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Influence of traffic loading and weathering effects on chemical and physical properties of asphalt and asphalt mixtures

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Abstract —*As the development of paved road is going on, natural resources are depleting and using reclaimed asphalt pavements is not only economical but also it is also corroborating to be very beneficial in preserving natural resources for future generations. It is a common practice to add a small amount of reclaimed asphalt in construction of flexible pavements without drastically altering properties such as stiffness and low temperature and thermal cracking of the mix. The use of RAP improve resistance against rutting and fatigue cracking. The main objective of this study is to assess and analyze the chemical and physical characterization of aged asphalt mix properties. Initially, asphalt was laid according to the prepared job mix formula. The RAP and asphalt core samples of both wearing and base surfaces were collected after three years from the same site. The feasibility of RAP for the use in flexible pavement was tested by physical mix tests and FTIR spectrometer. The results indicated a slight decrease in strength, durability and overall bitumen content with increase in sulfoxides groups' concentration.*

Keywords **–** *Fourier-Transform Infrared Spectrometry, Aging, Reclaimed Asphalt Pavements, Asphalt Mix properties, Bitumen, Functional groups, Federal Highway Administration*

I. INTRODUCTION

It has been evaluated through long-term pavement performances that performance of pavements which contain up to 30 percent reclaimed asphalt pavement (RAP) is comparable to that pavements constructed from virgin materials with no RAP (Reclaimed Asphalt Pavement in asphalt mixtures: State of the Practice; Publication No. FHWA-HRT-11-021, 2011).But, there is recurrent need to analyze the effects of RAP and the binder extracted from the RAP to be used in fresh HMA mixes and how does these aged materials affect the chemical and physical properties of such mixtures.

Volatilization and oxidation were two major factors that imparts changes in rheological properties of the binder that leads to long term aging of asphalt binders (Asphalt Institute, 1996; Peterson 2009; Traxler 1963). In 2000, Lu and Isacsson stated in their publication that these two mechanisms are irreversible chemical reactions due to binder chemical compositions change. Over the long durations at elevated temperature, traffic loads and continuous air flow over the road surface, the binder oxidizes and gradually Aging becomes more severe by this air flow and temperature variance (Chen and Huang, 2000; McGennis et al., 1994).

According to Petersen (1984), the major changes that occur after aging in the asphalt binders are following:

- Binder Composition Change through oxidation
- Maltenes content reduction due to volatilization
- Adsorption and slow crystallization of waxes
- Rearrangement of resins and asphaltenes

In literature, there is lack of research in aging impacts on physical and chemical behavior of reclaimed asphalt pavements. Different studies are presented in this research such as Fourier transform infrared spectrometer (FTIR),Scanning electron microscope (SEM) and asphalt mix properties tests. Among these tests, FTIR which is used to obtain the infrared spectrum of absorption or emission of any sample material i.e. solid, liquid or gas. Basically, it gives the analysis of infrared radiation interacting with the molecules of the sample.An FT-IR spectrometer simultaneously collects high-spectral-resolution typically of 4 cm^{-1} with frequency of 32 hertz. The data is collected over a wide spectral range of 350 cm^{-1} to 7800 cm^{-1}.

The Fourier transform converts raw data into presentable spectrographs i.e. wavenumbers in units of cm^{-1}. The techniques used in FTIR spectrometer are types of absorption spectroscopy which gives us information about the chemical properties of certain material by analyzing how much of light is absorbed at each wavelength. FTIR spectrometer is used to characterize functional groups of asphalt binder. These functional groups are important in imparting certain chemical behavior to bituminous material. Changes in concentration of these functional groups is the main marker of aging phenomenon.

FTIR gives useful information about the absorption of infrared radiations, which typically ranges from 400 to 4000 cm^{-1}. It states that bonds in molecules attract IR radiations at resonant rates that are evocative for their vibrations. The Figure shown below represents a typical spectrograph. On Y-axis, Transmittance % is plotted. Transmittance is the amount of light (in percentage) that is transmitted or passed through the sample material whereas on X-axis, frequency of the IR radiation or wavenumbers are plotted. Frequency and wavenumbers are proportional to each other so they can be used interchangeably. Higher the wavenumber of a functional group present in the compound, stronger will be the bond and vice versa. Similarly, if a lighter atom is bonded to particular atom, the wavenumber will be higher in this case relatively to a heavier atom attached to that particular atom.

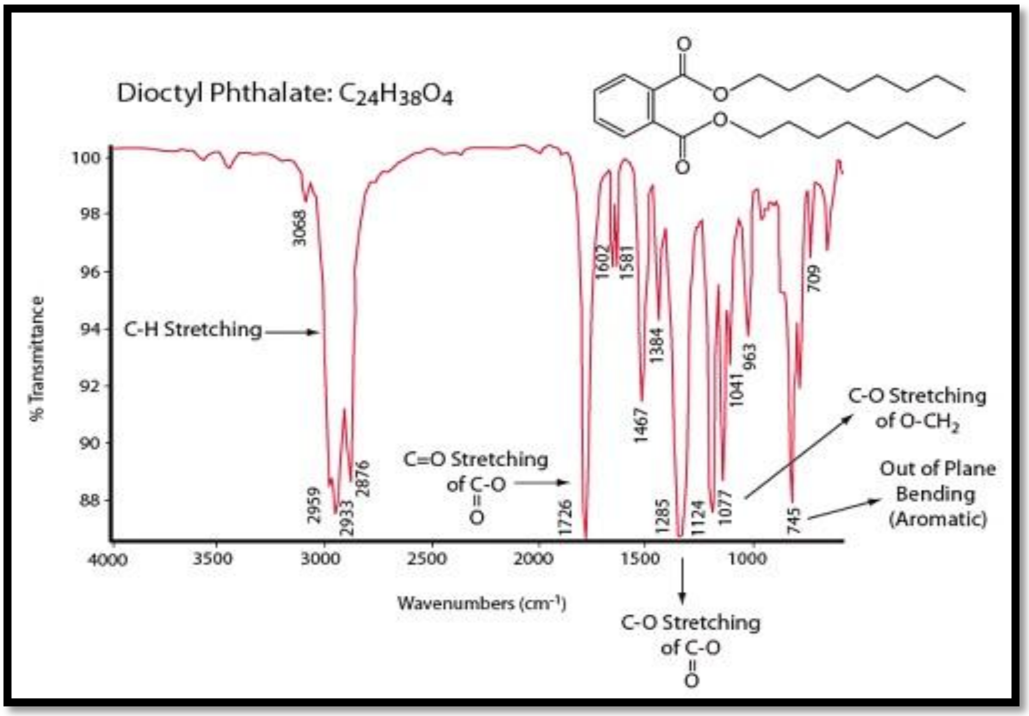

Figure 1. Typical image of spectrograph made by an FTIR Spectrometer

Saliani et al. (2019) evaluated chemical properties of virgin and RAP extracted binders by using FT-IR spectroscopy. It was concluded that bitumen absorption was impacted by virgin aggregates, binder and RAP. Coarse and fine RAP particles were separated and added into different hot mixed asphalt (HMA) mixtures for the testing purposes. The observed results showed that the fine and coarse recovered bitumen did not share same chemical characteristics, and the interaction of RAP bitumen with virgin bitumen significantly depended on RAP particle size. The amount of active RAP bitumen in coarse RAP particles was higher than in fine RAP particles.

II. METHDOLOGY

The asphalt mix was prepared in the laboratory and then laid at Grand trunk road near wazirabad bypass. Cores were extracted from the site and various tests were performed as shown in the figure 2 below.

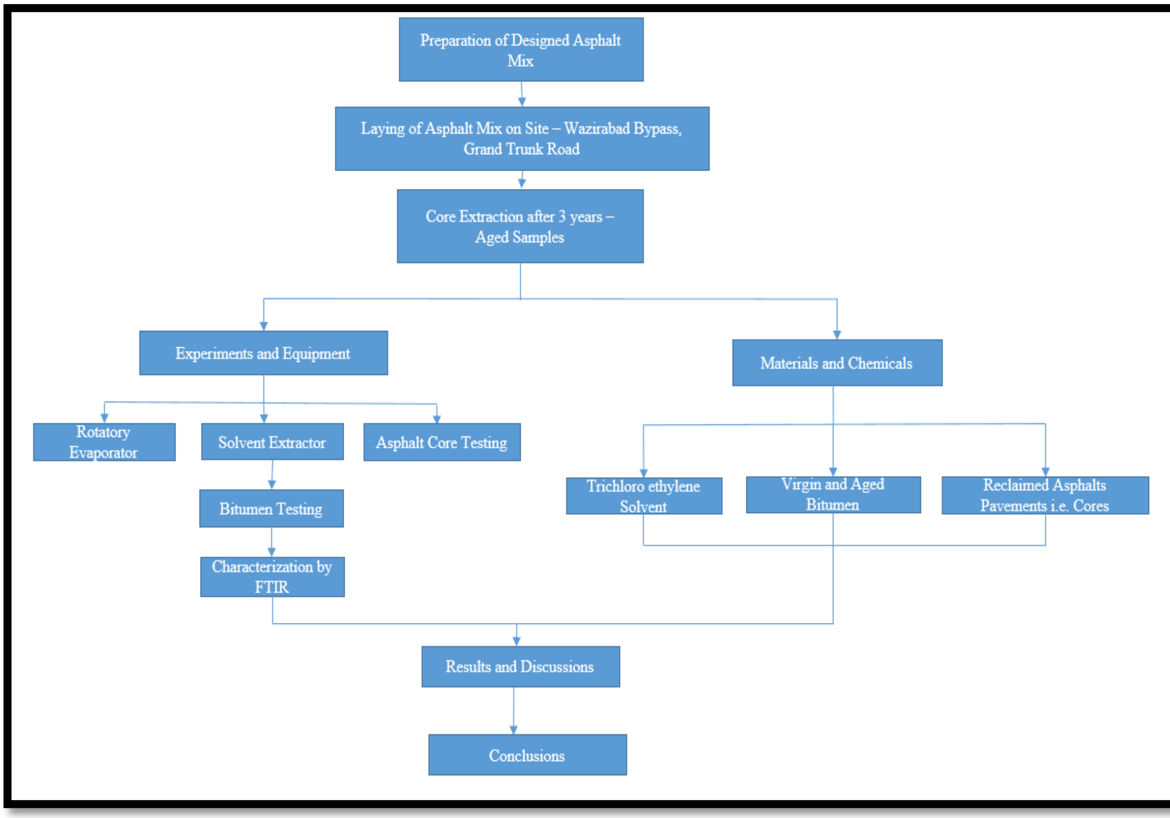

Figure 2. Research methodology flowchart

III. MATERIALS AND METHODS

Following methods and material were adopted in an orderly sequence for this research project. They all are listed down as under:

1. Premix asphalt samples of wearing and base course were prepared in the laboratory according to job mix formula. The standard acceptable limits for properties of bitumen, aggregates and mix were also determined for application as wearing and base course in accordance with ASTM D-8159, D-6927, D5404, D-2726 and D-2041.

2. A trial section of 30m length on north bound of G.T. road (two lane divided highway with TST on edges both sides) was selected for overlaying and necessary scarification as shown in figure 3. The width of each lane is 3.5 meters. The road was scarified in year 2016 up to layer of asphaltic base course.

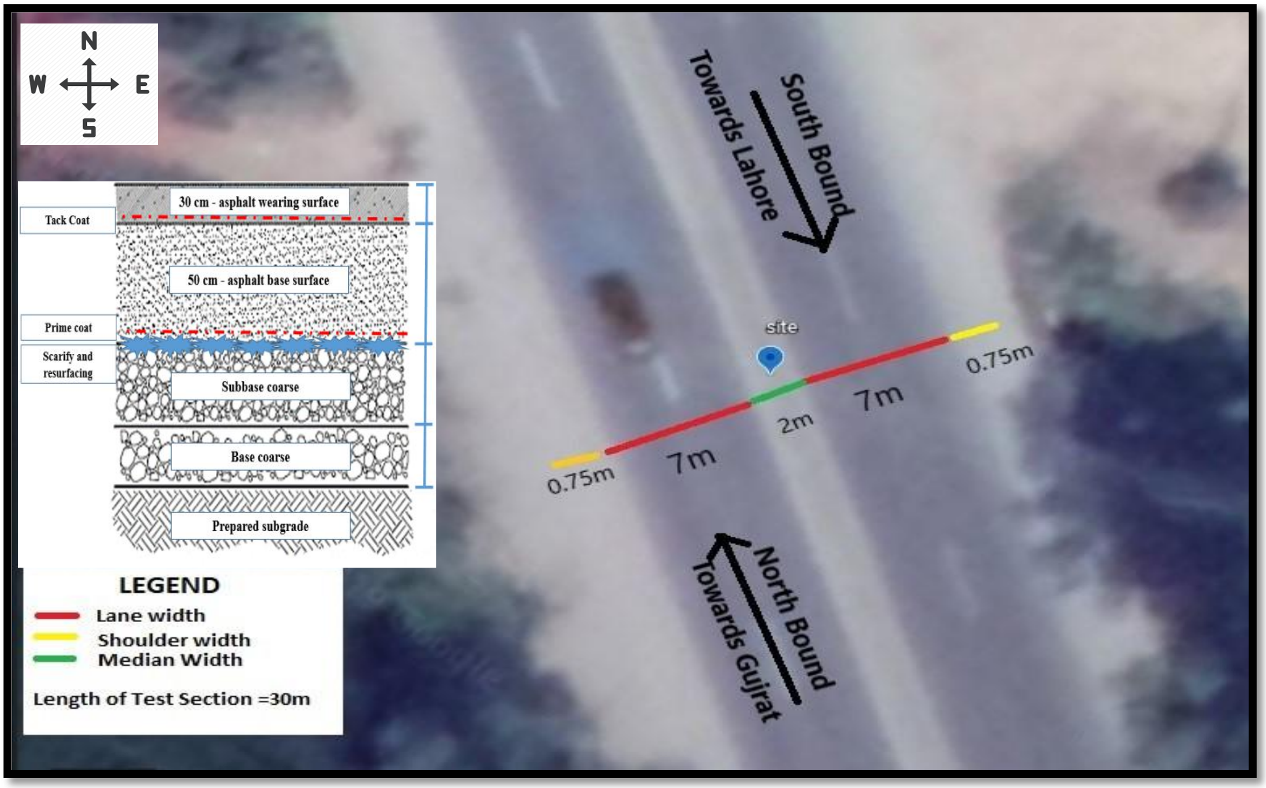

Figure 3. Site Location Map

3. An overlay of pre-mix asphalt plant manufacture asphaltic base course and asphaltic wearing course was laid in sectional dimensions as shown in sectional layout in figure 3 in accordance with the specification of AASHTO MS-2. After laying of asphalt mix, the images captured from the site are presented in figure 4.

4. Compaction was done by pneumatic tyre roller (PTR) and tandem roller respectively for asphaltic base course at the trial section. For asphaltic base course, the weight of PTR used was 24 tonnes and 7 to 8 passes were completed for required compaction along with the 1 to 2 passes of tandem roller (10 tonnes) over it. Further to remove any additional surface marks, one pass of static roller is applied.

5. Similarly, for asphaltic wearing surface, 7 to 8 passes of 24 tonnes PTR were applied along with 1 to 2 passes of 10 tonnes tandem roller over it. One pass of static roller was also applied for smoothening of the road surface.

6. Asphaltic wearing and base materials were subjected to traffic loading, environmental changes, temperature variation and climate change until 2019. Summary of which is shown in table 1, 2 and 3.

Figure 4. Site pictures

Table 1. Annual daily traffic (Communication and Works Department, Punjab)

Vehicle	No.s (2017)	No.s (2018)	No.s (2019)
AD Vehicles	129	134	140
Motor cycles & Rickshaws	2825	2938	3056
Cars and Pickups	690	718	746
Mini Buses and Wagons	165	172	178

Buses and Flying Coaches	35	36	38
Trucks 2-Axle	93	97	101
Trucks 3-Axle	29	30	31
Tractor	21	22	23
Tractor Trollies 3-Axle	169	176	183
Tractor Trollies 4-Axle	0	0	0
Passenger Car Equivalent (P.C.U.s)	6004	6244	6474

Table 2. Monthly average temperature variation table for site (Wazirabad)

Weather	**Month**											
	Jan	**Feb**	**Mar**	**Apr**	**May**	**Jun**	**Jul**	**Aug**	**Sep**	**Oct**	**Nov**	**Dec**
Avg. Temperature in 2019 (°C)	12.3	15	20.3	26	31.2	33.9	31.4	30.1	29.5	25.4	18.6	13.5
Avg. Temperature in 2018 (°C)	11.9	13.6	20	27	30.8	34.1	29	32.3	27.3	25.2	19.5	13.4
Avg. Temperature in 2017(°C)	12.4	14.1	21.5	27.5	30.9	36	32.2	34.5	25.1	24.7	17.8	12.9

There is little rainfall throughout the year. Between the driest and wettest months, the difference in precipitation is 176 mm. The variation in temperatures throughout the year is 21.6 °C. All this temperature and climate conditions present the fact that, the pavement itself was exposed to thermal aging and other weathering effects.

Table 3. Table 3. Annual rainfall data for site (Wazirabad)

Months	**Jan**	**Feb**	**Mar**	**Apr**	**May**	**Jun**	**Jul**	**Aug**	**Sep**	**Oct**	**Nov**	**Dec**
Rainfall (mm) in 2019	35	34	32	19	18	45	159	183	74	10	7	16
Rainfall (mm) in 2018	34	36	32	20	21	38	170	179	95	9	7	9
Rainfall (mm) in 2017	36	33	33	21	19	47	145	189	98	11	8	13

7. Samples from asphalt wearing surface and asphaltic bases were extracted in the form of asphalt cores and blocks after more than three years of asphalt laying using core cutter machine for laboratory testing. Samples were extracted from high speed lane as well as medium speed lane from both sides of road section (within 10 meter section). Location of core extraction points are marked as M1 and H1 in the figure 5. Pictures of extracted core samples are shown in figure 6. From each of these two extraction points, two cores were extracted, one from asphaltic wearing and second from asphaltic base surface labelled as M1, M1',H1 and H1' (Letters with subscript ' represent samples from asphalt base surface and vice-versa).

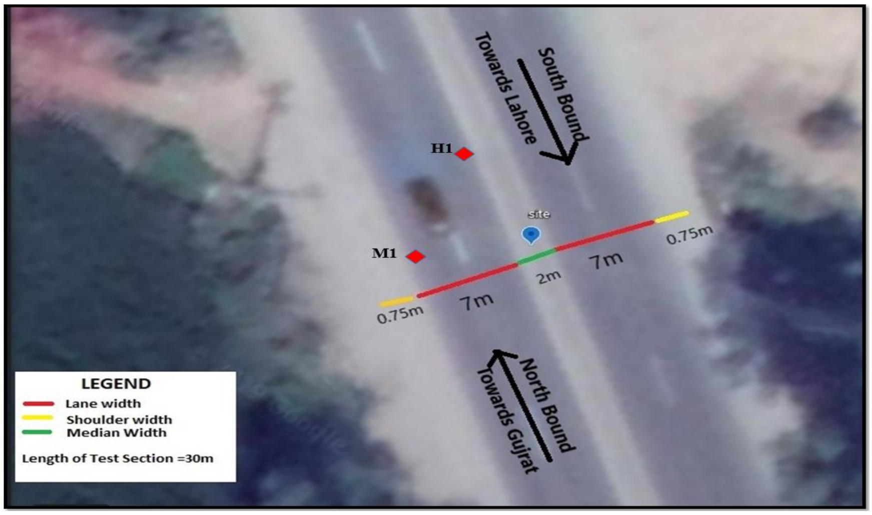

Figure 5. Location of core extraction

Figure 6. Extracted Asphalt Core from Site

8. Bulk specific gravity, Marshall stability and theoretical maximum specific gravity tests were performed on asphalt cores to determine the mix properties according to ASTM D-2726, D-6927, and D-2041. After performing these tests void-density relationship were calculated.

9. The solvent centrifuge extractor was used to extract bitumen from the reclaimed cores of asphalt wearing and base courses according to ASTM D-8159. Solvent (Trichloro-ethylene) was evaporated from the extracted bitumen using a rotatory evaporator according to ASTM D-5404.

10. Virgin bitumen (Grade 60/70) and extracted bitumen from asphaltic wearing sample were characterized in laboratory by performing Fourier transform infrared spectrometry.

11. Influence of traffic loading, temperature variation, moisture (precipitation) on chemical composition of binder were determined.

3.1. Fourier Transform Infrared Spectrometer

Fourier transform spectrometer will be used to classify the functional groups in the bitumen samples. The FTIR used for the testing is Attenuated total reflection (ATR) type spectrometer.
An ATR accessory operates by measuring the changes that occur in an internally reflected IR beam when the beam comes into contact with a sample. An IR beam is directed onto an optically dense crystal with a high refractive index at a certain

angle. This internal reflectance creates an evanescent wave that extends beyond the surface of the crystal into the sample held in contact with the crystal.

In regions of the IR spectrum where the sample absorbs energy, the evanescent wave will be attenuated. The attenuated beam returns to the crystal, then exits the opposite end of the crystal and is directed to the detector in the IR spectrometer. The detector records the attenuated IR beam as an interferogram signal, which can then be used to generate an IR spectrum.

IV. RESULTS AND DISCUSSIONS

1. Margalla crush (aggregates) were used in the preparation of asphalt mix that was laid in 2016 on the trial section. Results of aggregates properties for the asphalt mix design are tabulated in table 4.

Table 4. Aggregate tests

Test	Values	Limits
Elongation Index	10	10% Max
Flakiness	6	10% Max
Sand Equivalent	76	more than 45 %
Angularity	100%	90% minimum
Soundness in Sodium sulphate	Coarse – 7.2 Fine – 4.8	12% max
Los Angeles Abrasion	15%	40% max
Water Absorption	1.02	2% max

2. Typical ranges of aggregate passing percentage according to ASTM D-3515 were followed for the asphalt mix design as shown in table 5.

Table 5. Passing percentage limits for Asphalt Mix Design

Sieve Size	Percentage Passing by Weight – FHWA Limits (Wearing Course)	Percentage passing by weight – FHWA Limits (Base Course)
25mm	100	100
19mm	90-100	70-100
12.5mm	71-90	53-90
9.5mm	56-80	40-80
4.75mm	35-56	30-56
2.38mm	23-38	23-38
0.84mm	13-27	13-27
0.300mm	5-17	5-17
0.17mm	4-14	4-14
0.150mm	3-12	3-12
.075mm	2-10	2-10

3. The optimum design bitumen content for asphalt wearing is 4.3% whereas for asphaltic base course, design bitumen content is 4.00% as given by JMF design which was followed earlier.

4. The results of asphalt mix properties for both asphalt wearing and base samples are presented in table 8.

Table 6. Asphalt mix properties (AASHTO MS-2)

Description	JMF Specifications for Virgin Mix (Asphalt Wearing)	JMF Specifications for Virgin Mix (Asphalt Base)	Desirable Ranges (AASHTO MS-2)
Total Asphalt Content (Effective + Absorbed)	4.3%	4.00%	3-5 %
Bulk Specific Gravity	2.43	2.39	2.3-2.5
Theoretical Maximum Specific Gravity	2.53	2.50	2.5-2.7
Air Voids	4.00%	4.5%	4-7 %
Voids in Mineral Aggregates	12.93%	13.54%	12 minimum
Voids filled with Bitumen	69.06%	66.40%	60-75%
Marshall Stability	1802 Kg	1450 Kg	1000 Kg minimum
Marshall Flow	3.0 mm	3.30 mm	2-3.5 mm

5. Absorbance peaks of various functional groups for the virgin binder obtained from FT-IR spectrometer are shown below in figure 7 and are tabulated in table 9.

Table 7. FTIR: Absorbance peaks of various functional groups in virgin bitumen

Chemical Group	Transmittance %	Bond	Absorbance Peaks for Virgin bitumen sample		Typical range of functional groups
			Approximate Wavenumber (cm^{-1})	Intensity	
Alkanes	44%	C-H Stretch	2920.6	Strong	2850-3000
	53%	C-H Stretch	2851.4	Medium to Strong	2850-3000
α,β-Unsaturated Aldehydes, Ketones	85%	C=O Stretch	1680	Weak	1670-1720
Carbonyls	86%	C=C Stretch	1690	Weak	1660-1690
Amines	81%	N-H Bend	1650	Weak to Medium	1580-1650
Nitro Compounds	78%	N-O asymmetric stretch	1551	Weak to Medium	1300-1600
	72%		1375	Medium	
Aromatics	61%	C-C Stretch	1457	Medium	1300-1600
Sulfoxides	87%	S=O	1020	Very weak	1000-1070
Methyl Alkanes	80%	C-H rock	720	Weak	720-725

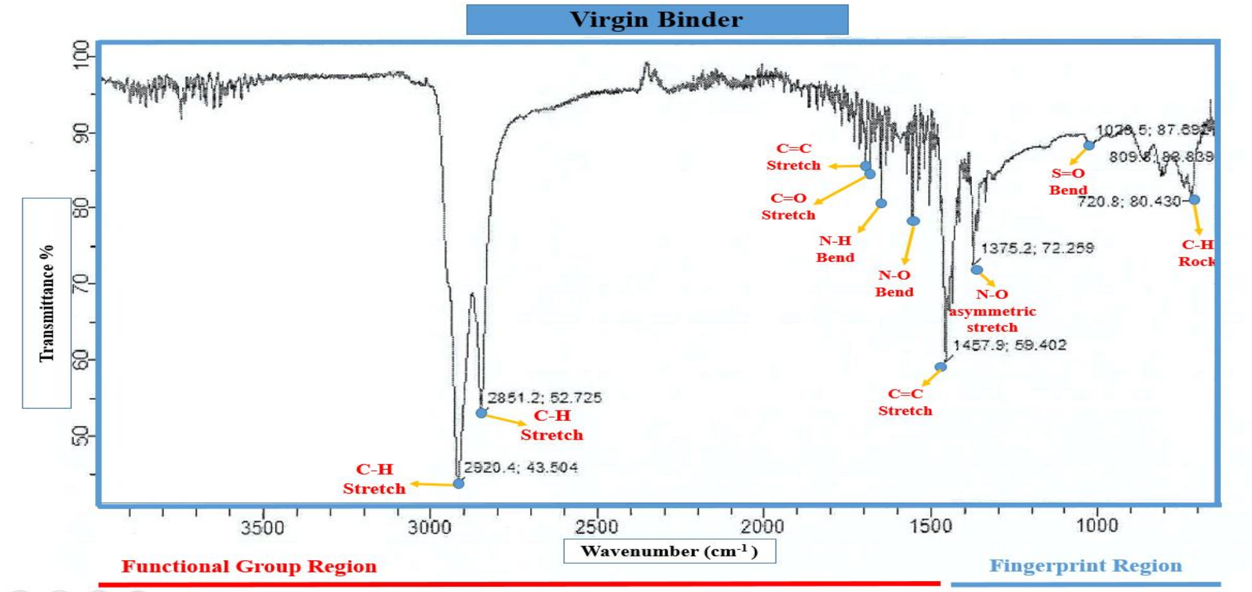

Figure 7. FTIR spectrograph of virgin bitumen sample

6. Gradation curves for both extracted asphalt wearing and base coarse samples obtained are shown in figure 8 and 9, whereas their passing percentages through different sieves are presented in table 10.

Table 8. Gradation comparison of asphalt base course and asphalt wearing course after aging

Sieve #	Sieve Size (mm)	Passing % (Asphaltic Base course)	Passing % (Asphaltic wearing course)	NHA Specification Limits for Passing %(Asphaltic Base course)	NHA Specification Limits for Passing %(Asphaltic wearing course)
#4	4.76	36.99	40.38	30-45	35-50
#10	2	16.49	22.56	15-35	20-35
#40	0.42	5.43	10.23	5-16	5-12
#100	0.15	2.38	3.24	2-8	2-8
#200	0.075	0.35	0.44	2-7	2-7

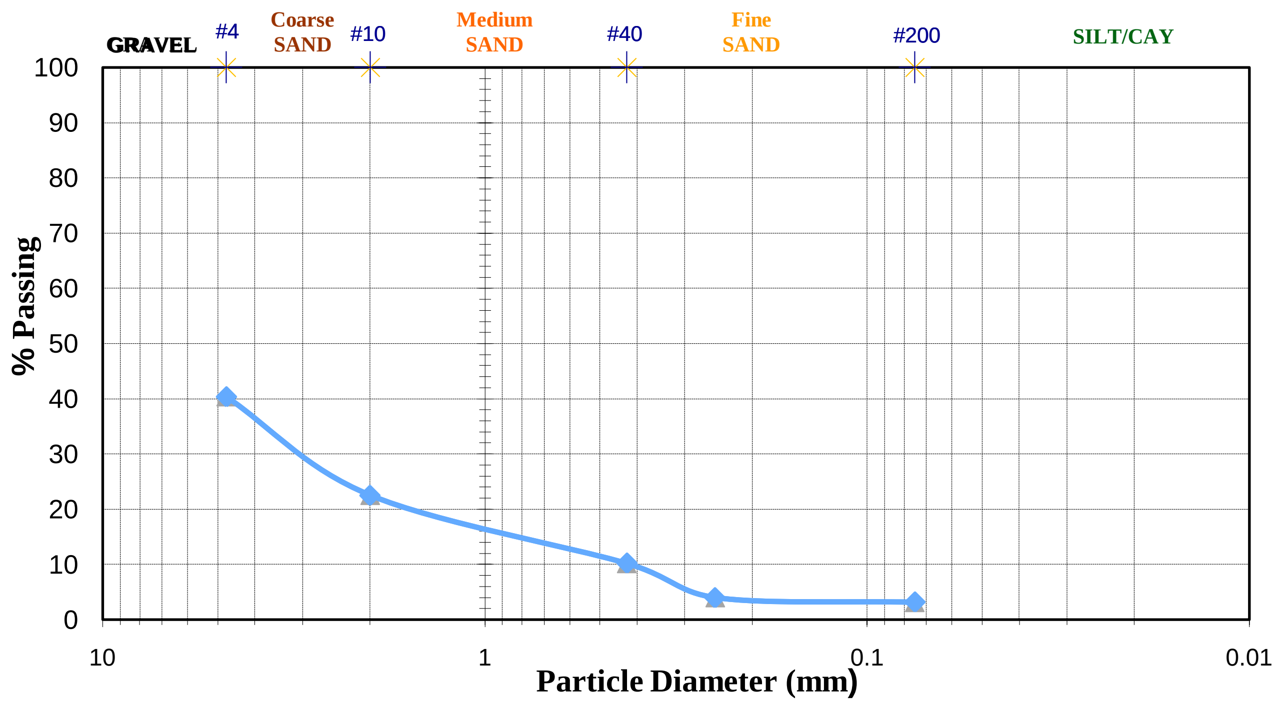

Figure 8. Grain size distribution in the asphalt wearing course

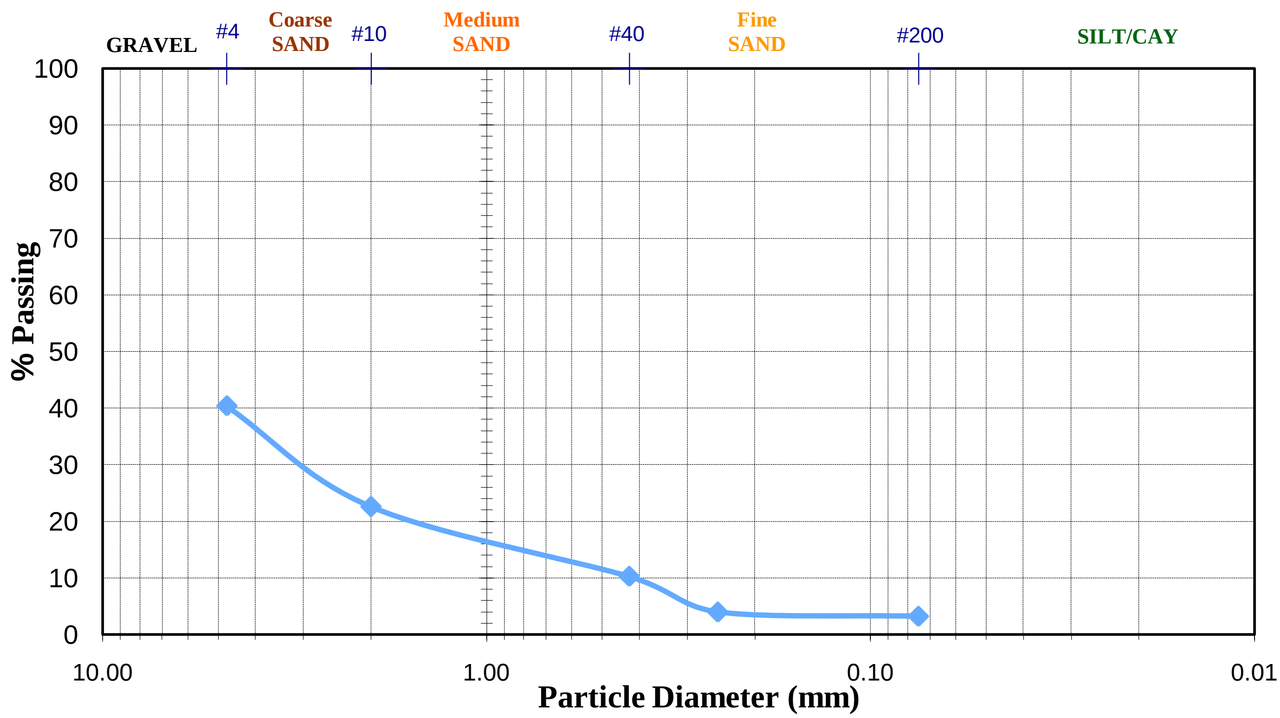

Figure 9. Grain size distribution in the asphalt base course

7. Asphalt mix properties for aged samples of both asphalt wearing and base course according to AASHTO MS-2 are presented in the table 11.

Table 11. Asphalt mix properties for aged samples

Description	**Asphalt Wearing Course**		**Asphalt Base Course**		**Desirable Ranges (MS-2)**
	Aged Sample M1	**Aged Sample H1**	**Aged Sample M1'**	**Aged Sample H1'**	
Total Asphalt Content (Effective + Absorbed)	3.78%	3.81%	3.61	3.64	3-5 %
Bulk Specific Gravity	2.31	2.32	2.28	2.29	2.3-2.5
Theoretical Maximum Specific Gravity	2.52	2.52	2.51	2.51	2.5-2.7
Air Voids	8.33%	7.93%	9.16	8.76%	4-7 %
Voids in Mineral Aggregates	16.82%	16.51%	17.20	16.82%	12 minimum
Voids filled with Bitumen	76.21%	75.77%	46.7	47.91%	60-75%
Marshall Stability	1420 Kg	1450 kg	1050 Kg	1070Kg	1000 Kg minimum
Marshall Flow	3.8 mm	3.7mm	1.50 mm	1.6 mm	2-3.5 mm

8. Absorbance peaks of various functional groups for the virgin binder obtained from FT-IR spectrometer are shown below in figure 7 and tabulated in table 12.

Table 12. Absorbance peaks for various functional groups of aged bitumen

Chemical Group	**Transmittance %**	**Bond**	**Absorbance Peaks for aged bitumen sample**		**Typical Range of Functional groups**
			Approximate Wavenumber (cm^{-1})	**Intensity**	
Alkanes	46%	C-H Stretch	2920.6	Strong	2850-3000
	55%	C-H Stretch	2851.4	Medium to Strong	2850-3000
α,β-Unsaturated Aldehydes, Ketones	78%	C=O Stretch	1680	Weak to Medium	1670-1720
Carbonyls Alkenes	80%	C=C Stretch	1690	Weak	1660-1690
Amines	81%	N-H Bend	1650	Medium	1580-1650
Nitro Compounds	78%	N-O asymmetric stretch	1551	Medium	1300-1600
	70%		1375	Medium to Strong	
Aromatics	57%	C=C Stretch	1457	Medium	1300-1600
Sulfoxides	74%	S=O Stretch	1018	Medium, Broad	1000-1070
Methyl Alkanes	76%	C-H rock	721	Medium	720-725

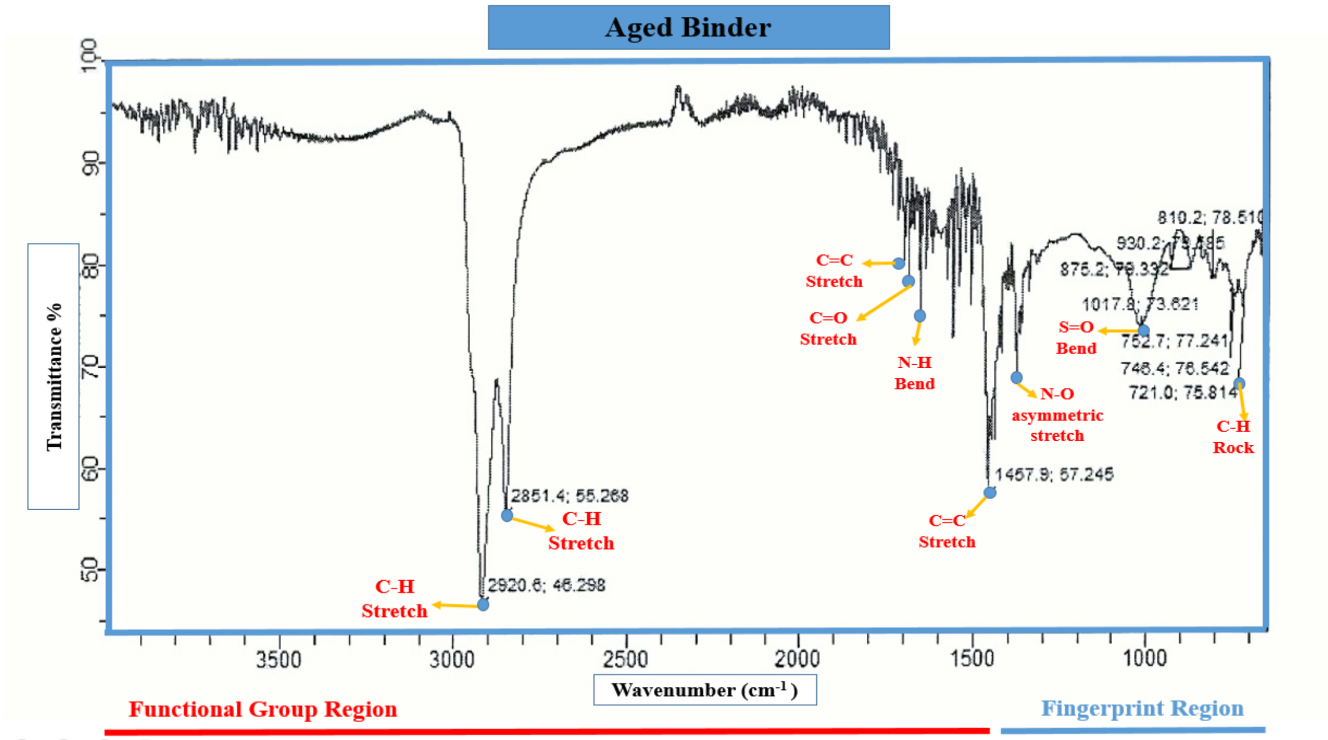

Figure 10. FTIR spectrograph of aged bitumen sample

9. On comparing gradation of extracted asphalt core samples to standard gradations for asphalt mixes, it can be seen that the aggregate surface area was increased, caused by disintegration of aggregates by high traffic loading and weathering effects over the years.

10. Asphalt content in the aged wearing coarse sample is 3.78% as compared to 4.3% of virgin mix as specified by JMF design. This shows a reduction of 15% in bitumen content has occurred due to volatilization and oxidation which affects its rheological properties.

11. Bulk specific gravity is reduced by 5% because of loss of bitumen and wear and tear of aggregatesas compared to the time when it was laid initially where compaction requirements were fulfilled.

12. Theoretical maximum specific gravity have undergone negligible change because it is air tight or void-less density of the mix. Voids in both conditions i.e. virgin and aged should be negligible and hence these values are very close to each other as shown in the table 11.

13. Voids in total mix have increased immensely from 4 % at the time of laying to 8.33% due to gradual decrease in density because of bitumen loss, disintegration of aggregates, increased surface area which in turn causes loos packing.

14. Marshall stability for the wearing core sample is 1420 Kg which is a downgrade of 26% from its freshly prepared asphalt mix test value whereas in case of base coarse, stability is decreased to 1050 kg from its freshly prepared mix test value of 1450 Kg. This is understandable because the aged asphalt mix has a lesser density due to irregularities that has happened in its structure under the varyingly distributed traffic loads and stripping due to water absorption.

15. It was observed that the low quality aggregates can often produce mixes with maximum Marshal stability because higher quality aggregates are more prone to crushing earlier due to high brittleness. It also conforms with the research conducted byAbdul Rahman S. Al-Suhaibani.

16. Marshall flow value for both asphalt wearing and base layers vary. As for Asphalt wearing, it shows a deformation of 3.8 mm before it cracked. This is significantly higher than 1.5 mm value of asphaltic base. It is because asphaltic wearing surface consists of higher quality material with a very dense packing so, it sustained more load and deformed more before cracking. Whereas, asphaltic base includes slightly larger aggregates particles as well which imparts less bearing strength to the mix compared to wearing surface. This is why asphaltic base cracked earlier showing much less deformation.

17. The difference in test values between asphalt base and wearing cores lie in their gradation and packing characteristics. Asphaltic base consists of slightly coarser aggregates particles which gives more void space in between aggregates. On the other hand, asphaltic wearing layers are made up of smaller, more angular particles imparting more surface area and less air voids.

18. In the spectrograph of virgin binder and aged binder, obvious peaks are at 2920 cm^{-1}, 2851 cm^{-1}, 1457 cm^{-1}, 1375 cm^{-1} and 720 cm^{-1}. In the functional group region of the spectrograph, from wavenumber 3100 cm^{-1} to 3500 cm^{-1}, there is a slight increase in concentration of hydroxyl O-H ions due to some likely retention of water molecules (hydrogen bonding) as a result of weathering effects over the years. Moreover, bitumen oxidation was found very complex because, there were not isolated molecules but rather thousands of compounds sharing a similar aromaticity and length of carbon chains up to staggering c^{150} atoms. But it is only possible to analyze few proportions of carbon, hydrogen and Sulphur bonds. It was seen that sulfoxides functional groups were formed earlier as a result of short term aging, whereas carbonyl groups formed later after long term aging of the bitumen. The major difference lie in the fingerprint region, where carbonyl and sulfoxides bands are usually present. There are additional peaks created due to creation of C=O and S=O. These additional carbonyl (C=O) groups were produced due to absorption of oxygen in unsaturated carbon chain. Their concentration is increased as evident from the spectrographs which shows that the oxidative aging has occurred. From a chemical point of view, aging is caused by irreversible oxidation and volatilization, itself due to the presence of oxygen in air, thus creating carbonyl (C=O) and sulfoxide (S=O) groups, modifying bitumen composition (increase in molecular size, bitumen polarity and aromaticity) and hardening the bituminous binder which becomes more brittle.

19. Carboxylic acids, ketones, anhydrides, sulfoxides, pyrrolic types, 2-quinolone types and phenolic types are the hetro-atom functional groups which were present initially in bitumen. After oxidation average increase in concentration of compounds of ketones, anhydrides, carboxylic acid, and sulfoxides is 0.55, 0.20, 0.005 and 0.29 mol/L respectively.

20. From the table 9 and 12, it is deduced that the transmittance % is decreased for most of the functional groups except alkanes. This decrease in transmittance is accounted for evaporation of volatile components due to surrounding temperature, as a result of which binder mass and volume is reduced. The bond strength for these functional groups have increased i.e. bonds have become rigid, making bitumen hard and eventually brittle.

21. It can also be seen that there is 15% increase in carbonyl and sulfoxide absorption. This increase in these functional groups is due to oxidation. As a result of oxidation C-H bonds break. Oxygen atoms reacts with free carbon, sulfur and hydrogen atoms thus creating increased concentration of S=O, C=O and O-H functional groups in aged bitumen. Due to oxidation there is an evolution in molecular structure of bitumen due to which in aged bitumen the threads have become more angular and very small in size as compared to that of virgin bitumen.

22. The moisture only affects the surface of bitumen that is indicative by FT-IR spectrograph as there are peaks at 3300 cm^{-1} and 1600 cm^{-1}. But after, 4 hours of drying, most of the water was evaporated easily at room temperature. This shows that water was only absorbed on to the surface of bitumen and did not affect the internal molecular structure. The most prominent peaks, remained even after the normal drying were OH and NH stretch, carbonyl stretching, sulfoxides, alcohol, ether, ester and sulfate groups. But, these peaks were also completely reversed by oven drying the sample at 150 °C for 30 mins.

V. CONCLUSIONS

It is concluded from this research project that the difference between virgin bitumen as well as virgin asphalt mix from aged specimens is evident by the resulting test values. The asphalt mix design properties varied significantly from the fresh prepared mixture properties mainly because of oxidation, consistent traffic loads and weathering effects which disturbs the internal aggregate packing characteristics of pavements. As a result, the pavement structure deteriorates over time.

As far as bitumen is concerned, Fourier transform infrared spectrometer clearly shows the increase in carbonyl and sulfoxides band peaks which is a major marker of aging in the bitumen. Aging through oxidation is clearly an irreversible chemical reaction which tends to increase the molecular size, polarity and aromaticity in the bitumen sample making the bitumen hard and brittle. Asphalt becomes brittle due to aging. Its damage tolerance is reduced and it becomes less durable. Hence, resulting bitumen then neither have required binding properties nor any strength imparting properties.

VI. ACKNOWLEDGEMENTS

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