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Surface morphology, Raman spectroscopy and chromaticity analysis of Indian automobile paint samples and its forensic application

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Abstract

Trace evidence, such as paint flakes, is considered substantial evidence in criminal and road accident investigations. Comparison and identification of paint evidence is considered the most challenging task in forensic analysis. The paint evidence is very often transferred onto the clothing, automobiles, weapons, etc. from the victim as well as the crime scene when there is an impact with automobiles. The present work establishes surface morphology of paint samples using a Raman microscope and vibrational energy shift using a laser Raman spectrometer instrument. Also, the chromaticity of paint samples was analyzed using a microspectrophotometer instrument (MSP), and their chromaticity coordinates (u', v') were mapped to assess color composition and forensic significance. The surface texture of the paint samples exhibits fine granularity with dispersed dark spots, likely corresponding to pigment particles. The Raman spectral analysis of the paint sample suggests a composition primarily consisting of various pigments such as hematite (α-Fe_2O_3), iron oxide-based pigments, titanium dioxide, etc., and the presence of aromatic compounds, or binder materials. The chromaticity coordinates (u',v') of the analyzed paint samples are concentrated in the yellow-green spectral region, with values ranging approximately from (0.222, 0.530) to (0.312, 0.540). This suggests that the dominant hues are in the yellowish-green to yellow-orange range.

Keywords: Automobile paint flakes, Laser raman spectrometer, MSP**,** chromaticity**.**

Introduction

The trace evidence plays an important role to link the suspected vehicle to crime scene in the investigation of crimes related to road accidents, robbery, hit and run cases[1]. One of the most often analysed criminalistic traces is small paint flakes that were found on a victim's clothing or taken from the scene of an automobile accident. It is possible to identify the vehicle involved in the collision by comparing them to a paint sample taken from a suspected vehicle. Car paint samples are routinely examined using a variety of optical microscopy and spectroscopy techniques that use the entire spectrum of electromagnetic radiation[2].

Main purpose of the paint coated on automobiles is for protection and decoration. Due to its less detrimental effects on the environment, water-based acrylic polyurethane enamel paint is currently the most extensively used paint type[3]. Modern used automobile paint is applied with a thickness of 100 µm (0.1mm). Automobile paint consists of mainly three components; binder, pigment and additives[4].

Differences in primers and topcoats result in noticeable disparities among car coatings. Some variations in paint formulation are due to requirements such as durability and high quality physical appearance. Acrylic lacquers, acrylic enamels, and alkyd enamels are the major categories present in the automobile paints presently.

The use of high-solid-content acrylic enamels with the clear coat/base coat system[5-6] is the current trend in automotive finishing, although general motors' vehicles were covered with acrylic lacquers[7] for many years. This research focuses on automobile paint analysis with the primary utilization of Laser Raman spectrometer and microscope, micro spectrophotometer incorporated into the data analysis which covers a variety of approaches in its pursuit of discriminating between paint samples.

Materials and Methods

Collection of samples: In the present study, total ten paint flakes of vehicles such as Hyundai cars, Maruthi, Ashok Leyland and Force including trucks and buses, were collected. All the ten samples were chosen to analyze surface morphology, vibrational energy shift and their chromaticity coordinates (u', v') were mapped to assess color composition and forensic significance.

The registration number, model number, name, and paint collecting points of the individual automobile were recorded for references. The different paint samples were further marked as follows for further reference.

Table-1: Markings of paint flakes with automobile details.

Sample code	Vehicle model	Color of the paint	Vehicle name	Sample code	Vehicle model	Color of the paint	Vehicle name
APF1	2021	White	Hero Destine 125 vx (se)	APF6	2018	White	Maruthi Swift Dzire
APF2	2015	White	Chevrolet Cruze LTZ	APF7	2014	Grey	Chevrolet Enjoy
APF3	2005	Silver Color	Hyundai Elantra	APF8	2020	White	Force Traveller
APF4	2016	Cream Color	Tata Ace Facelift Ht BS III	APF9	2018	Red	Ashok leyland ltd,
APF5	2020	Red	Maruthi Swift	APF10	2018	Green	Ashok leyland ltd

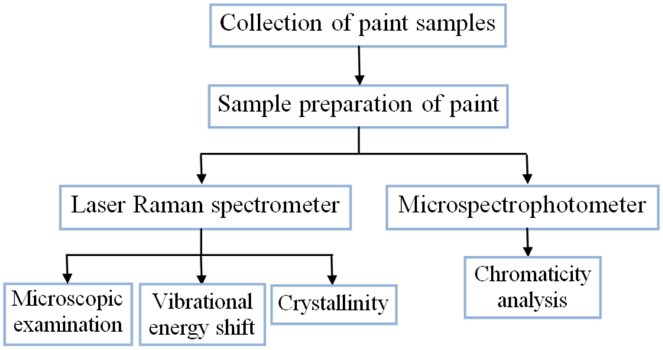

Figure-1: Analysis flow chart of paint flakes.

Experimental techniques: Microscopic examination: All the paint flakes were examined using Leica microsystems built in Laser Raman spectrometer under different magnification and recorded the surface colour, surface morphology and other information.

Vibrational spectroscopy using Laser Raman spectrometer: Vibrational spectroscopy involves the study of molecular vibrations to gain insights into chemical composition, molecular structure, and intermolecular interactions. Laser Raman spectroscopy is a powerful vibrational spectroscopic technique used for qualitative and quantitative analysis of materials. The vibrational spectra of paint samples were analyzed using Renishaw inVia Laser Raman spectrometer instrument. The Raman spectra of Raman shift/cm-1 as function of counts were recorded in laser wavelength 785 nm with laser power 0.5% and grating 1200 l/mm.

Microspectrometry: Microspectrometry is an advanced analytical technique that integrates optical microscopy with spectroscopic methods (such as Raman, infrared, or UV-Vis spectroscopy) for high-resolution chemical and structural analysis at the microscopic level.

It is widely used in forensic science, materials research, and life sciences. The chromaticity coordinates (u', v') were mapped to assess color composition using Lucia Microspectrometry instrument.

Results and Discussion

Surface morphology of paint samples using Raman microscope: The micrograph of the paint sample labeled as APF-1 is shown Figure-1(a). The micrograph having minimal irregularities with relatively smooth. The surface morphology exhibits fine granularity with dispersed dark spots may due to the presence of pigment particles or impurities within the paint matrix. The uniformity suggests a well-mixed composition with minimal mechanical wear or exposure to environmental degradation.

The micrograph of the paint sample labeled as APF-2 is shown in Figure-1(b). The micrograph shows a network of tiny scratches and lengthy grooves that indicate a more noticeable surface roughness. The abrasions found on paint surface potentially due to various environmental factors such as external force. The dark spots dispersed on surface represent pigment clusters or impurities. The linear features of abrasions making it suitable for forensic analysis in comparative trace evidence studies.

The micrograph of the paint sample labeled as APF-3 is shown Figure-2(a). The micrograph of the paint sample presents a highly heterogeneous and coarse structure with aggregate texture having bright and dark spots suggesting different pigment components and fillers present in the paint formulation. The morphology also exhibits green and shiny silver/ white spots over the area may indicating the chromium-based pigments or phthalocyanine green and titanium dioxide.

The micrograph of the paint sample labeled as APF-4 is shown Figure-2(b) reveals a relatively smooth, non shiny and homogeneous surface with few dark spots scattered over area. These spots could correspond to embedded pigment particles or impurities in the paint matrix.

The micrograph of the paint sample labeled as APF-5 is shown Figure-3(a) shows the relatively homogenous and dominated by pink to reddish matrix which usually represents a single layer pigments and sometimes a undercoat. The paint texture also exhibits the faint linear streaks over the surface, likely due to the brushing during the paint deposition.

The micrograph of the paint sample labeled as APF-6 is shown Figure-3(b) reveals a relatively smooth, non shiny and homogeneous surface with few dark spots scattered over area. These spots could correspond to embedded pigment particles or impurities in the paint matrix.

The micrograph of the paint sample labeled as APF-7 is shown Figure-4(a) reveals a high reflectivity surface. The micrograph appears to be bright spots with reflective particles distributed over area. Such type morphology generally suggests likely metal flakes such as metallic finish used in metallic or pearlescent automotive finishes**.**

The micrograph of the paint sample labeled as APF-8 is shown Figure-4(a) exhibits smooth and pale surface. The appearance of the surface is uniform, homogenous and light colored possibly beige or off-white, suggesting a non-pigment-intensive layer**.** The presence of minor dark inclusions, possibly contaminants, indicates a relatively homogeneous but slightly worn surface.

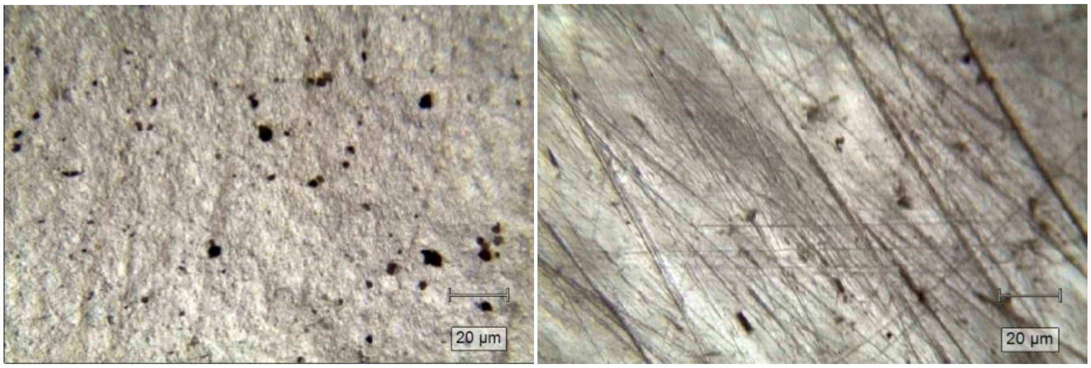

Figure-1: Surface morphology of paint samples a) APF-1, b) APF-2.

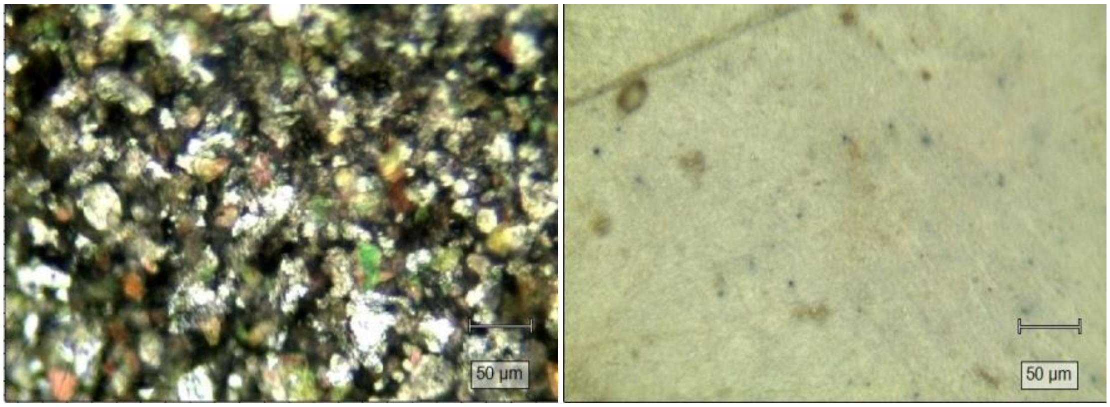

Figure-2: Surface morphology of paint samples a) APF-3, b) APF-4.

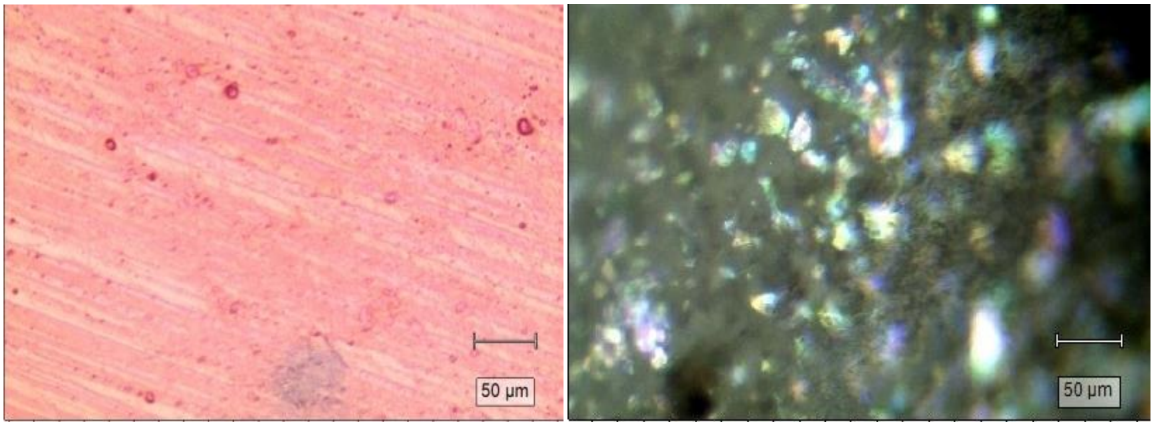

Figure-3: Surface morphology of paint samples using Raman microscopy a) APF-5, b) APF-6.

The micrograph of the paint sample labeled as APF-9 is shown Figure-5(a) exhibits uniform reddish hue. The paint texture appears to be dominant reddish or terracotta coloration which commonly representing presence of iron oxide pigments. On top right corner of the surface exhibits the slightly mottled area which may due to the indicate wear, corrosion, or variation in pigment dispersion.

The micrograph of the paint sample labeled as APF-10 is shown Figure-5(b) exhibits uniform green appearance. The entire texture area having consistence green tone which suggests the presence of green pigments such as Chromium oxide green or phthalocyanine green.

Vibrational spectroscopy using Raman spectrometer of paint samples: The Raman spectra of the APF-1 paint sample is shown in Figure-6(a) as a function of Raman shift/cm^{-1}. The peak observed at 142cm^{-1} indicates low-frequency peak is likely belongs to metal-oxide stretching vibrations usually observed in iron oxide pigments mainly hematite or binding media used in the paint. The peak observed at 232 cm^{-1} suggests iron oxide based pigments or titanium dioxide in the rutile phase. Since TiO_2 is widely used as white pigment and which is Raman-active mode in this region. The strong peak observed at 447cm^{-1} is a characteristic peak of rutile-phase titanium dioxide, which is widely used in white paint composition mainly due to its great opacity and stability. The peak positioned at 661cm^{-1} is corresponding to iron oxide phases such as Fe_2O_3 hematite or carbon black. Carbon-based pigments usually indicates the existence of colorants or additives[8-10].

The Raman spectrum of the white APF-2 paint sample is presented in Figure-6(b) as a function of Raman shift/cm^{-1}. The peak positioned at 220cm^{-1} indicates presence of metal oxides mainly related to iron oxide pigments or binding agent. The strong peak observed at 447cm^{-1} is a characteristic peak of rutile-phase titanium dioxide, which is widely used in white paint composition mainly due to its great opacity and stability. The peak positioned at 612cm^{-1} belongs to iron oxide such as Fe_2O_3, hematite or chromium-based pigments. The peak observed at 1004cm^{-1} suggests the presence of organic compounds which are originating from binders, polymeric additives or resin components[11-12].

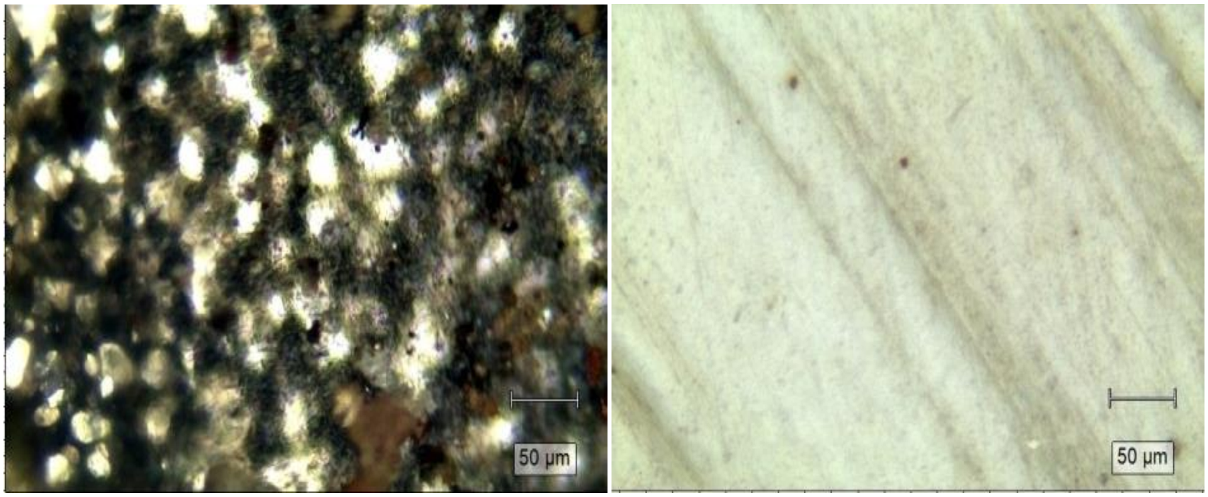

Figure-4: Surface morphology of paint samples using Raman microscopy a) APF-7, b) APF-8.

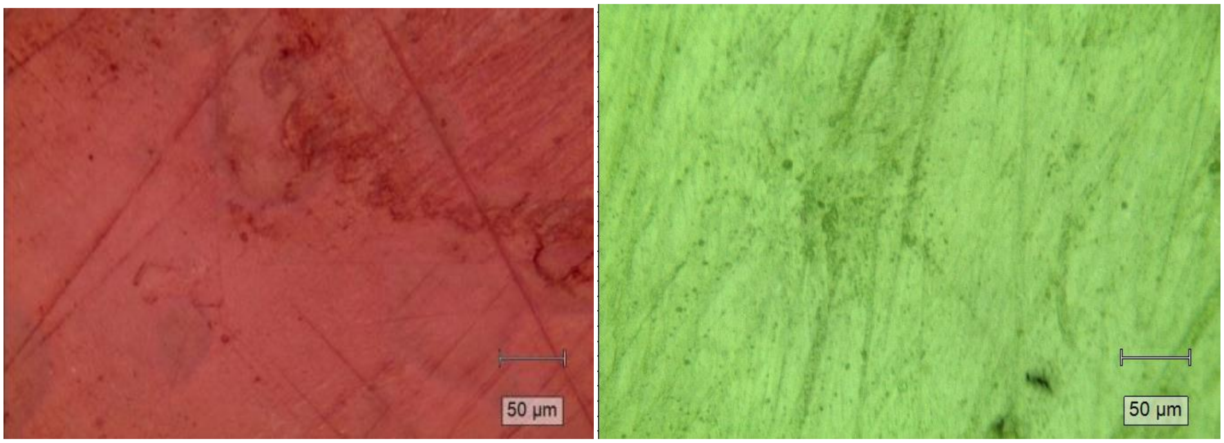

Figure-5: Surface morphology of paint samples using Raman microscopy a) APF-9, b) APF-10.

Table-2: Comparison of micrographs of paint samples.

Sample code	Surface morphology description	Key observations	Forensic significance
APF-1	Smooth with minimal irregularities and fine granularity	Dark spots possibly due to pigment particles or impurities; uniform texture	Suggests well-mixed paint composition with minimal wear; useful for matching to source
APF-2	Rough surface with scratches and grooves	Abrasions and pigment clusters visible; linear features present	Surface wear indicative of environmental exposure; valuable for trace comparisons
APF-3	Coarse and heterogeneous texture	Bright and dark spots; presence of green and silver/white pigments	Suggests multi-pigment formula with fillers (e.g., titanium dioxide, chromium-based pigments); useful in pigment analysis
APF-4	Smooth, non-shiny and homogeneous	Few scattered dark spots indicating pigment particles or impurities	Homogeneous surface aids in layer and pigment consistency checks
APF-5	Homogeneous reddish surface with faint streaks	Brush-induced linear marks; uniform pink to reddish hue	Indicative of single pigment layer or undercoat; brush pattern useful in application technique analysis
APF-6	Smooth, non-shiny and homogeneous	Sparse dark inclusions; pigment or impurity origin	Paint uniformity supports pigment ID and matrix homogeneity
APF-7	Reflective surface with bright shiny spots	Presence of reflective particles, likely metal flakes	Suggests metallic/pearlescent automotive finish; crucial for vehicle paint matching
APF-8	Smooth and pale (beige/off-white)	Minor dark inclusions suggest light wear or contamination	Non-intensive pigment layer; can assist in substrate or primer layer identification
APF-9	Uniform reddish/terracotta surface	Slight mottling in top corner; iron oxide pigment presence	Helps identify corrosion or pigment variation; significant for origin determination
APF-10	Uniform green surface	Consistent tone, suggests chromium oxide or phthalocyanine green pigment	Strong indicator of specific pigment use; assists in classifying paint type and potential source

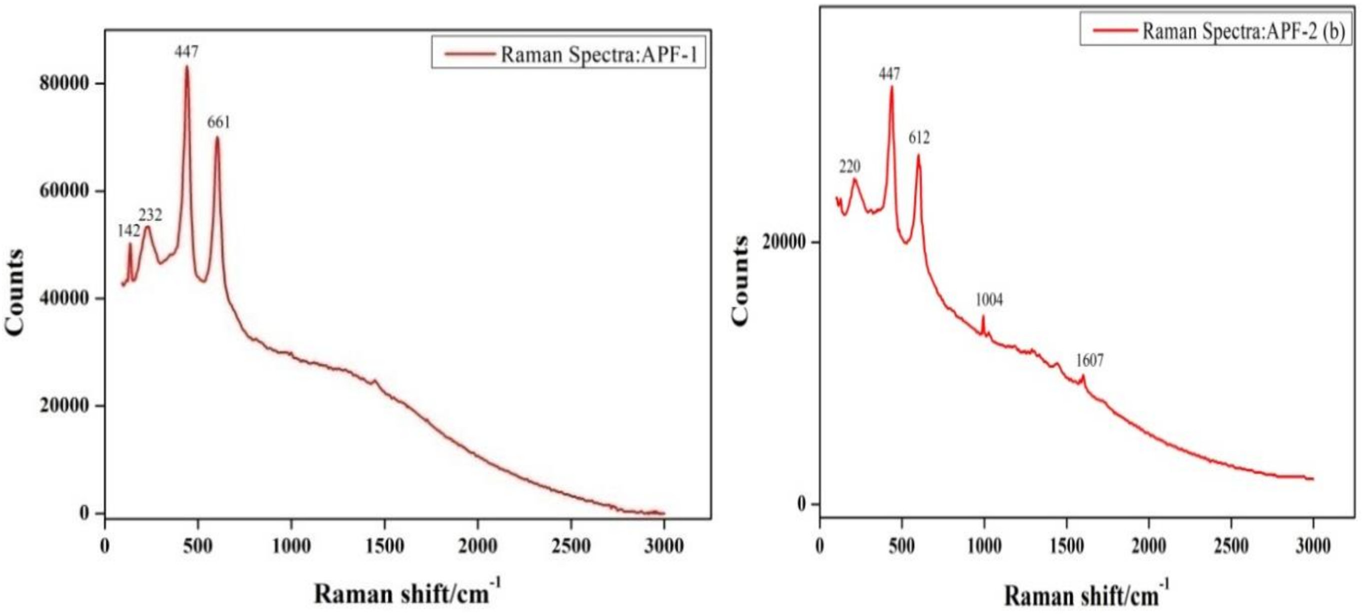

Figure-6: Raman spectra of paint samples a) APF-1, b) APF-2.

The Raman spectra of the APF-3 paint sample is presented in Figure-7(a) as a function of Raman shift/cm^{-1}. The peak observed at 622cm^{-1} is associated with iron oxide such as Fe_2O_3, hematite or chromium-based pigments[9]. The strong and characteristic peak positioned at 1005cm^{-1} is attributed to organic compounds such as styrene, aromatic resins, or polymer-based additives[13]. The peak at 1036cm^{-1} peak is due to the presence of organic pigments, aromatic compounds, or binder materials[14]. The peak positioned at 1193cm^{-1} is C–H bending vibrations from aromatic hydrocarbons present in organic pigments/ polymer-based binders/ quinacridone-based pigments. A peak position 1455cm^{-1} belongs to C–H deformation vibrations in alkyd resins or acrylic-based paints. A peak at 1609cm^{-1} is typically linked to carbon-based pigments or aromatic C=C stretching modes.

The Raman spectra of the APF-4 paint sample is presented in Figure-7(b). The strong at 229cm^{-1} is low-frequency vibrational modes associated to iron oxide pigments such as hematite. Another significant peak at 449cm^{-1} indicates presence of titanium dioxide in its rutile phase. The presence of this peak is consistent with previous studies on rutile-phase TiO_2. Additionally, the peak at 618cm^{-1} may be attributed to iron oxide phases such as magnetite (Fe_2O_3) or hematite (Fe_2O_3). The peaks at 1005cm^{-1}, 1320cm^{-1}, and 1606cm^{-1} strongly suggests the presence of organic binders, polymeric additives, and carbon-based materials in paint matrix.

The Raman spectra of the APF-5 paint sample is presented in Figure-8(a). The peak positioned at 127cm^{-1} indicates the existence of metal-oxide stretching vibrations such as iron oxide or lead-based pigments. The peaks at 351cm^{-1} and 452cm^{-1} are indicating titanium dioxide in the rutile phase[15]. Additionally, the peaks positioned at 582cm^{-1} and 622cm^{-1} are due to the contributions from hematite/magnetite (Fe_2O_3) which are commonly found in red and brown pigments[16]. The peaks positioned at 1201cm^{-1}, 1310cm^{-1} and 1349cm^{-1} exhibits existence of the aromatic C=C stretching vibrations, suggesting organic dyes or polymeric resins. The most intense peak at 1598 cm^{-1} is characteristic of carbonaceous materials, possibly from organic pigments, polymeric binders, or degradation products.

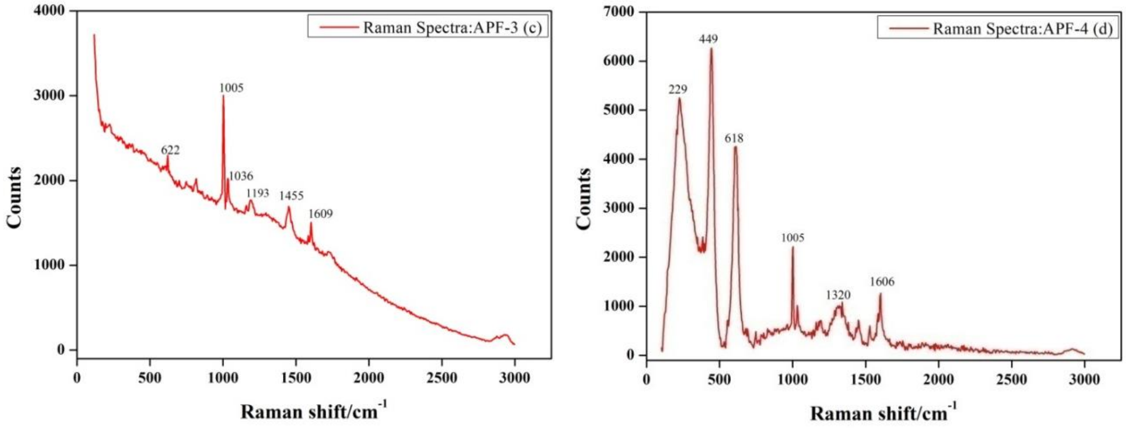

Figure-7: Raman spectra of paint samples a) APF-3, b) APF-4.

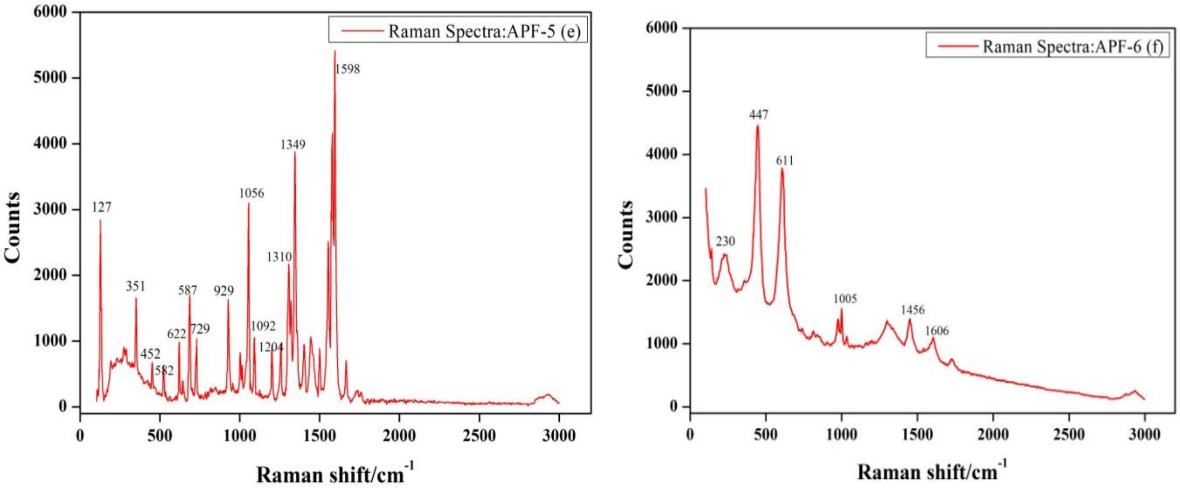

Figure-8: Raman spectra of paint samples a) APF-5, b) APF-6.

The Raman spectra of the APF-6 paint sample is presented in Figure-8(b). The peak exist at 230cm⁻¹ associated to metal-oxide stretching vibrations. The prominent peak at 447cm⁻¹ belongs to the symmetric stretching modes of titanium dioxide in the rutile phase. The peak at 611cm⁻¹ indicates iron oxide phases.

The Raman spectra of the APF-7 paint sample is presented in Figure-9(a). The positioned at 622cm⁻¹ is mainly due to the metal-oxide stretching vibrations. Another strong peak found at 1005cm⁻¹ belongs to the aromatic ring breathing mode, suggesting the presence of organic pigments/ binders[13]. The peak at 1312cm⁻¹, 1607cm⁻¹ is associated to C–H deformation vibrations and to C=C stretching in aromatic rings.

The Raman spectra of the APF-8 paint sample is presented in Figure-9(b). The peak positioned at 228cm⁻¹ is associated to lattice vibrations of metal oxides mainly titanium dioxide in its rutile form. A prominent peak at 447cm⁻¹ supports the existence of rutile-phase. The peak positioned at 612cm⁻¹ indicates the presence of hematite/ magnetite and peak 1005 cm⁻¹ is indicative of aromatic ring breathing modes.

The Raman spectra of the APF-9 paint sample is presented in Figure-10(a). The peaks positioned at 128cm⁻¹ is associated metal oxides which is mainly present in pigments such as titanium dioxide (TiO_2) and peak and 351cm⁻¹ suggests the contribution of iron oxide phases like hematite/ goethite, which are often used as red and yellow pigments in paints. The peaks at 622cm⁻¹ and 688cm⁻¹ is mainly due to the stretching modes of metal-oxygen bonds such as chromium/ iron oxide pigments. The peak at 930cm⁻¹ is indicative of C–C stretching vibrations and notable peak positioned at 1056cm⁻¹ belongs to sulfate stretching vibrations indicating presence of barium sulfate as paint extender or filler. The peaks at 1310cm⁻¹ and 1348cm⁻¹ can be attributed to C–H bending vibrations. The most intense peaks at 1581cm⁻¹ and 1598cm⁻¹ attributed to stretching vibrations aromatic ring suggesting existence of organic pigments i.e., azo dyes or phthalocyanines.

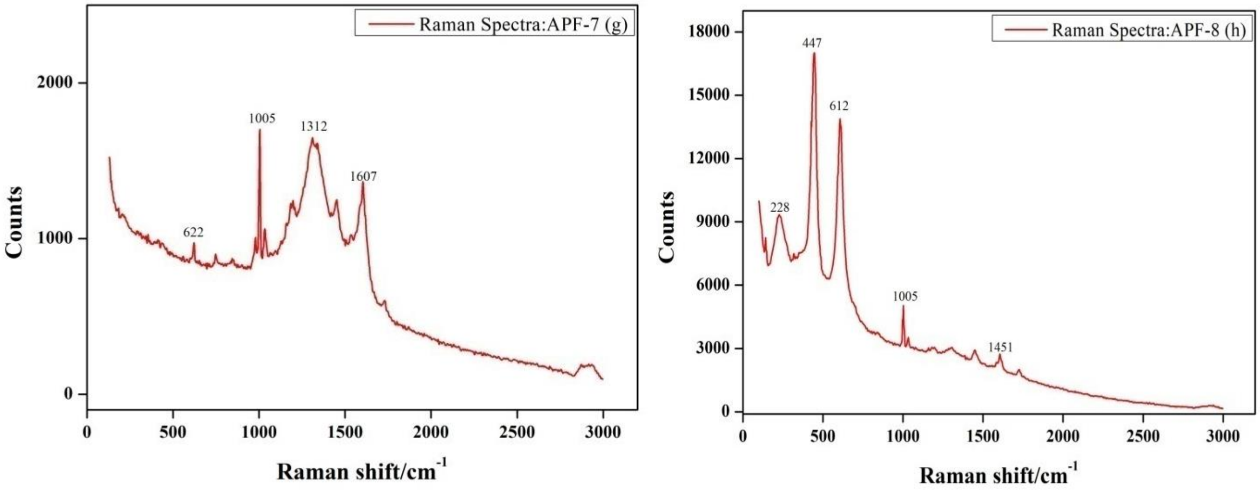

Figure-9: Raman spectra of paint samples a) APF-7, b) APF-8.

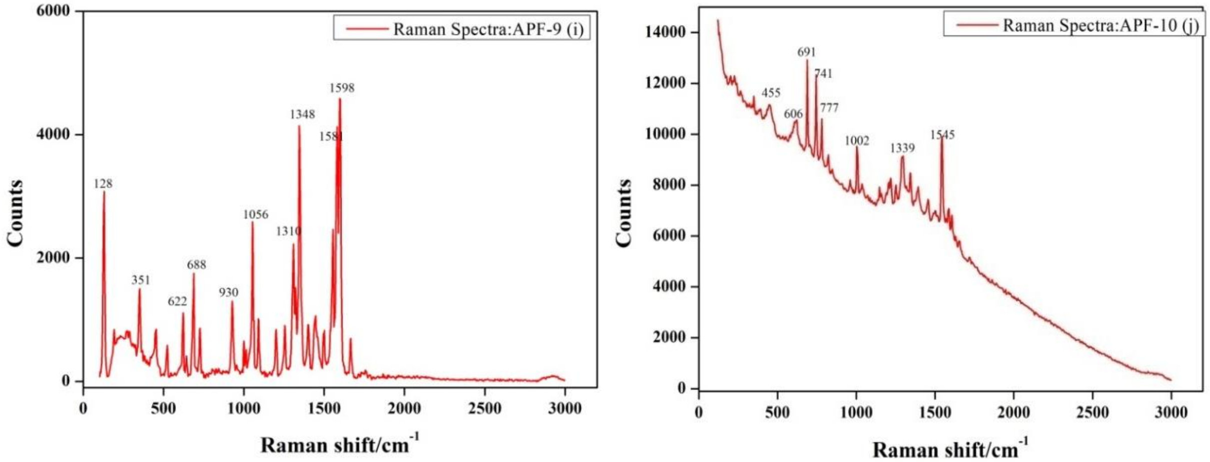

Figure-10: Raman spectra of paint samples a) APF-9, b) APF-10.

The Raman spectra of the APF-9 paint sample is presented in Figure-10(b). The peaks positioned at 455cm^{-1} and 606cm^{-1} attributed to the metal-oxygen stretching vibrations associated with TiO_2 and Fe_2O_3. The peak observed at 691cm^{-1} is due to symmetric stretching modes of metal-oxygen bonds likely from Cr_2O_3 or ferric oxide pigments. The 741cm^{-1} and 777 cm^{-1} peaks are indicative of ring breathing modes in organic pigments i.e., phthalocyanine-based or azo dyes.

Crystallinity analysis of paint samples: The application of Raman spectroscopy in characterization of crystallinity is also effective. The Raman spectra assist in identifying effect of crystallinity by providing information about the structural arrangement of binders and pigments. Similar research studies have shown that drying methods, pigments dispersion and processing parameters have a major influence on crystallinity and turn paint coating performance and durability[17]. The crystallinity percentage can be estimated using the intensity ratio of crystalline to total spectral intensity. The formula employed for this calculation is:

$$Crystallin\ ity\% = \frac{I_C}{I_C + I_A} \times 100$$

Where: I_C-intensity of crystalline peak, while IA-intensity of amorphous peaks.

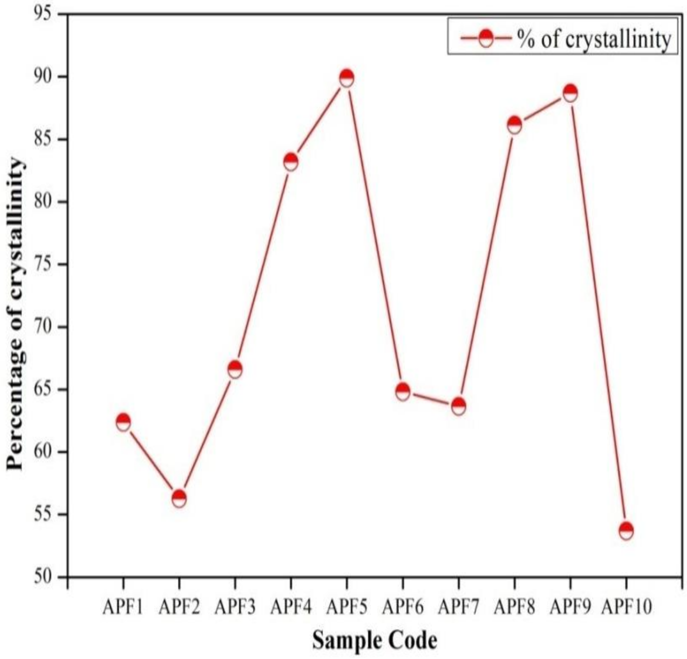

Figure-11: Crystallinity of paint samples.

The % of crystallinity of paint samples was shown in Figure-11. The % of crystallinity of paint sample labeled as APF1 starts at about 63%, followed by decrease in APF2 about 58%. The increase was noticed for paint sample APF3 about 66% and APF4 about 80%, peaking at APF5 about 91%. However, a significant fall is observed in paint sample APF6 about 65% and APF7 is about 64%, before the rises sharply again in APF8 about 88% and APF9 about 89% suggests the ore ordered molecular structure, which can enhance properties like scratch resistance, chemical stability, and adhesion to surfaces[18]. The drastic decline in % of crystallinity was noticed in APF10 is about 55% found to be lowest crystallinity suggesting higher proportion of amorphous regions, which may lead to greater flexibility but reduced hardness and durability[19]. The above rise and decline of % of crystallinity may attribute to the differences in pigment composition, binder properties and solvent interaction. Further analysis using complementary techniques like X-ray diffraction (XRD) and differential scanning calorimetry (DSC) could provide a more comprehensive understanding of the microstructural characteristics and thermal behavior of these paint formulations which may be added in future scope.

Chromaticity analysis of automobile paint samples using Microspectrophotometry: In examination of automobile paint samples, the color characterization is critical aspects of forensic investigation. The chromaticity diagram exhibits a perceptually uniform color space that enables the precise identification and differentiation of pigments. In present analysis, the automobile paint samples labeled from APF-1 to APF-10 were analyzed using a Microspectrophotometer instrument and their respective chromaticity coordinates in the form (u', v') were mapped to assess color composition and their forensic significance.

The chromaticity coordinates (u', v') of the analyzed paint samples are situated in the range of yellow-green spectral region with coordinates approximately ranging from (0.222, 0.530) to (0.312, 0.540). This suggests that the dominant hues are in the yellowish-green to yellow-orange range. The probable existence of pigment in the paint sample was identified based on the chromaticity diagram: i. Chromium-based yellow pigments such as lead chromate, bismuth vanadate are mainly observed in industrial coatings[20]. ii. Organic yellow pigments such as Benzimidazolone/ Isoindolinone Yellow are more likely present in automotive finishes due to their stability and weather resistance[21]. iii. Iron oxide-based pigments usually corresponds to orange and reddish-yellow hues in paints[22-24].

The variation in the coordinates suggest the slight differences in the paint composition. For instance: APF-5 and APF-9 having coordinated (u'=0.308 & 0.312, v'=0.524 & 0.525) indicate exhibit higher u' values, positioning them closer to the yellow-orange region, which suggesting presence of pigments such as iron oxide or lead chromate. APF-6 (0.222, 0.530) and APF-10 (0.244, 0.540) are more greenish-yellow, possibly indicating a stronger presence of bismuth vanadate or chromium-based yellow pigments. APF-3 (0.281, 0.536) and APF-7 (0.264, 0.532) are centrally clustered, likely representing standard yellow formulations used in automobile coatings.

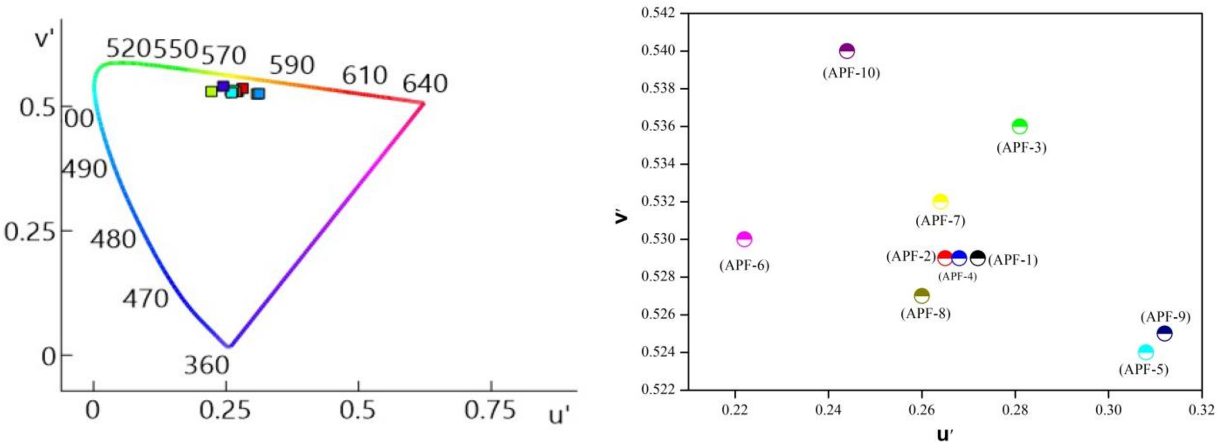

Figure-12: Chromaticity coordinates (u', v') map of paint samples.

Conclusion

In this study, a systematic forensic analysis of Indian automobile paint samples was conducted using surface morphology examination, Raman spectroscopy, and chromaticity analysis. The research aimed to establish a reliable and effective methodology for paint characterization, which is critical in forensic investigations involving hit-and-run cases, vehicular accidents, and criminal activities. The surface morphology analysis performed using Raman microscopy revealed distinct layer structures, confirming the variations in paint compositions among different automobile manufacturers. These findings are crucial in differentiating between paint samples and establishing their origin. The Raman spectroscopy analysis provided valuable insights into the vibrational energy shifts of different pigment components present in the paint samples. The obtained Raman spectra successfully distinguished between organic and inorganic pigments, confirming the presence of characteristic peaks associated with TiO_2, Fe_2O_3, and other commonly used automotive pigments. The best Raman shift values observed were at 514 cm^{-1}, 640 cm^{-1}, and 1440 cm^{-1}, which correspond to key pigment compositions. The chromaticity analysis conducted using a micro spectrophotometer (MSP) mapped the chromaticity coordinates (u', v') of the paint samples. This approach effectively categorized paint colors based on their spectral characteristics, ensuring precise differentiation. The highest accuracy in colorimetric identification was achieved with average CIEUV ΔE value 0.031725 and standard deviation CIEUV ΔE value 0.02166. Furthermore, the recorded Raman spectral database and chromaticity values can significantly enhance forensic databases, facilitating faster and more precise identification of paint samples in forensic casework. Overall, this study successfully validates the use of Raman spectroscopy, surface morphology analysis, and chromaticity mapping as powerful tools in forensic paint analysis. Future research may focus on expanding the dataset by exploring advanced spectroscopic techniques to further refine forensic paint characterization.

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