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Vehicle Paint as Trace Evidence: Spectroscopic and Microscopic Characterization for Forensic Identification

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Abstract

Vehicle paint is a common trace evidence in crime scene investigations of car accidents. The importance of car paints in terms of Forensic Sciences is highlighted by the transfer of paint samples from one car to another in car accidents, theft, or hit-and-run cases. The paint is considered evidence in all cases where vehicle paint is found as residue. Analyses are performed with FT-IR, Raman, Pyr-GC/MS, and SEM/EDX. With the analysis methods, the basic characteristics of the paints on the vehicles involved in the incident can be determined. In this study, paint samples of five vehicles obtained from the auto industry in Çorum were pulverized and analyzed using FT-IR Spectroscopy and SEM/EDX methods. As a result of the analysis, it was observed that vehicles with different brands, models, and production years gave different results. SEM images of five vehicle paints provided information about surface topography. SEM results show that the paint samples have rough, cracked, and heterogeneous surface morphology. The analysis information for each vehicle varies, proving the importance of vehicle trace evidence. The vehicles involved in the crime can be identified even without eyewitnesses or camera footage. There is a need to create a vehicle database in our country. This study aims to contribute to the tool database library that can be used in forensic sciences in our country in the future.

Keywords: Vehicle Paint, Forensic Science, FT-IR, SEM/EDX, Trace Evidence

INTRODUCTION

Paints, or surface coatings, as they are technically called, are ubiquitous in daily life. They are applied to many surfaces, such as wood, paper, metal, and plastic, to increase the properties and durability of products [1].

There are no strict rules for the application of architectural paint, and situations where architectural paint is applied, often encountered in judicial cases, can be done by amateurs. On the other hand, vehicle paint applications are quite technical processes and are usually carried out by professionals [2]. Paint transfer, which is most common in automobile accidents, is a good example of material transfer that occurs when two objects come into contact [3]. In hit-and-run cases, a mutual paint change usually occurs between the offender's vehicle and the victim's vehicle involved in the collision.

Watercolors are a potential source of trace evidence in crime scene investigations. Due to their water-based nature, watercolor stains can easily contaminate the surfaces they come into contact with, and in this respect, they can carry other types of evidence. Watercolors can be essential, especially in detecting microscopic traces such as hair strands, cartilage pieces, and fibers. In addition, it is possible to detect individual-specific biometric traces such as fingerprints, palm prints, or ear prints on the watercolor layer. Such traces can make significant contributions both to the identification of the perpetrator and to establishing his connection with the incident [4].

Paint shavings and paint transfer are two types of evidence frequently used in forensic material analysis. Especially after traffic accidents, paint transfer between colliding vehicles contributes to illuminating the incident by providing information about the contact direction, impact intensity, and contacting surfaces. These analyses are essential in identifying the perpetrator's vehicle or the parties involved in the accident. In incidents such as theft, paint traces on the tools used by the perpetrator at the scene help establish a connection between the suspect and the crime scene. Paint layers transferred from door, window, or vehicle surfaces onto the tool can be analyzed with microscopic and spectroscopic

techniques to reveal unique structural features [5]. For forensic analysis of multilayered paint chips of hit-and-run cars, composition analysis, including trace chemical components in multilayered paint chips, is crucial for identifying the getaway car. Depending on the car manufacturer and year of production, the number of layers, painting process, and paints used are specific to the car types, color, and surface protection. [6].

The most crucial element that makes the paint attractive is its color. Different types and amounts of minerals are used in paint formulas. Pigments are one of the cornerstones of paint, in the form of small particles that are insoluble in a solvent or a binder. Ordinary paint is a mixture of pigment particles, polymers, polyelectrolytes, and surfactants [7]. There are differences in brightness, chemical stability, cost, and durability between organic and inorganic pigments. Organic pigments contain carbon, while inorganic pigments do not contain carbon. Organic pigments such as azo, diazo, polycyclic, phthalocyanine, and quinacridone contain carbon, while inorganic pigments do not contain carbon (i.e., titanium dioxide, iron oxide and chromium oxide are inorganic pigments). [8]. Copper phthalocyanine blue and titanium dioxide are very common pigments used in paints. Most pigments used in paint produce color by selective absorption of specific wavelengths of visible light [9].

Paint chips and stains are left as physical evidence at hit-and-runs, crime scenes, and accidents that can be transferred from one vehicle to another or from one vehicle to the person hit. Before a paint sample is taken, a photograph is taken of the area containing the sample without disturbing the evidence. Crime scene investigators then collect the paint sample by carefully scraping or peeling it from the surface where it was found. Therefore, the investigating officer must be competent and effective in collecting paint evidence and be aware of paint's importance as physical evidence [10].

Paint chips taken during the paint collection process should contain all paint layers, if possible. While the collected physical evidence (glass containing paint samples, headlights, etc.) is sent to the laboratory with the help of plastic bags

or glass bottles, if there are paint chips on the clothes, they are wrapped in clean paper and sent. Dry paint chips brought to the laboratory from the scene are subjected to a series of processes for analysis. The choice of method used depends on the product quantity and condition. The paint chip to be analyzed is evaluated visually and macroscopically. Suppose the sample is a paint chip as a result of macroscopic examination. In that case, the examination is continued with the help of a microscope, and the number and order of layers of the paint sample are determined. If there is a reference paint sample, the comparison is made. If a physical match is made, a report is kept. If there is no physical match, the sample is evaluated for the analytical approach. A sample suitable for the method to be used is prepared. The analyst applies solvent tests to the paint chip to see if the paint reacts by swelling, curling, softening, or other ways.

Significant diversity was found from primary surface coats using FT-IR microspectroscopy to characterize and evaluate the chemical diversity of paint samples from 75 vehicles representing a range of automobile manufacturers. Information such as manufacturing country, manufacturing facility, and vehicle production year were obtained in the classification made using principal component analysis. Obtaining this information from the analysis of interrogated automotive paint samples found at a crime scene or on the victim's clothing has been shown to assist in developing avenues of investigation [11]. The paint sample can be analyzed by taking a thin section with the help of a microtome or by pulverizing it. FT-IR spectroscopy, SEM, PC/GC-MS, and Raman spectroscopy can be used to analyze the paint chip instrumentally. Two analysis methods must give parallel results for the paint sample to be presented to the court as evidence. Since automotive paint parts are essential in forensic science, procedures have been defined to analyze vehicle paint samples. A new methodology is proposed to automate visual analysis using image acquisition. Based on the microscopic image of a paint sample, color and texture information was obtained and compared with paints with the same properties for the paint database. Experiments have shown that the proposed methodology is valid, and the same type of pigment is present both in the paint samples and the database [12].

PDQ (Paint data query) is an international automotive paint database used by forensic scientists at 102 forensic laboratories in 24 countries (including 58 forensic laboratories in 32 states in the United States) to help identify possible suspect vehicles with paint evidence left at crime scenes. The binder and pigment formulations in each layer of the paint create unique combinations that determine the vehicle's likely make, model, and year range. Each paint layer is separated and placed between two diamonds for infrared analysis when analyzing an automotive paint sample. Each component has a characteristic fingerprint known as the infrared spectrum. From there, it includes complete source information for each sample, color descriptions of each paint, the chemical composition of each layer, and layer sequence information, including images of each spectrum [13].

Comparative analysis of paint traces can identify pigment

transfer between vehicles during a crash, objects such as crowbars or screwdrivers used to break windows, cars or safes, or the source of spray paint. This allows for tracking vehicle types in hit-and-run accidents and catching thieves who unknowingly transfer paint from the robbery area to any tools they use [14].

To distinguish different pigments with similar tones, microscopic traces of the samples must be taken with high chemical specificity and analyzed non-destructively and comparatively. Raman microscopy meets both criteria due to its high spectral resolution, spatial resolution in the submicron range available, and no need for sample preparation [14].

Duarte et al.'s research examines studies on paint trace evidence published between 2010 and 2019. Information on paint layers, model validation, chemical characterization, and sample origins is available. In addition to FT-IR and Raman spectroscopies, they mentioned optical coherence tomography as an alternative. They also emphasized that chemometric techniques were used when analyzing the spectra [15].

Since auto paints of the same brand and color differ depending on the year of production in terms of chemical content, pigment type, layer structure, elemental content, and resin type, thanks to these differences, a year-based distinction can be made with forensic analysis. Different resin types, binder changes, and solvent bases emerge thanks to the determination of functional groups. Changes in pigment formulations over the years (e.g., a different shade instead of blue or another metal-based pigment for stability) can be distinguished by Raman Spectroscopy. When the elemental composition (e.g., Ti, Zn, Cr) changes yearly, this difference is seen in the EDX analysis [16].

This study aims to emphasize the importance of vehicle paints in terms of forensic science and to characterize paint residues that are considered trace evidence in crime scene investigations. Paint samples taken from vehicles of different brands, models, and production years were examined using FT-IR spectroscopy and SEM/EDX analysis methods, and the chemical and morphological properties of the paints were determined. It has been shown that even paints with similar colors can show differences. The study aims to contribute to future forensic practices to draw attention to the necessity of creating a reference database of vehicle paints in Türkiye.

MATERIAL AND METHODS

Reagents

Vehicle paint samples were obtained from the Çorum automobile industry zone. Among these samples, those suitable for examination were selected and examined in a laboratory environment. The samples were pulverized with a mechanical sanding machine and analyzed with FT-IR, and SEM/EDX images were taken. IR spectra were taken with the device in the chemistry department laboratory of Hitit University. SEM/EDX results were obtained through service procurement at Hitit University Scientific Technical and Application Research Center.

Table 1. Characteristics of the paint samples used in the research

Order	Vehicle brand	Production	Color
1	Renault Toros	1997	Blue
2	Volkswagen Polo	2014	Red
3	BMW 3	2016	White
4	Kia Sportage	2017	Gray
5	unknown	unknown	Red

Apparatus and instrumentation

Paint samples of five vehicles obtained from the auto industry zone were pulverized. Powdered samples were studied directly.

FT-IR Spectroscopy

IR spectroscopy was analyzed using a Fourier Transform Infrared Spectrometer (FT-IR) Thermo Scientific Nicolet 6700 device. FT-IR spectroscopy deals with the infrared region of the electromagnetic spectrum. Because each functional group consists of different atoms and bond strengths, the vibrations are specific to the functional group. FT-IR microspectrometers can be routinely applied in forensic samples because they are less costly and easy to use. Most paints exhibit strong absorbance between 1650 cm^{-1} and 1750 cm^{-1} (carbon-oxygen double bond stretching, sometimes a doublet for polyurethanes) and multiple absorbances (carbon-oxygen single bond stretching) between 1000 cm^{-1} and 1300 cm^{-1} . Although paints are quite complex, it is important to visually distinguish between simple acrylic, PVAc, polyurethane, and alkyd paints. Peaks at 815 cm^{-1} and 1550 cm^{-1} indicate the presence of melamine crosslinking [2].

Scanning Electron Microscope (SEM)

SEM analysis was made with Jeol JSM-5600, and EDX analysis was made with the FEI brand Quanta FEG 250 model. While performing EDX analyses, insulating vehicle paints were coated with gold-palladium coating and analyzed. Images with nanometer-scale resolution are produced and aid in interpretation. In the scanning electron microscope, the interaction between the electron beam and the material is used to obtain information from the sample's surface taken from living or non-living materials [17].

The sample is exposed to a high-energy electron beam in SEM, and the sample's morphology, topography, crystallographic information, etc., are determined. The most important problem at this stage is that electrons accumulate on the surface of the material taken, causing image loss, or the electron beam does not reflect sufficiently from the material. This problem is prevented by the coating materials used during sample preparation. Aluminum, gold, silver, palladium, and carbon can be used as coatings. The coating method prevents particularly sensitive materials from being damaged under the current generated by the electron beam.

RESULTS AND DISCUSSION

FT-IR Analyses

By referring to the relative intensity or shapes of certain absorption bands, the studied automobile paint samples were distinguished based on the Infrared spectra. The prominent

peaks in the FT-IR spectrum of car paints, except for a few inorganic pigments, generally originate from the polymer binder. The FT-IR spectra of car paints are given in Figure 1, and the characteristic (diagnostic) bands in the infrared spectrum originating from the main polymer resins generally found in paint binders are given in Table 2. Modern automotive paint coatings commonly use amino resins, acrylic and/or alkyd resins in the top coat, polyurethanes and epoxy resins in the inner layers are used for corrosion protection, and it is well known that they contain crosslinking agents such as styrene [18]. While the paint in the top coat in the metal section consists of a styrene-acrylic-urethane polymer, the plastic section paint is a styrene-acrylic-melamine resin mixture. Therefore, one of the polymers that constitute the binder makes up the difference in chemical composition [18]. In the literature, the N-H Stretching peak observed for Urethane was determined as 3379 cm^{-1} and 3398 cm^{-1} , respectively, it was determined as 3548 cm^{-1} and 3406 cm^{-1} in the 1997 model Toros brand car [18]. The C=O ($1724\text{--}1719\text{ cm}^{-1}$) and C-O (1230 cm^{-1} - 1012 cm^{-1}) stretching peaks in the ester structure observed in almost all of the II, III, IV, and V numbered automobile paints given in Table 1 indicate the presence of acrylic and alkyd resins in the paint samples [18, 19, 20, 21]. The C=C stretching peaks detected as firm peaks at 1640 cm^{-1} and 1520 cm^{-1} in all paint samples (I-V) indicate Styrene and epoxy resins [19, 20, 21]. When the FT-IR spectrum of the unknown brand vehicle is examined, it is seen to have similar values to the Kia brand car. From here, we can guess that it may be a Kia brand or collided with a Kia brand vehicle.

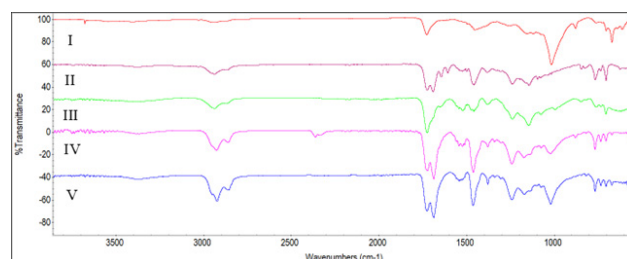


Figure 1. FT-IR spectra of cars. I. Renault Toros brand 1997 model blue colored car; II. Volkswagen Polo brand 2014 model red colored car; III. BMW 3 series brand 2016 model white colored car; IV. Kia Sportage brand 2017 model gray colored car; V. Red colored Unknown brand car

Table 2. FT-IR data of the vehicles (cm^{-1})

Complex	$\nu_{\text{N-H}}$	$\nu_{\text{O-H}}$	$\nu_{\text{C-H}}$	$\nu_{\text{C=O}}$	$\nu_{\text{C=C}}$	$\nu_{\text{C-H bending}}$	$\nu_{\text{C-O}}$
I	3548 3406	3354	2921 2854	1723 1683	1539	1460	1238 1016
II	-	3536	2932 2867	1724	1617	1452	1154 1012
III	-	3393	2936 2859	1722	1520	1456 1374	1236 1142
IV	-	3378	2935 2873	1722 1688	1640 1521	1456	1235 1141
V	-	3378	2924 2857	1719 1684	1521	1458	1238 1169

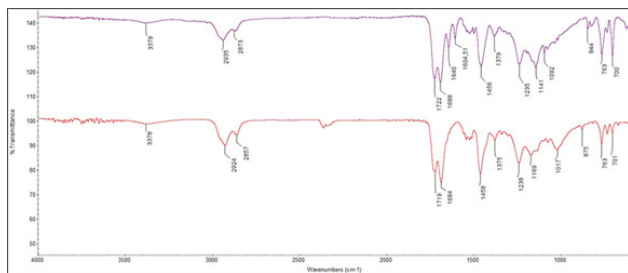
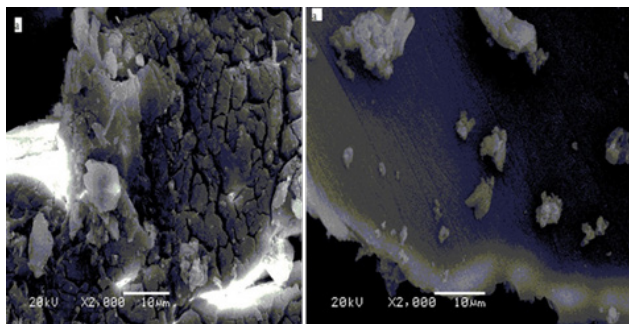


Figure 2. Spectra from top to bottom belong to the purple Volkswagen polo, and the red color belongs to the vehicle of an unknown brand and model.

SEM results



Picture 1. Electron microscope images of the blue paint of the Renault Toros brand, 1997 model, and Volkswagen Polo brand 2014 model red vehicle paint

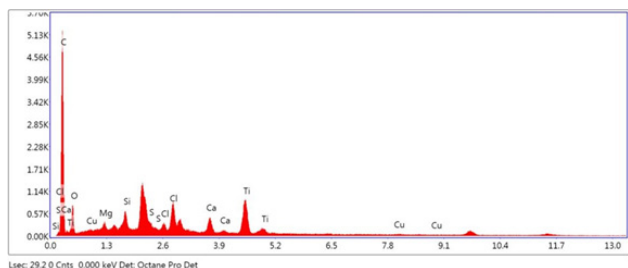


Figure 3. EDX results of Renault Toros brand 1997 model blue paint

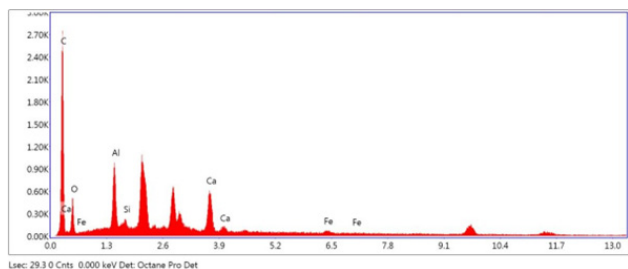
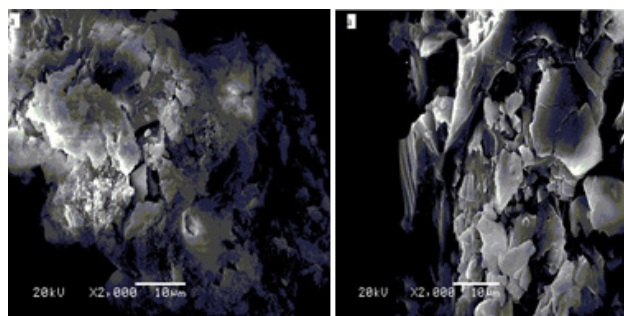


Figure 4. EDX results of Volkswagen Polo brand 2014 model red vehicle paint



Picture 2. Electron microscope images of the white vehicle paint of the BMW 3 Series brand, 2016 model, and the gray paint of the 2017 model Kia Sportage

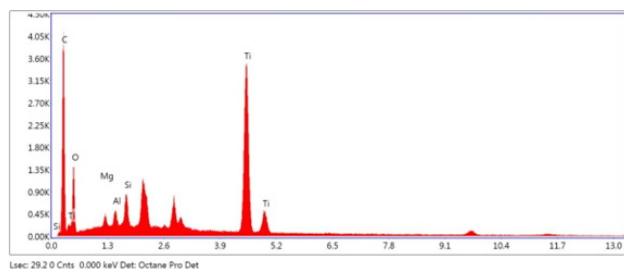
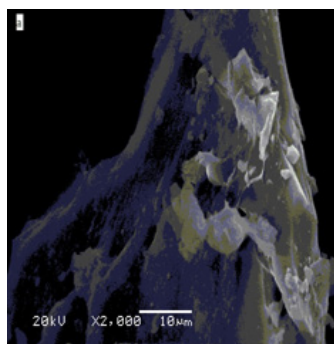


Figure 5. EDX results of BMW 3 Series brand 2016 model white vehicle paint



Picture 3. Electron microscope images of the red vehicle paint hitting a 2017 model gray Kia Sportage vehicle.

When the SEM results (Picture 1, Toros brand vehicle) are evaluated, the cracked and rough morphological structure can be seen in the first picture. The white and shiny appearance may be due to TiO_2 structured pigments. According to the EDX results in Figure 3, the presence of TiO_2 pigments is confirmed. C and O density shows that the paint content is highly organic. Detection of chlorine (Cl) and sulfur (S) elements may result from stabilizers, driers or chemical additives added to the paint system. According to the SEM image of the Volkswagen Polo vehicle in Picture 1, the surface has a wavy structure. As

Table 3. Elemental analysis results of Renault Toros, Volkswagen Polo, and BMW 3 Series vehicle paints

Order	Vehicle, model brand	Color	Production	Atomic Percentage of Elements									
				C	O	Al	Mg	Si	Ca	Ti	Fe	Cu	Cl
1	Renault Toros	Mavi	1997	68.23	16.58	-	0.41	1.16	1.88	6.38	-	0.40	0.68
2	Volkswagen Polo	Kırmızı	2014	65.29	22.44	4.76	-	0.35	6.81	-	0.35	-	-
3	BMW 3	Beyaz	2016	45.72	27.20	1.08	1.11	1.84	-	23.05	-	-	-

a result of the damage, a non-homogeneous, multilayered structure is formed. Silica or aluminum-based pigments or additives are available according to EDX (Figure 4). The SEM information of the BMW brand vehicle in Picture 2 shows fragmented, fractured, and porous areas on the surface. This can be seen when the paint peels off from the surface. When the SEM results of the red vehicle of an unknown model and brand that crashed into the Kia Sportage brand vehicle are examined, thin layers can be seen on the edges. Pointy-looking structures are related to brittle and cracked surfaces.

In this study, standard spectra were observed in the paint scales. The band seen at 1730 cm^{-1} is most likely from alkyd resins used in modern paints. Again, the bands observed around 3600 cm^{-1} and 1000 cm^{-1} in all paint samples indicate the presence of talc. Talc is a widely used filler in high-quality paint. Talc-filled compounds are common paint extenders due to their chemical inertness, allowing pigments to adhere to the vehicle surface and increasing durability. Alkyd resin is a widely used binder in the automotive industry. It is a synthetic resin made by a condensation reaction (water release) between a polyhydric alcohol and dibasic acid (phthalic anhydride). The polymer structure of alkyd resins makes them suitable for use as a base for enamels and paints with different specific properties. The structure molecules form strong structural interactions that include forming films that can cover any surface, providing protection against weathering and wear.

In our study, SEM images provided information about the surface topography of five paint samples. The red vehicle of an unknown brand that hit the 2017 model Kia Sportage is not the 2014 model red Volkswagen Polo we analyzed. We found that the FT-IR spectra and SEM images of the two-vehicle paints are different. As a result of the elemental analysis of the paints, the most common elements found in blue Renault Toros, red Volkswagen Polo, and white BMW 3 Series vehicles are C element, O element, and Si element. FT-IR results confirm the talc we found in the white BMW and the blue Renault Toros Mg and Si. The Fe element found in the red-colored Volkswagen indicates the presence of red iron oxide. The Ca element in Volkswagen Polo and Renault Toros vehicle paints may be related to calcium carbonate. Calcium carbonate has a tiny particle size. It is applied to protect the vehicle against impacts such as sand and gravel.

CONCLUSION

In this study, paint samples of five different vehicles were examined. It has been determined that the molecular structures of vehicle paints of different colors, brands, models, and production years are different. It has been determined that the microscopic images and FT-IR bands of two red vehicle paints that appear to be the same color are different. Epoxy and styrene resin were detected in all paint samples. SEM images provided information about the surface topography of the paint samples. Images: It has been shown that rough, cracked, and particle structures are present. According to the elemental analysis results, Ti, Si, Ca, and Al elements were found in the paint samples. Paint samples, one of the trace evidence frequently encountered in forensic cases, can be analyzed and used to create a paint database.

Our study is planned to contribute to this issue.

Kochanowski (2000) analyzed and interpreted 100 vehicle paints in 5 colors with Py-GC-MS [22]. McIntee (2008) analyzed 110 automobile paints with LIBS (laser interactive plasma spectroscopy), FT-IR, and SEM/EDS and distinguished 88% of the samples [23]. Zieba-Palus (2010) examined 18 new and repainted solid and metallic paints [24]. Skenderovska et al. (2008) contributed to the police investigation by examining four different hit-and-run accidents with Raman and FT-IR spectrometers [25]. Gelder et al. (2005) analyzed paint flakes with Raman spectroscopy and evaluated their contributions to forensic science [26].

A study also shows that the new multimodal approach has a strong potential to elucidate the chemical and physical properties of multilayered vehicle paint particles and could be useful in detecting illegal vehicles. In this study, Fourier transforms infrared (ATR-FT-IR) imaging, Raman micro spectrometry (RMS), and scanning electron microscopy/energy dispersive X-ray spectrometric (SEM/EDX) techniques were performed in combination for detailed characterization of three car paint chip samples, providing complementary and comprehensive information about multilayer paint chips [6]. Maric et al. used FT-IR to characterize and evaluate the chemical diversity of electrocoating primer, primer surface coating, and base coats in automotive paint samples from 75 vehicles. Classification using principal component analysis revealed 14 distinct groups that could be correlated to country of manufacture, specific manufacturer and manufacturing facility, year of vehicle production, and in some cases, number of layers in the paint system [11]. Verma et al. (2019), in their study to distinguish automobile paint samples belonging to Maruti company in India, analyzed 20 paints with solubility tests and a UV spectrophotometer. In their study, unlike the literature, they worked on vehicle paints that had not been studied before. They observed the benefits of FT-IR and SEM analyses in hit-and-run situations with a high probability of paint chip change [27].

The paint particles detected in vehicle accidents are compared using global databases such as the number of layers, color pigments, FT-IR/UV-Vis spectra, and PDQ/EPG. With this study, we wanted to draw attention to the necessity of creating a reference database in order to make reliable comparisons in evaluating vehicle paints as trace evidence in forensic cases. Chemical and morphological analyses of paint samples from different vehicles reveal that each vehicle has its unique paint structure, and this brings up the need for a systematic archive to determine which vehicle the paint residues obtained from the scene belong to. Suppose a national vehicle paint database is created. In that case, it will be possible to identify vehicles through paint evidence in cases where there are no eyewitnesses or camera recordings, and it will make significant contributions to criminal investigations.

There is a need to create a vehicle paint database in our country. This study is intended to be a preliminary study for the vehicle paint database library that can be used in forensic sciences in our country in the future and to pioneer and contribute to new studies.

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