT of the mix.

Despite its shortcomings, red mud can effectively be used as filler in asphalt concrete mixes and its usage could be considered economical in places where it can be found in considerable quantities.

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