

John V. Goodpaster,¹ Ph.D.; Amanda B. Sturdevant,² M.S.; Kristen L. Andrews,³ B.S.; and
Leanora Brun-Conti,¹ B.S.

Identification and Comparison of Electrical Tapes Using Instrumental and Statistical Techniques: I. Microscopic Surface Texture and Elemental Composition*

ABSTRACT: Comparisons of polyvinyl chloride electrical tape typically rely upon evaluating class characteristics such as physical dimensions, surface texture, and chemical composition. Given the various techniques that are available for this purpose, a comprehensive study has been undertaken to establish an optimal analytical scheme for electrical tape comparisons. Of equal importance is the development of a quantitative means for sample discrimination. In this study, 67 rolls of black electrical tape representing 34 different nominal brands were analyzed via scanning electron microscopy and energy dispersive spectroscopy. Differences in surface roughness, calendaring marks, and filler particle size were readily apparent, including between some rolls of the same nominal brand. The relative amounts of magnesium, aluminum, silicon, sulfur, lead, chlorine, antimony, calcium, titanium, and zinc varied greatly between brands and, in some cases, could be linked to the year of manufacture. For the first time, quantitative differentiation of electrical tapes was achieved through multivariate statistical techniques, with 36 classes identified within the sample population. A single-blind study was also completed where questioned tape samples were correctly associated with known exemplars. Finally, two case studies are presented where tape recovered from an improvised explosive device is compared with tape recovered from a suspect.

KEYWORDS: forensic science, explosives, electrical tape, scanning electron microscopy, energy dispersive spectroscopy, agglomerative hierarchical clustering, principal components analysis, discriminant analysis

Polyvinyl chloride (PVC) electrical tape offers a number of potential types of evidence to a criminal investigator. For example, both the tape backing and adhesive can serve as substrates for latent fingerprints, with various methods for their recovery available (1–3). In addition, trace evidence such as human hair can be recovered from the tape adhesive. Hair lodged between two pristine layers of tape can become significant circumstantial evidence if it is compared with a suspect via microscopic and genetic analysis (4).

Of course, the tape itself possesses potentially valuable evidential characteristics. For instance, a physical match between two torn or cut tape ends is considered to be a conclusive association of the two pieces of tape (5). This is of particular importance when associating tape recovered from a crime scene and tape found in a suspect's possession. However, the high plasticity and lack of a fibrous backing in electrical tape can make physical matches difficult. When a physical match cannot be established, the class characteristics of tape can be examined and the possibility of two tape samples sharing a common origin can be estimated.

In order for electrical tape to be probative evidence in this situation, three prerequisites must be satisfied: (1) electrical tape must

have a reasonable frequency of occurrence in matters of legal relevance; (2) significant and well-documented diversity must be present in the tape population; and (3) laboratory methods must exist that can reliably discern this diversity. It has long been clear that these conditions are met. The frequency of occurrence of electrical tape, particularly in explosives investigations, is such that its forensic importance has been well recognized. For example, of the incidents reported to the United States Bomb Data Center from 1/1/2001 to 6/1/2006, electrical tape was specifically recorded as a component in almost 600 of those cases (A. Purpura, Personal Communication, The United States Bomb Data Center, June 1, 2006). In addition, pressure-sensitive tape represents a multi-billion dollar industry in the United States, of which electrical tape is an important part (6). The sheer size of this market has resulted in numerous companies that produce numerous tape brands with differing physical and chemical characteristics. Finally, various methods have been developed and proposed for the differentiation of electrical tape samples, as will be discussed below.

The composition, manufacturing, and distribution of electrical tape have a fundamental effect on its analysis and value as physical evidence. Electrical tape consists of two main layers: a plasticized PVC film backing and an elastomeric adhesive (see Table 1). The backing is composed of *c.* 60% PVC resin and 40% liquid and powder additives that are included to adjust the physical and/or chemical properties of the PVC. The powder additives are used in smaller amounts and include stabilizers to prevent oxidation and/or degradation, fillers to reduce cost, colorants, and flame retardants (7,8). A large quantity of liquid plasticizer, generally aromatic or aliphatic in nature, gives PVC the flexibility and

¹Bureau of Alcohol, Tobacco, Firearms and Explosives (ATF), Forensic Science Laboratory—Washington, Amundson, MD 20705-1250.

²Troop C Crime Lab, Missouri State Highway Patrol, Park Hills, MO.

³Forensic and Investigative Science Program, West Virginia University, Morgantown, WV.

*Portions of this paper were presented at the 2005 AAFS Annual Meeting in New Orleans, LA.

Received 17 June 2006; and in revised form 28 Oct. 2006 accepted 5 Nov. 2006; published 5 April 2007.

TABLE 1—*Examples of electrical tape components.*

Polyvinyl Chloride (PVC) Backing		Adhesive	
Component	Examples	Component	Examples
Aromatic plasticizers	Dialkyl phthalate esters Trialkyl trimellitate esters	Elastomers	Polyisoprene (PIR) Polybutadiene (PBR) Polyisobutylene (PIB) Poly(styrene-co-isoprene) (SIR) Poly(styrene-co-butadiene) (SBR) Polybutylacrylate
Aliphatic plasticizers	Dialkyl adipate esters Tricresyl phosphate		
Stabilizers	Lead carbonate (PbCO_3) Lead sulfate (PbSO_4)		
Flame retardants	Antimony oxide (Sb_2O_3) Aluminum hydroxide ($\text{Al}(\text{OH})_3$)	Fillers	Carbon black
Fillers	Carbon black Calcium carbonate (CaCO_3) titanium dioxide (TiO_2) Barium sulfate (BaSO_4) kaolin ($\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$) talc ($\text{Mg}_3\text{Si}_4\text{O}_{10}(\text{OH})_2$) dolomite ($\text{CaMg}(\text{CO}_3)_2$)	Tackifier Resins	Wood rosin terpene resins Petroleum resins

workability common to electrical tape. The main components of the adhesive layer are an elastomer, tackifier resins, and plasticizer that has “migrated” from the backing layer. An elastomer is a natural and/or synthetic rubber polymer which, at room temperature, is capable of recovering its size and shape after removal of a deforming force. Tackifier resins improve the stickiness, or tack, of the polymer by lowering its viscosity and allowing it to form an immediate bond with a surface under low pressure. As in the backing, there are various other additives that lend certain chemical and physical characteristics to the adhesive layer (7,8).

The first process in the manufacture of electrical tape is mixing PVC resin with the appropriate liquid and powder additives and pressing (or “calendering”) the plasticized PVC into a thin film. The remainder of the manufacturing process involves the roll coating of various layers (primer, release, and adhesive) onto the PVC film; see the discussion by Kee (8) for additional details. It is important to note that throughout the manufacturing process, each piece of machinery that comes into contact with the tape has the potential to leave visible and reproducible markings on the final product. For example, marks such as striations, divots, craters, and bubbles can arise from rollers, cutters, the coating application, and the winding of the tape. These markings can provide fast eliminations based on microscopic backing texture (8,9).

Following the manufacturing process, the means by which electrical tape is distributed can be fairly complex. Electrical tape is commercially available in three different grades, depending on the quality and/or quantity of raw materials used in the manufacturing process. These grades can be designated as general, mid-range, and premium. Each grade of tape meets a certain set of standards that affects its suitability for use under various conditions. 3M is the market leader in pressure-sensitive tapes (6) and the specifications for its electrical tape product line are shown in Table 2.

Despite the plethora of grades and nominal brands that are available at the retail and wholesale level, the number of manufacturers who actually produce tape from its raw ingredients is rather small. These producers may sell their products directly to consumers under their own brand names. However, they may also enter into agreements with tape converters who purchase large rolls of tape from the manufacturer, slit it into the desired roll size,

and package the tape under a different brand name. Such “second-tier” manufacturers are not uncommon among mass produced products of all types.

Understanding and documenting this phenomenon is particularly crucial for a forensic population study of electrical tape. If such product distribution is ignored, the presumed heterogeneity of the population (as measured by the number of different nominal brands included in the study) is likely to be inflated. This would arise from nominally different brands that are found to be chemically indistinguishable, not because of a failure of the methodology employed, but because both brands originated from the same primary manufacturer.

While information from market research firms (6) and trade groups (10) is available, such distribution networks are often not readily apparent. Luckily, there are some means by which the origin of a particular brand can be elucidated. In this way, the degree to which a reference collection represents the population of electrical tape can be more accurately assessed. The primary means of product tracing stems from the fact that electrical tapes sold in the United States must be rated by a recognized testing laboratory. Most commonly, Underwriter’s Laboratories (UL) tests electrical tapes to ensure that the tape will adequately perform both as a tape and as an electrical insulator. In addition to UL, the Canadian Standards Association (CSA) also rates electrical tapes for sale in either Canada or the United States. Both UL and CSA measure and certify various adhesion, temperature, flame resistance, and dielectric properties of electrical tapes. Upon successful completion of these tests, a tape is given a UL and/or CSA listing and

TABLE 2—*Grades, product examples and specifications for 3M electrical tape.*

	General Use	Mid-Range	Premium
Products	Tartan 1710 Temflex 1700	Commercial 700	Super 88 Super 33+
Application temperature	0 to 80°C	– 10°C to 90°C	– 18°C to 105°C
Application lifetime	Temporary	Permanent	Permanent
Flexibility	Low	Good	Superior
Initial tack	Good	High	Excellent

allowed to display the applicable logo and identifying file numbers on its packaging. The alphanumeric UL control number, file number, and product number (e.g., 362K, E52811, and FR 101, respectively) can be associated with a particular manufacturer, as can the CSA file number (e.g., LR32044). These file numbers are accessible via the UL (www.ul.com), and CSA (www.csa-international.org) web sites. Generally, a one-to-one relationship can be assumed between a UL/CSA listing and a particular manufacturer. However, it is possible for a tape converter to apply to UL to be “co-listed” and therefore obtain their own UL file number for use on their products, despite the fact that the tape was manufactured by another company. These types of relationships can usually be deciphered, however, by cross-referencing information from UL and CSA and/or contacting the tape supplier directly.

Numerous analytical techniques are in regular use for the analysis of electrical tape, generally focusing on analysis of the tape backing, adhesive, and plasticizers (J. Smith, Personal Communication, Missouri State Highway Patrol, September 24, 2001). However, there have been only a few published studies regarding their relative effectiveness (11). Kee analyzed over 100 samples of PVC adhesive tape (8) based on physical properties such as tape dimensions, surface textures and edge markings, as well as the inorganic and organic content of the tape backing using X-ray fluorescence (XRF) and multiple internal reflectance-infrared spectroscopy (MIR-IR). Samples were separated into four broad classes based on the presence or absence of calcium (a common filler component) and lead (a common stabilizer component). These classes were further divided based on the presence or relative amounts of other elements such as phosphorous, antimony, silicon, sulfur, and titanium. Overall, 131 tapes were segregated into 15 groups based solely on elemental composition. Using MIR-IR and physical characteristics, two of these groups could be further divided, resulting in a total of 17 distinguishable classes.

Keto analyzed 18 rolls of black PVC electrical tape, three rolls from each of six nominal brands: LePage, Tuck, Manco, Nashua A-7, Vanguard and 3M Scotch 33 (9). All brands were differentiable using microscopic examination of surface texture, attenuated total reflectance-fourier transform IR (ATR-FTIR) or XRF. Furthermore, it was observed that within-roll variations were smaller than between-roll variations for a given nominal brand. In addition, 3M tapes were easily differentiable from other brands based on their FTIR spectra. From FTIR analysis of the adhesive side, the rubber component was identified most often as polyisoprene (PIR), polybutadiene (PBR), styrene/isoprene copolymer (SIR), or styrene/butadiene copolymer (SBR). In addition, the plasticizer oil was extracted using a nonpolar solvent such as chloroform or pentane, analyzed as a cast film, and identified as aromatic or aliphatic. Merrill and Bartick (12) have since evaluated six configurations of ATR-FTIR and analyzed the backings and adhesives of various pressure-sensitive tapes, including six different electrical tapes. Overall, a single-reflection diamond/KRS-5 IRE was recommended as the technique of choice.

Williams and Munson (13) analyzed 30 black PVC tapes with pyrolysis-gas chromatography-mass spectrometry (Py-GC-MS). Twenty-six of the tapes were distinguishable based on their pyrograms. One indistinguishable pair was found to be from the same manufacturer with the only differences being in the amount of adhesive applied and the thickness of the backing material used. The tapes in the other indistinguishable pair had different brand names. Although slight differences did exist within a single roll, the samples taken from the same roll were still more similar to each other than to any others in the sample set. All samples re-

covered and tested postblast from an improvised explosive device (IED) could be linked back to the appropriate tape, with only small differences noted.

Based on these prior studies, one may conclude that electrical tape is highly diverse and differentiable. Therefore, the probability of a coincidental association between a known and unknown exhibit should be low. The burden, however, is on the forensic scientist to evaluate this risk by demonstrating the diversity of the sample population. Given the various analytical techniques that are available for this purpose, a comprehensive study has been undertaken to establish an optimal instrumental methodology for electrical tape comparisons. Of equal importance is the development of a quantitative means for sample discrimination. In the case of PVC electrical tapes, a successful analytical scheme will discern differences between samples at a relatively small scale, whether that is between tape manufacturers, nominal tape brands, manufacturing batches of the same nominal brand, or individual tape rolls. Ironically, an overly precise analytical method that could discern differences between samples from the same roll of tape (intrasample microheterogeneity) would not be desirable. Such a level of analytical precision surpasses the relevant sample sizes encountered in crime laboratories and would make associations between a questioned and known length of tape difficult or even impossible. The ability to compare samples in a quantitative manner is also desirable, as statistical tests could then be used to assess similarity or dissimilarity.

Materials and Methods

Bureau of Alcohol, Tobacco, Firearms and Explosives (ATF) Laboratory Electrical Tape Reference Collection

The ATF Laboratories maintain large collections of materials that are of interest to explosives investigations. These collections include batteries, timers, switches, fuse and detonating cord, device containers such as steel and PVC pipe, commercial and military explosives, adhesives and tapes (e.g., duct, Teflon[®] [Du Pont, Wilmington, DE], and electrical). The ATF electrical tape reference collection currently includes over seventy rolls of tape representing over 30 nominal brands, including those exemplars previously analyzed by Keto (9).

Table 3 lists the tape samples that were analyzed along with the manufacturing information that was present on the product packaging. The state from and year in which the exemplar was acquired are listed in the “SOURCE” column. For some 3M (St. Paul, MN) products (e.g., Temflex 1700, Scotch Super 88, Scotch 700, and Scotch Super 33+), the year of manufacture is reported as it is printed on the tape roll core. Among the rolls analyzed are nine exemplars from different production batches that were received from the 3M Hutchinson plant in 2004. These consisted of three rolls each of three different brands (rolls A, B, and C for Super 33+, Super 88, and Temflex 1700). Overall, 34 nominal brands are represented (67 rolls in total). By tracing manufacturing information as described above, at least seven manufacturers can be inferred: 3M, AChem Technology (Taipei, Taiwan, and Shanghai, China), Globe Industries (Taipei, Taiwan), Hebei Huaxia Enterprise (Hebei, China), Ningbo Sincere Adhesive Products (Zhejiang, China), Nitto Denko (Osaka, Japan), and Symbio (Taoyuan, Taiwan).

Although the reference collection includes tape with various colors and widths, only black tapes with a nominal width of 0.75 in. were included in this study. These samples are visibly indistinguishable upon coarse examination and are the most

TABLE 3—*Electrical tape exemplars used in this study (67 rolls total).*

Brand	Roll	Source	Origin	UL	CSA	Listed Manufacturer
ACE Hardware	—	GA (05)	Taiwan	362K E49341 #33180	32044	ACHEM Technology
All-Temp	—	VA (04)	Taiwan	362K E52811 FR 101	32044	ACHEM Technology
Auto Solutions	—	DE (05)	China	74HK E206648	202432	ACHEM Technology
Bengal	A, B, C	—	Taiwan	327K #220	—	Nitto Denko Corporation
Champion Quality Tools	A, B C, D	PA (04) PA (05)	China	57RJ E220094 #1920	—	Ningbo Sincere Adhesive Products
Duck	A	PA (04)	Taiwan	362K E49341	32044	ACHEM Technology
	B	MD (05)	China	74HK E49341	202432	
Duck (668 Pro)	—	MD (05)	Taiwan	362K E49341	32044	ACHEM Technology
Electro Tuff	—	PA (04)	Taiwan	362K E52811 #101	—	ACHEM Technology
Frost King (ET60)	A	—	Taiwan	206T	—	Globe Industries
	B	MD (01)				
Frost King (ET60FR)	C, D	MD (05)	China	57RJ	—	Ningbo Sincere Adhesive Products
	E	MD (05)	China	906B	—	—
GE	—	GA (04)	Taiwan	206T E62265 #230	—	Globe Industries
Globe	—	—	Taiwan	206T E62265 #220	—	Globe Industries
Intertape	—	PA (04)	Taiwan	362K E52811 #101	—	ACHEM Technology
Permacel (P-29 Plus)	—	—	—	705B	—	—
Permacel (AW 8 1/2)	—	—	—	705B	—	—
Power First	A, B, C	IL (05)	China	98LJ E174965	—	Hebei Huaxia Enterprise
Powerworks	—	PA (04)	Taiwan	590J E50292 #33546	—	Symbio
LePage's	—	MD (84)	—	906B	—	—
Leviton	—	MD (85)	U.S.A.	—	—	—
Manco	—	MD (84)	Taiwan	590J	—	—
Michigan Industrial Tools	—	PA (05)	Taiwan	590J E50292 #33546	—	Symbio
Nashua A-7	—	MD (84)	U.S.A.	116A	—	—
Radio Shack	—	MD (05)	Taiwan	362K E52811 #101	32044	ACHEM Technology
Radio Shack (Temflex 1700)	—	MD (05)	U.S.A.	539H	—	3M
Scotch 33	A, B, C, D	MD (84)	U.S.A.	539H	—	3M
Scotch Super 33+	A, B C D, E, F G A2 B2	MN (04) MN (03) IL (05) MN (05) MD (01) (01)	U.S.A.	539H	48769	3M
3M Cold Weather (Scotch Super 33+)	CW	MD (05)	U.S.A.	539H	48769	3M
Scotch Super 88	A, B, C A2 B2	MN (03) MD (01) (00)	U.S.A.	539H	48769	3M
Scotch 700	A, B C D	— MD (04) MD (05)	U.S.A.	539H	48769	3M
Shurtape	—	—	—	859X	—	—
Tartan 1710	A, B	—	Taiwan	9Z53	702174	3M
Temflex 1700	A, B C	MN (04) MN (03)	U.S.A.	539H	48769	3M
Vanguard	A, B	MD (84)	U.S.A.	521D	—	—
WUL	—	MD (84)	Taiwan	590J #33546	—	Symbio

commonly encountered color and width. The vast majority of these tapes were nominally 7 Mils (0.007 in.) in thickness; the two known exceptions were Scotch Super 88 and Permacel (New Brunswick, NJ) AW 8 1/2, which were nominally 8.5 Mils (0.0085 in.) thick. While this difference in thickness can serve as a nonchemical means of discrimination, practical experience shows that the high plasticity of PVC tape makes accurate thickness measurements rather difficult, particularly when comparing the thickness of pristine tapes with those that have been stretched or damaged in any way.

Instrumental Analysis

All samples were handled with gloves to avoid any contamination of the backing. In addition, a length of tape encompassing the circumference of the roll was removed and discarded before sampling to ensure a clean surface for analysis. All tape samples were also immediately categorized according to the color of their adhesive (i.e., black or clear). It is known that 3M is the only manufacturer that uses black filler in its adhesive layer; therefore, a careful visual examination of the adhesive color can rapidly discriminate 3M tapes from other brands. Each tape sample was cut off the roll in an *c.* 5 cm strip and placed directly onto a round 75 mm diameter aluminum disk. Three separate locations centered along the width and length of the tape segment were analyzed. In preliminary studies, acquiring elemental data within 2 mm of the edge of the tape resulted in artificially high levels of aluminum due to backscatter from the aluminum stub.

The scanning electron microscopy and energy dispersive spectroscopy (SEM-EDS) instrumentation consisted of a JEOL JSM-5910LV electron microscope with a tungsten source (JEOL, Peabody, MA). An accelerating voltage of 30 kV was used and the working distance was set at 15 mm. The electron beam spot size was varied in order to optimize dead time, but typically was set to a value of *c.* 40. The magnification was set at $65\times$ and the electron image resolution was 512×400 . Given the insulating characteristics of electrical tape, all samples were imaged and analyzed under low vacuum conditions with a sample chamber pressure of 20 Pa. The energy dispersive spectrometer utilized an EDAX SUTW detector with EDAX Phoenix analyzer and EDAX Genesis software (EDAX, Mahwah, NJ). The acquisition time was set at 300 live seconds with a typical dead time of *c.* 30%. The detector resolution was calibrated monthly and resolution typically ranged from 130–135 eV.

The relatively low magnification and long working distance described above were intended to maximize the sample size that was examined by the SEM (*c.* 1.6 mm $\times$ 2 mm). In turn, this large sample size helped to avoid any microheterogeneity that may be present in the tape backing due to poor mixing of inorganic fillers. The relatively long acquisition time and high accelerating voltage were designed to enhance peak-to-background ratios, particularly for heavier elements such as Pb and Sb, whose detection by EDS is more challenging than by XRF. In general, the reproducibility of the conditions and resultant data are of paramount importance, particularly when comparing a known and unknown sample.

Selected tape samples were analyzed via X-ray powder diffraction (XRPD) to confirm the presence of particular inorganic fillers. The instrumentation utilized was a D8 Discover with a General Area Detector Diffraction System (GADDS) (Bruker AXS, Madison, WI). Samples were irradiated using a Ceramic Long Line Focus Cu anode X-ray tube set at 45 kV and 35 mA using a 0.5 mm pinhole slit. Data processing was conducted using EVA software.

Following the accumulation of a large data set, a single-blind study was conducted whereby small sections of tape were presented as unknowns to a co-author (K. L. A.) and analyzed. Based on surface texture and elemental composition, these samples were associated with their possible source. In this case, a set of 3M tapes with black adhesives was utilized. Additional sets of tape were examined on two different occasions separated by 4 days (Michigan Industrial Tools, Kentwood, MI), 3 weeks (Super 88, roll C), and 6 months (Frost King [Thermwell Products Co., Mahwah, NJ], rolls A and B) as a measure of the temporal stability of the SEM-EDS technique.

Data Analysis

The standardless quantitation algorithm of the EDAX Genesis software was used to generate de-convoluted net peak areas for the following elements of interest: magnesium (Mg), aluminum (Al), silicon (Si), sulfur (S), lead (Pb), chlorine (Cl), antimony (Sb), calcium (Ca), titanium (Ti), and zinc (Zn). Carbon and oxygen were not included in this study as they largely reflect the organic portion of the tape and were found to be less useful in discriminating tape samples. Chlorine was included as a convenient measure of the amount of PVC in a tape sample versus the amount of inorganic fillers.

Generally, the presence or absence of an element was self-evident. However, some interpretation was necessary in cases of low spectral resolution and/or where peak intensity approached the detection limit of the instrumentation. For example, given the resolution of the EDAX detector, the principal emission lines for S ($K_{\alpha} = 2.31$ keV) and Pb ($M_{\alpha} = 2.34$ keV) were not completely resolved from one another, although the presence of Pb was usually verified by observing its higher energy L lines. While the EDAX software was able to provide deconvoluted peak areas for these two elements, these values exhibited relatively poor precision and were found to be highly correlated. Therefore, the peak areas for S and Pb were included as a summed variable when they were present resulting in better precision and no significant loss of information. In contrast, the principal emission lines for Sb ($L_{\alpha} = 3.60$ keV) and Ca ($K_{\alpha} = 3.69$ keV) were sufficiently resolved and were able to be reliably deconvoluted.

A criterion for consideration of elements present in trace quantities was also needed. Hence, the peak-to-background ratios (P/B) were calculated for each element based on their net peak areas (P) and the computed area of the underlying background (B). Based on working expressions for detection limit derived from counting statistics, any peak whose net peak area exceeded a value of $(B)^{1/2}$ was considered to be detected (with a corresponding confidence level of *c.* 85%) (14,15). While this level of confidence is relatively low, the P/B criterion was intended as an initial screen before further examination using the statistical methods outlined below. Depending on what region of the spectrum was considered, this translated into minimum P/B values of 0.3–0.4 for most elements.

The data was normalized by summing the squares of all peak areas for a given sample and then dividing each peak area in that sample by the square root of this sum (16,17). This procedure eliminated any variability in the data due to sample amount and instrument response. Statistical evaluation of the data was completed using Microsoft Excel and a separately available add-in, XLSTAT-Pro Version 7.5 (Addinsoft SARL, Paris, France). Three stages of statistical analysis were utilized: (1) agglomerative hierarchical clustering (AHC), (2) principal components analysis (PCA), and (3) discriminant analysis (DA) (16,17) Similar

pattern recognition approaches have been used in the forensic analysis and classification of polyester fibers (18), sheet and container glasses (19), and photocopy and printer toners (20,21).

AHC is a useful exploratory technique for grouping observations according to their similarity and/or dissimilarity. Both the reproducibility of the data and the extent to which it may be classified is revealed in this way. The AHC methodology used here relied on the Euclidean distances between data points. For simplicity, each data point corresponded to the average of three observations from an individual roll of tape expressed as normalized peak areas. The formation of clusters was based on Ward's aggregation criterion (20,21). A horizontal dendrogram resulted, where each observation begins as a unique group at a Euclidean distance of zero. As the distance increases, observations begin to cluster together and nodes occur between groups of samples until ultimately all data points are joined at a sufficiently large distance. An automatic truncation, based on the histogram of node positions, is then calculated. If interpreted strictly, any nodes that occur at distances less than this point would be considered insignificant and all affected samples would be clustered together as a homogenous group. This assumption should be made with caution, however, as such an automatic truncation may be too conservative (clustering samples that are in fact differentiable) or too liberal (differentiating samples that are actually indistinguishable). AHC is considered an "unsupervised" pattern recognition technique in that the elemental data is analyzed for similarities and dissimilarities without assigning any observations to a particular class, such as a nominal brand of tape. In this way, the intrinsic heterogeneity of the data can be visualized as a dendrogram where each sample is clustered with others of similar composition.

Inspection of the intercorrelations between the elemental variables allows for significant trends in the data to be discerned. For example, variables that are highly correlated (directly proportional) may appear together in a subset of the population. Variables that are negatively correlated (inversely proportional) may be mutually exclusive. PCA accounts for these intercorrelations among the observed variables and represents them as a smaller set of uncorrelated composite variables, or principal components (PC). The relative contributions of the original variables to the calculated principal components can be discerned by examining the squared cosines and/or factor loadings for each variable. PCA is optimized such that the first PC describes the maximum amount of variation in the original data set and each additional PC accounts for a successively smaller portion. Hence, a plot of the first few PCs will often encapsulate the majority of the variation in the data set in only two or three dimensions, such that visual inspection will reveal its underlying structure.

DA is a useful follow-up to PCA as it can utilize a set of uncorrelated PCs to construct canonical discriminant functions or canonical variates (CV). An important advantage of utilizing PCA as a preprocessing step for DA is that the latter PCs will by definition describe only a small portion of the variance in a data set and hence largely consist of noise. If these latter PCs are excluded from subsequent DA processing, some amount of noise is thereby filtered out. For the data described below, the quantity of PCs selected for the DA algorithm was sufficient to describe at least 99% of the variance.

DA is considered a "supervised" pattern recognition technique in that a learning sample containing observations with assigned groups is used to construct the CVs (17). The ability of the CVs to discriminate between these assigned groups is optimized by maximizing between-group variance and minimizing within-group variance. DA may then classify new observations according to the

CVs generated from the learning sample and report the probability that each new observation belongs to one of the established groups. The validity of the model can be verified by checking for any reclassification of the original observations following construction of the CVs. Analyzing a set of test samples with known groups and allowing them to be classified as unknowns by the DA algorithm is also an effective means of verifying the structure of the data. If significant classification errors are seen between two groups, then it may be concluded that they are easily confused using the instrumental methodology in question. Ideally, good agreement should be obtained between the unsupervised (AHC) and supervised (DA) pattern recognition techniques. Specifically, distinct clusters in AHC should be easily differentiable via DA whereas groups that are significantly comingled in an AHC dendrogram will likely be confused in DA.

Results and Discussion

Surface Texture

The manufacturing process for tape involves the mixing of ingredients, formation of a thin film that will become the tape backing, and many interactions of that backing with various rollers and other surfaces. The nature of the materials in the tape backing is such that its macroscopic and microscopic features will be affected by these treatments. Systematic microscopic features that are discernable by SEM include surface roughness, elliptical calendaring marks, and the nature of inorganic fillers (e.g., the distribution of particle sizes and their spatial density).

Following examination of many backing surfaces via SEM, it has been concluded that the surface texture of any given roll of electrical tape is highly reproducible over both short (mm) and long (cm) distances. For example, Fig. 1 contains six SEM images from a single roll of tape (3M Tartan 1710, Roll A) acquired over 2 days. All sample locations were c. 20 mm apart, but locations one to three were separated from locations four to six by at least 20 cm. This particular roll of tape exhibits moderate surface irregularity with no noticeable surface defects. The inorganic filler has a fine, narrowly distributed particle size. Most importantly, these characteristics are consistent among all six locations. Such consistency was regularly observed with all other tape samples over similar spatial scales.

Figure 2 contains six representative SEM images from six different brands of tape to illustrate the degree to which surface texture can differentiate tape samples. Figure 2A and B depict two rolls of Champion Quality Tools ("Champion") brand, purchased at the same retail location in two different years. A clear differentiation can be made between the two Champion rolls based on surface texture. The most obvious differences are a finer defect structure in the newer roll as well as a lack of large, elliptical calendaring marks—indicating that the two rolls encountered decidedly different manufacturing conditions. This also serves as an excellent example of surface morphology being sufficiently specific so as to discern differences between rolls of tape manufactured under the same brand name. Similar differences were noted between otherwise identical rolls of older brands such as Bengal and Scotch 33.

Another example of the ability of surface texture analysis to differentiate two different nominal brands with indistinguishable chemical compositions is shown in Fig. 2C (Duck 668 Pro) and 2D (3M Tartan 1710, roll A). These two brands share the same country of origin (Taiwan) as well as similar elemental compositions. However, the tape backings have differentiable surface

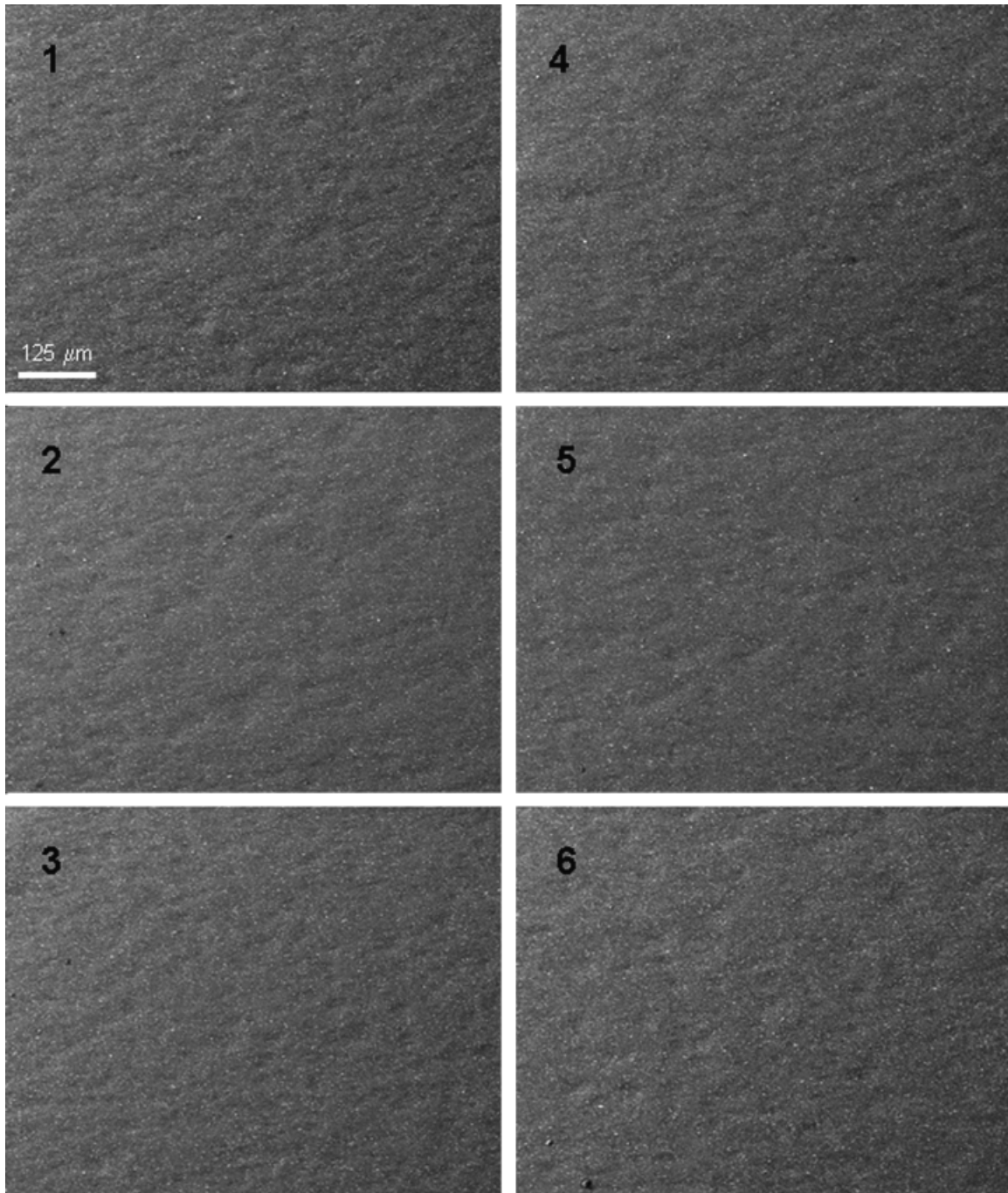


FIG. 1—Six replicate scanning electron microscope images of the surface texture of a single roll of general-use tape (3M Tartan 1710, roll A).

textures based on the smoother surface of the Duck tape as well as its more pronounced calendering marks.

A final example is shown in Fig. 2*E* and *F*, which depict Scotch Super 33+ and Scotch Super 88, two different premium brands with the same listed manufacturer (UL 539 H, 3M Company). The premium-grade tapes manufactured by 3M represent an important exception to the use of surface texture for tape discrimination. This arises from the characteristics of the tapes themselves—exceptionally smooth surface texture, fine, uniform filler particle sizes, and lack of any physical defects. This presents a homogeneous and therefore less specific surface that is largely indistinguishable within and between rolls of the two brands. While the backing thickness of Super 33+ (7Mils) and Super 88 (8.5Mils) is an obvious difference that should not be overlooked, the reli-

ability with which the thickness can be measured decreases rapidly with sample size and elongation.

Elemental Composition

Between-Brand Discrimination (Black Adhesives)—The compiled results for those tapes with black adhesive (and hence manufactured by 3M) are displayed in Table 4. All results are expressed as normalized peak areas and each tape roll was sampled three times. Super 88 roll C (marked with an asterisk) was sampled on two separate occasions separated by 3 weeks' time.

The AHC dendrogram for 3M tapes is shown in Fig. 3, where four clusters are evident: Clusters A and B contain Pb-stabilized

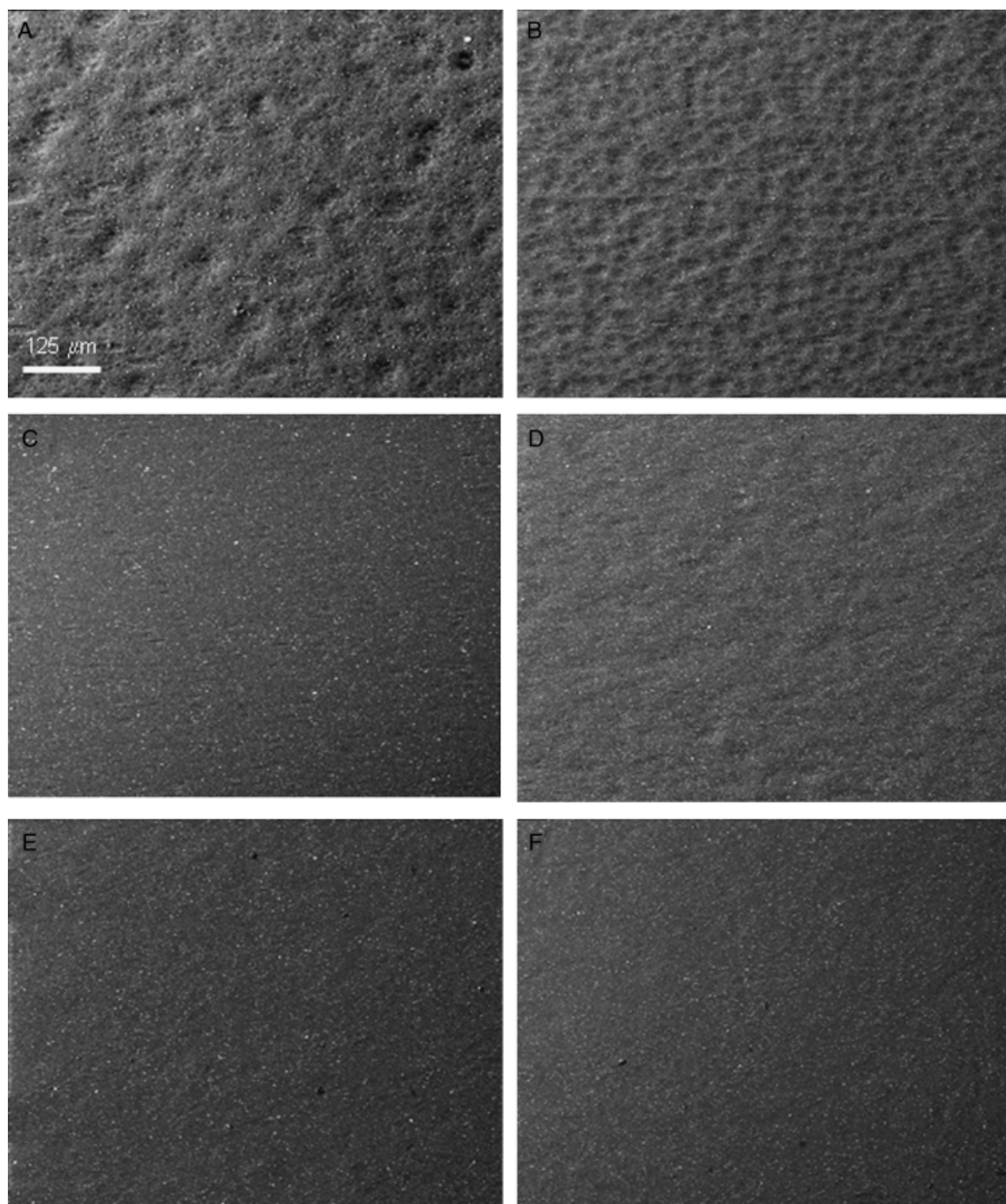


FIG. 2—Comparison of surface textures: (A) and (B). Two rolls of Champion Quality Tools brand (rolls A and C), purchased at the same retail location in different years; (C) and (D) Duck 668 Pro and 3M Tartan 1710 Roll A, two chemically indistinguishable general-use brands manufactured in Taiwan; (E) and (F) Scotch Super 33+ (roll A) and Scotch Super 88 (roll A), two chemically indistinguishable premium brands manufactured by 3M.

tapes whereas Clusters C and D contain Pb-free tapes that have a Mg- and Al-based filler. Cross-referencing this data with sample information in Table 3 indicates a correlation with the year in which the tape was manufactured. In particular, tapes manufactured before 2002 contain Pb, while those manufactured after 2002 do not. This conforms to information obtained from 3M, who discontinued the use of Pb-based stabilizers in their electrical tapes in 2001. All of the 3M tapes tested contained some amount of Sb and Ca, which are attributable to antimony oxide (flame retardant) and calcium carbonate (filler), respectively. XRPD analysis of samples from each of the 3M brands confirmed this

assumption. The AHC dendrogram provides an additional level of differentiation between general-use and mid-grade tapes (Clusters A and D) versus the premium grades (Clusters B and C). This distinction is based on the relative amount of fillers, with the premium grade brands having higher levels of Mg, Al, Sb, and Ca than brands in the mid-range or general grades.

The underlying structure of the data can be further elucidated by examining the correlations between the variables. For example, for the data set that includes all 3M tapes regardless of year of manufacture, the relative amounts of Mg and Al are directly proportional to one another ($R^2 = 0.98$). In contrast, the relative

TABLE 4—Elemental composition of tapes with black adhesive (27 rolls analyzed).

Brand (Rolls)	Year (s)	n		Normalized Peak Areas ($\times 100$)					
				Mg	Al	S+Pb	Cl	Sb	Ca
Commercial 700 (A, B)	Pre-2002	6	Mean	—	—	1.58	99.98	1.11	0.21
			s	—	—	0.09	0.001	0.05	0.03
			% RSD	—	—	6.0	0.001	4.9	11.9
Commercial 700 (C, D)	2004–2005	6	Mean	0.49	0.42	—	99.99	0.99	0.27
			s	0.03	0.04	—	0.001	0.08	0.03
			% RSD	5.4	8.7	—	0.001	7.7	10.1
Scotch 33 (A–D)	1984	12	Mean	—	—	2.02	99.98	0.60	0.33
			s	—	—	0.20	0.004	0.07	0.03
			% RSD	—	—	10.0	0.004	11.2	8.6
Super 33+ (A–G, CW)	2003–2005	24	Mean	0.56	0.62	—	99.99	1.02	0.89
			s	0.04	0.05	—	0.001	0.05	0.08
			% RSD	7.3	8.5	—	0.001	4.4	9.5
Super 33+ (A2, B2)	2001	6	Mean	—	—	2.16	99.97	1.12	0.92
			s	—	—	0.09	0.002	0.03	0.07
			% RSD	—	—	4.1	0.002	3.1	7.7
Super 88 (A–C*)	2003	12	Mean	0.55	0.61	—	99.99	1.11	0.89
			s	0.05	0.06	—	0.001	0.08	0.11
			% RSD	9.6	9.7	—	0.001	7.2	12.2
Super 88 (A2, B2)	2000–2001	6	Mean	—	—	2.26	99.96	1.11	0.90
			s	—	—	0.03	0.001	0.03	0.05
			% RSD	—	—	1.5	0.001	3.0	5.1
Temflex 1700 (A–C, RS)	2003–2005	12	Mean	0.49	0.45	—	100.00	0.45	0.52
			s	0.03	0.04	—	0.001	0.04	0.06
			% RSD	5.8	8.3	—	0.001	9.8	12.1

*Roll C was sampled in triplicate on two separate occasions.
RSD, relative standard deviation.

amounts of these two elements are inversely proportional to S and Pb ($R^2 = -0.95$ to -0.97), which is consistent with the two major sub-groups of these tape samples. Lastly, moderate positive correlations exist between Cl and Mg as well as Cl and Al ($R^2 = 0.72$ – 0.79), and a negative correlation exists between Cl

and S+Pb ($R^2 = -0.88$). These correlations indicate that when they are present, S+Pb occur at higher relative peak areas than do Mg and Al, as can be seen in the original data.

For the purposes of PCA, rolls of tape from the same nominal brand and manufactured in the same time period were qualitatively identified as members of the same group. The result of PCA showed that nearly all (99.0%) of the variance is accounted for in terms of the first three principal components. The squared cosines for PC 1 show that it is strongly linked to the relative amounts of Mg and Al versus S+Pb. Hence, it differentiates between tapes with a Pb-based stabilizer and those with Mg- and Al-based filler, as described above. PC 2, and to a lesser extent PC 3, account for differences in the relative amounts of Sb and Ca.

DA of the first four principle components formed from this data set further refined the modeling of the data. In this case, the vast majority of the variation (95.6%) could be represented in only two dimensions. A plot of the first two CVs appears in Fig. 4 with the scores for the third CV indicated as positive via an unfilled data label or negative via a filled data label. In accordance with AHC and the principle components, the major division in the data arises from older Pb-stabilized tapes, which appear to the left of the y-axis, versus newer Pb-free tapes, which appear to the right of the y-axis. Subtle but discernable differences along CV 2 allow for further differentiation of the premium (Super 88 and Super 33+), mid-range (Commercial 700), and general-use brands (Scotch 33 and Temflex 1700). While the differentiation between brands was generally obvious, there were no reliable differences between Super 88 and Super 33+ other than a small difference in CV 3 for recently manufactured rolls.

The extent to which DA can correctly classify closely related samples in this data set is shown in Table 5. This table lists a set of specific observations, the correct groups to which they were assigned before the calculation of the CV, the groups to which they were reassigned following discriminant analysis, and the probability that the observation belongs to its new, albeit incorrect,

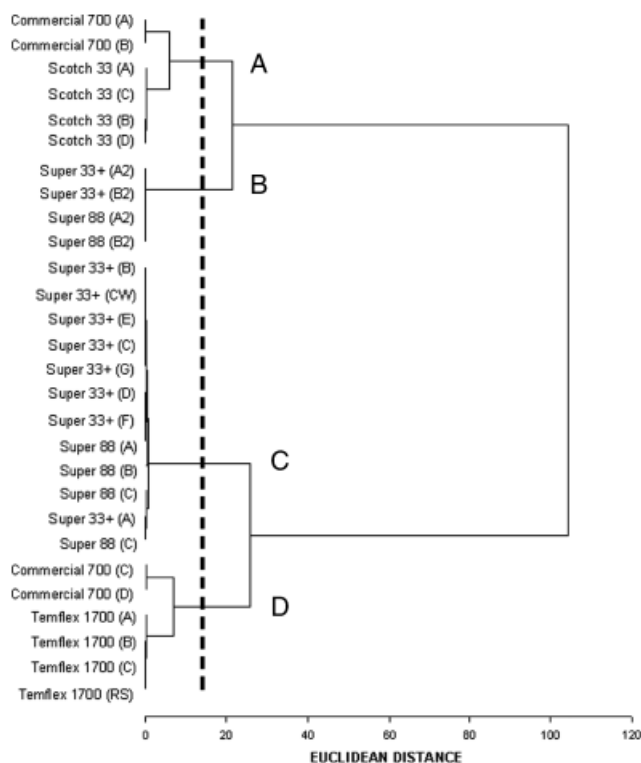


FIG. 3—Agglomerative hierarchical clustering of elemental data for tapes with black adhesive.

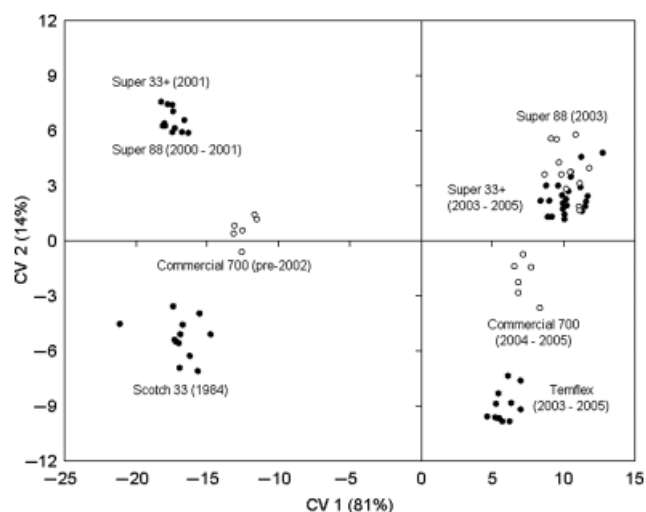


FIG. 4—The observations of tapes with black adhesives expressed in terms of the first two canonical variates (CV). The categories used to group the data were the nominal brand and year of manufacture. Filled data labels correspond to those observations with negative scores in CV 3, while unfilled data labels correspond to positive scores.

class. Notably, all of the incorrect reclassifications involve the brands Super 88 and Super 33+. In particular, Super 88 (roll C) was sampled on two occasions separated by 24 days. While the initial observations were correctly classified, two observations made on the second occasion were incorrectly classified as Super 33+. This is one indication that the temporal variation for the technique can be larger than the brand-to-brand variation that exists between Super 88 and Super 33+. The additional reclassification errors involving these two brands clearly demonstrate that the discrimination of Super 88 and Super 33+ poses the greatest risk of error due to their highly similar chemical compositions. Notably, no reclassification errors were seen with other brands. The apparent error rate was 10.7% for the 84 observations of tapes with black adhesive, or an apparent error rate of 18.8% if only the 48 observations involving the two premium brands are considered.

Between-Brand Discrimination (Clear Adhesives)—The compiled results for tapes with clear adhesive are displayed in Table 6. All results are expressed as normalized peak areas and each tape roll was sampled three times, with three rolls (marked with an asterisk) being sampled on two separate occasions: Frost King (A), Frost King (B), and Michigan Industrial Tools. Before statistical analysis of the elemental data for these tapes, three rolls were excluded based on their unique chemical composition. Frost King (C) and Frost King (D) were the only rolls with clear adhesive that contained Mg in their backing ($P/B = 0.5\text{--}0.7$). Nashua

A-7 brand was the only tape that contained zinc ($P/B = 0.7\text{--}0.8$). Given that a simple visual inspection of the elemental data eliminates these samples as being similar to any other tape, they were not included in further statistical comparisons.

Initial analysis using AHC indicated that a distinct subset of the tapes contains both Al and Si, which is consistent with the use of kaolin or other aluminosilicate-based filler. Hence, given the relatively large number of nominal brands present in this data set (26), the sample population was divided into two sub-classes: those tapes with aluminosilicate-based filler (20 rolls) and those without any detectable Al (20 rolls). AHC of the sample population that contained Al and Si is shown in Fig. 5. In addition to Al and Si, tapes in Cluster A contain S+Pb, Sb and Ca whereas tapes in Cluster B contain only Ca. Cluster C contains the only tapes with detectable levels of Ti, and Cluster D contains tapes with high levels of Ca-based filler. The truncation of this dendrogram is rather conservative, indicating that the tapes may actually be differentiable with additional statistical analysis.

Analysis of the correlation between the variables for aluminosilicate tapes shows that the relative amounts of Al and Si are directly proportional ($R^2 = 0.98$), whereas the relative amounts of these elements are negatively correlated with chlorine ($R^2 = -0.88$). Subsequent PCA results in the first three principal components of the aluminosilicate tapes capturing 79.4% of the total variance of the population. As suggested by the correlations described above, the squared cosines for PC 1 indicate it is heavily influenced by the relative amounts of Al/Si (i.e., kaolin) versus Cl. PC 2 is influenced by the relative amounts of S+Pb and Ca. Finally, PC 3 is heavily influenced by the relative amount of Ti.

The results of a DA using the first five principal components are shown in Fig. 6, with 73.4% of the variance described using the first two CVs. Note that the relatively small portion of the variance depicted in this two-dimensional plot should inspire caution in its interpretation. For example, three brands (Shurtape, Leviton, and All-Temp) are tightly clustered. Despite their proximity in two dimensions, the observations associated with these brands are unambiguously classified by DA with high membership probabilities (All-Temp = 91.8–99.6%, Leviton = 96.4–99.7%, Shurtape = 100%). Similar clusters of other brands were also competently discriminated. In fact, as shown in Table 7, only one observation was erroneous reclassified for an overall apparent error rate of 1.7%. This includes tape rolls that were sampled on two different occasions separated by 6 months (Frost King rolls A and B). Hence, the day-to-day variation for the SEM-EDS measurements is less than the variation seen between these brands and other similar tapes.

AHC analysis of those tape rolls that did not contain Al is shown in Fig. 7. As with Fig. 5, the tapes have been grouped into four clusters based on their fillers. Tapes in Clusters A, B, and C

TABLE 5—Apparent error rate when classifying tapes by brand (black adhesive).

Brand (Roll), Sample	Actual Group	Assigned Group	p
Super 33+ (A), 1	Super 33+ (2003–2005)	Super 88 (2003)	0.79
Super 33+ (A), 3	Super 33+ (2003–2005)	Super 88 (2003)	0.55
Super 33+ (C), 1	Super 33+ (2003–2005)	Super 88 (2003)	0.56
Super 33+ (B2), 2	Super 33+ (2001)	Super 88 (2000–2001)	0.82
Super 88 (A), 1	Super 88 (2003)	Super 33+ (2003–2005)	0.95
Super 88 (A), 3	Super 88 (2003)	Super 33+ (2003–2005)	0.88
Super 88 (C), 5	Super 88 (2003)	Super 33+ (2003–2005)	0.64
Super 88 (C), 6	Super 88 (2003)	Super 33+ (2003–2005)	0.88
Super 88 (A2), 3	Super 88 (2000–2001)	Super 33+ (2001)	0.53

Apparent error rate = $9/84 = 10.7\%$.

TABLE 6—Elemental composition of tapes with clear adhesive (40 rolls analyzed).

			Normalized Peak Areas (×100)								
Brand (Roll)	<i>n</i>		Mg	Al	Si	S+Pb	Cl	Sb	Ca	Ti	Zn
ACE	3	Mean	—	4.40	5.23	1.07	99.76	—	0.43	0.45	—
		<i>s</i>	—	0.03	0.04	0.04	0.001	—	0.04	0.03	—
		% RSD	—	0.7	0.7	3.5	0.001	—	8.5	6.9	—
All-Temp	3	Mean	—	3.13	3.78	1.05	99.84	0.92	2.41	—	—
		<i>s</i>	—	0.05	0.02	0.05	0.001	0.04	0.08	—	—
		% RSD	—	1.7	0.6	4.3	0.001	4.0	3.5	—	—
Auto Solutions	3	Mean	—	—	—	—	98.88	—	14.91	—	—
		<i>s</i>	—	—	—	—	0.007	—	0.05	—	—
		% RSD	—	—	—	—	0.007	—	0.3	—	—
Bengal (A–C)	9	Mean	—	—	—	1.07	99.99	—	—	—	—
		<i>s</i>	—	—	—	0.05	0.001	—	—	—	—
		% RSD	—	—	—	5.1	0.001	—	—	—	—
Champion (A–D)	12	Mean	—	—	—	1.07	99.68	0.99	7.82	—	—
		<i>s</i>	—	—	—	0.04	0.009	0.14	0.12	—	—
		% RSD	—	—	—	3.6	0.009	13.9	1.5	—	—
Duck (A)	3	Mean	—	3.10	3.62	1.06	99.86	—	2.15	—	—
		<i>s</i>	—	0.05	0.09	0.04	0.003	—	0.05	—	—
		% RSD	—	1.7	2.4	4.1	0.003	—	2.2	—	—
Duck (B)	3	Mean	—	—	—	1.11	99.59	—	9.02	—	—
		<i>s</i>	—	—	—	0.04	0.004	—	0.04	—	—
		% RSD	—	—	—	3.4	0.004	—	0.5	—	—
Duck (668 Pro Series)	3	Mean	—	—	—	1.27	99.99	1.03	—	—	—
		<i>s</i>	—	—	—	0.02	0.000	0.03	—	—	—
		% RSD	—	—	—	1.9	0.000	2.6	—	—	—
ElectroTuff	3	Mean	—	3.06	3.42	1.57	99.86	0.37	2.16	—	—
		<i>s</i>	—	0.07	0.07	0.06	0.004	0.06	0.14	—	—
		% RSD	—	2.4	2.0	3.9	0.004	17.2	6.4	—	—
Frost King (A)*	6	Mean	—	4.46	5.35	1.45	99.67	—	3.91	—	—
		<i>s</i>	—	0.08	0.09	0.27	0.011	—	0.09	—	—
		% RSD	—	1.7	1.7	18.4	0.011	—	2.3	—	—
Frost King (B)*	6	Mean	—	4.23	5.03	—	99.78	—	—	—	—
		<i>s</i>	—	0.13	0.10	—	0.010	—	—	—	—
		% RSD	—	3.1	2.0	—	0.010	—	—	—	—
Frost King (C, D)	6	Mean	0.91	0.83	0.81	2.08	84.31	—	53.66	1.84	—
		<i>s</i>	0.11	0.15	0.08	0.12	0.696	—	1.09	0.05	—
		% RSD	11.8	17.9	10.4	5.7	0.825	—	2.0	2.9	—
Frost King (E)	3	Mean	—	—	0.71	—	95.71	—	28.97	—	—
		<i>s</i>	—	—	0.04	—	0.060	—	0.20	—	—
		% RSD	—	—	5.0	—	0.062	—	0.7	—	—
GE	3	Mean	—	4.41	5.31	1.49	99.58	0.39	5.79	—	—
		<i>s</i>	—	0.21	0.05	0.08	0.009	0.05	0.04	—	—
		% RSD	—	4.8	0.9	5.3	0.010	12.3	0.7	—	—
Globe	3	Mean	—	2.93	3.54	1.11	99.89	—	0.23	—	—
		<i>s</i>	—	0.12	0.15	0.08	0.009	—	0.03	—	—
		% RSD	—	4.0	4.1	7.1	0.009	—	15.0	—	—
Intertape	3	Mean	—	3.27	3.82	1.51	99.82	—	2.83	0.55	—
		<i>s</i>	—	0.04	0.04	0.24	0.008	—	0.10	0.01	—
		% RSD	—	1.2	1.0	15.8	0.008	—	3.7	1.5	—
Lepage's	3	Mean	—	5.00	6.54	1.39	99.64	1.17	—	—	—
		<i>s</i>	—	0.05	0.07	0.05	0.003	0.04	—	—	—
		% RSD	—	1.0	1.1	3.6	0.003	3.4	—	—	—
Leviton	3	Mean	—	3.16	3.69	0.99	99.85	0.94	2.15	—	—
		<i>s</i>	—	0.04	0.11	0.08	0.005	0.03	0.08	—	—
		% RSD	—	1.4	2.9	7.6	0.005	3.1	3.7	—	—
Manco	3	Mean	—	3.50	4.07	0.83	99.83	—	2.16	—	—
		<i>s</i>	—	0.03	0.05	0.05	0.003	—	0.10	—	—
		% RSD	—	1.0	1.2	6.3	0.003	—	4.7	—	—
Michigan Industrial Tools*	6	Mean	—	3.54	4.10	—	99.82	—	2.50	—	—
		<i>s</i>	—	0.19	0.21	—	0.019	—	0.21	—	—
		% RSD	—	5.3	5.2	—	0.019	—	8.4	—	—
Nashua A-7	3	Mean	—	—	1.70	2.36	99.93	0.50	2.09	—	0.29
		<i>s</i>	—	—	0.12	0.40	0.009	0.06	0.14	—	0.03
		% RSD	—	—	7.1	16.9	0.009	12.5	6.5	—	11.5
Permacel (P-29 Plus)	3	Mean	—	—	4.39	1.69	99.89	—	—	—	—
		<i>s</i>	—	—	0.13	0.06	0.006	—	—	—	—
		% RSD	—	—	3.1	3.5	0.006	—	—	—	—
Permacel (AW 8 1/2)	3	Mean	—	—	3.60	1.50	99.92	—	—	—	—
		<i>s</i>	—	—	0.08	0.04	0.002	—	—	—	—
		% RSD	—	—	2.2	2.6	0.002	—	—	—	—

TABLE 6—Continued.

			Normalized Peak Areas (× ¹⁰⁰)								
Brand (Roll)	<i>n</i>		Mg	Al	Si	S+Pb	Cl	Sb	Ca	Ti	Zn
Power First (A–C)	9	Mean	—	—	0.48	—	98.75	0.77	15.71	—	—
		<i>s</i>	—	—	0.05	—	0.046	0.06	0.29	—	—
		% RSD	—	—	11.1	—	0.047	8.0	1.9	—	—
Powerworks	3	Mean	—	3.54	3.88	—	99.84	—	2.18	—	—
		<i>s</i>	—	0.07	0.02	—	0.003	—	0.02	—	—
		% RSD	—	1.9	0.4	—	0.003	—	0.9	—	—
Radio Shack	3	Mean	—	3.21	3.75	—	99.85	—	2.43	—	—
		<i>s</i>	—	0.09	0.06	—	0.005	—	0.01	—	—
		% RSD	—	2.9	1.7	—	0.005	—	0.2	—	—
Shurtape	3	Mean	—	3.71	4.18	1.02	99.81	0.99	2.35	—	—
		<i>s</i>	—	0.03	0.07	0.05	0.001	0.04	0.06	—	—
		% RSD	—	0.9	1.7	5.3	0.001	4.2	2.6	—	—
3M Tartan 1710 (A, B)	6	Mean	—	—	—	1.18	99.99	0.98	—	—	—
		<i>s</i>	—	—	—	0.09	0.002	0.09	—	—	—
		% RSD	—	—	—	8.0	0.002	9.0	—	—	—
Vanguard (A, B)	6	Mean	—	—	—	1.08	99.99	1.10	—	—	—
		<i>s</i>	—	—	—	0.04	0.001	0.04	—	—	—
		% RSD	—	—	—	4.0	0.001	3.4	—	—	—
WUL	3	Mean	—	3.32	3.73	1.34	99.84	—	2.09	—	—
		<i>s</i>	—	0.01	0.07	0.01	0.004	—	0.08	—	—
		% RSD	—	0.3	1.8	0.9	0.004	—	3.7	—	—

*These rolls were sampled in triplicate on two separate days.
RSD, relative standard deviation.

all contain S+Pb, whereas tapes in Cluster D do not. Clusters A, B, and C can be differentiated based on their other fillers, specifically Si in Cluster A, Ca in Cluster B, and Sb and Ca in Cluster C. Tapes in Cluster D contain Si, Sb, and high levels of Ca. The truncation of this dendrogram is conservative with multiple nominal brands of similar composition clustered together.

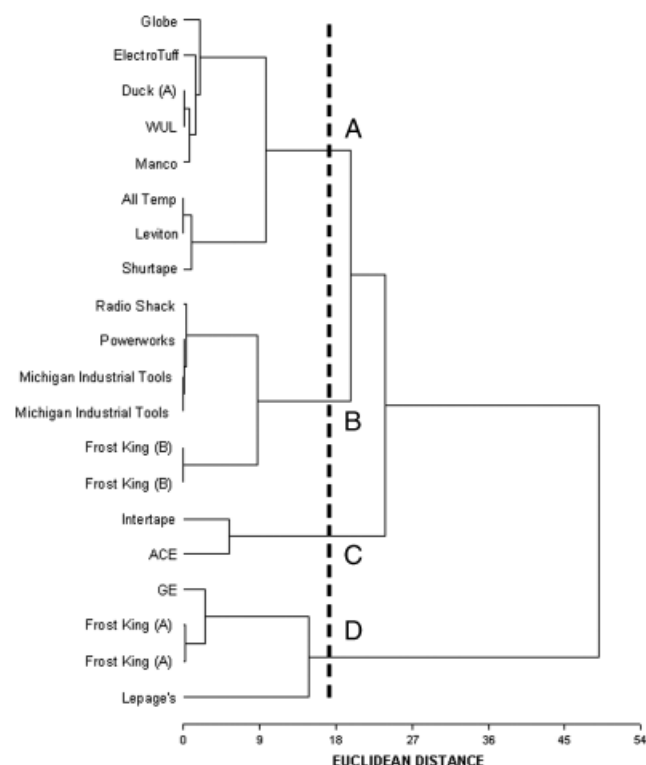


FIG. 5—Agglomerative hierarchical clustering of elemental data for tapes with clear adhesive and aluminosilicate filler.

Analysis of the correlation between the variables for tapes that do not contain Al shows that the relative amount of Ca is negatively correlated with S+Pb ($R^2 = -0.85$) as well as Cl ($R^2 = -0.91$), which implies that tapes with high levels of Ca-based filler tend to be Pb-free and necessarily contain lower relative levels of Cl (PVC). In turn, PCA results in the first three principal components capturing 94.7% of the total variance of the population. As suggested by the correlations described above, the squared cosines for PC 1 indicate it is heavily influenced by the relative amounts of Ca, S+Pb and Cl. PC 2, and to a lesser extent PC 3, are influenced by the relative amounts of Si and Sb.

The results of a DA using these principal components are shown in Fig. 8, with 98.0% of the variance accounted for in

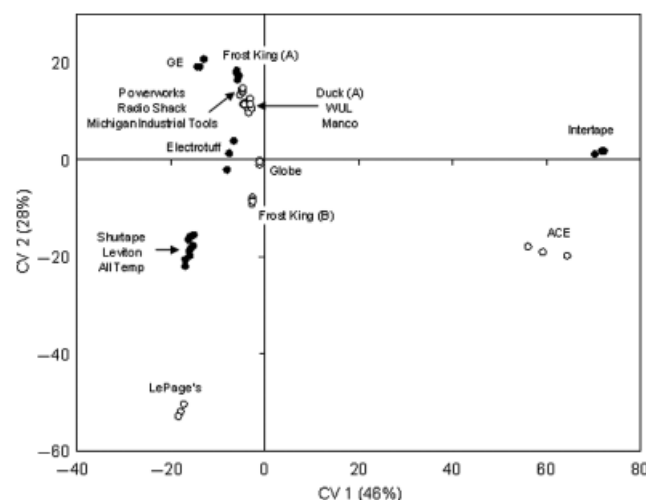


FIG. 6—The observations of tapes with clear adhesives and aluminosilicate filler expressed in terms of the first two canonical variates (CV). The categories used to group the data were the nominal brand with subcategories established for brands in which a significant qualitative difference was observed between rolls. Filled data labels correspond to those observations with negative scores for CV 3, while unfilled data labels correspond to positive scores.

TABLE 7—Apparent error rate when classifying tapes by brand (clear adhesive).

Brand (Roll), Sample	Actual Group	Assigned Group	p
Michigan Industrial Tools, 2 Apparent error rate = 1/60 = 1.7% (clear adhesives with aluminosilicate filler)	Michigan Industrial Tools	Powerworks	0.99
3M Tartan 1710 (A), 1 3M Tartan 1710 (A), 3 Apparent error rate = 2/60 = 3.3% (clear adhesives without aluminum)	3M Tartan 1710 3M Tartan 1710	Duck (668 Pro Series) Duck (668 Pro Series)	0.94 0.88

two dimensions. As was seen with the aluminosilicate tapes, the projection of the observations in this 2D space largely discriminates between most of the tape brands. Consistent with the AHC results above, three brands are tightly clustered: Vanguard, Tartan 1710, and Duck (668 Pro Series). Consequently, the observation membership probabilities for these tapes are lower (Duck 668 Pro = 56–86%, Tartan [roll B] = 89–97%, and Vanguard = 70–100%). In fact, inspection of the reclassification errors for this data set shows that two of the three observations for Tartan 1710 (roll A) were confused with Duck 668 Pro Series (see Table 7). Despite this chemical similarity, it should not be overlooked that the contrasting surface textures of these two samples can serve as a reliable means of differentiation as seen in Fig. 2. In contrast, the tight cluster of observations from Champion and Duck (roll B) do not indicate that they are easily confused. Champion brand contains an Sb-based flame retardant, whereas Duck (B) does not. Hence, there is a large separation between these two brands along CV 3 which is not apparent in two dimensions. Classification by DA was also unambiguous for all observations of these two brands.

Within-Brand Discrimination—Based on the PCA and DA results above, some obvious within-brand heterogeneity has already been noted. In the case of black adhesive tapes, a major formula

change occurred in 2001 and hence tapes with the same nominal brand but different manufacturing dates will have dramatically different compositions. Similarly, some clear adhesive tapes have also undergone changes in composition. For example, the chemical compositions of the two rolls of Duck brand tape are clearly differentiable. Not surprisingly, their manufacturing information and country of origin are also different. Frost King brand is another example, with two clear changes in formulation for each of the two products studied (ET60 and ET60FR). Interestingly, the change in composition for the ET60 rolls (roll A vs. roll B) is not reflected in the manufacturing information, whereas the change in composition for the ET60FR rolls (rolls C/D vs. roll E) is associated with a change in UL Listing.

Previous authors have noted that the variance within any given roll of tape may be less than the variance between rolls of the same brand—leading to the possibility of interroll heterogeneity (9,13). As was observed with surface texture analysis, the elemental composition of any given roll of electrical tape was reproducible over the distances used in this study. While the precision of the EDS data varied with each sample and element, the relative standard deviation (RSD) for any element (excluding Cl) for tapes with black adhesive ranged from 0.4% to 15%. The precision in tapes with clear adhesives ranged from 0.2% to 17% RSD. Therefore, for those rolls with high intraroll reproducibility, some subtle

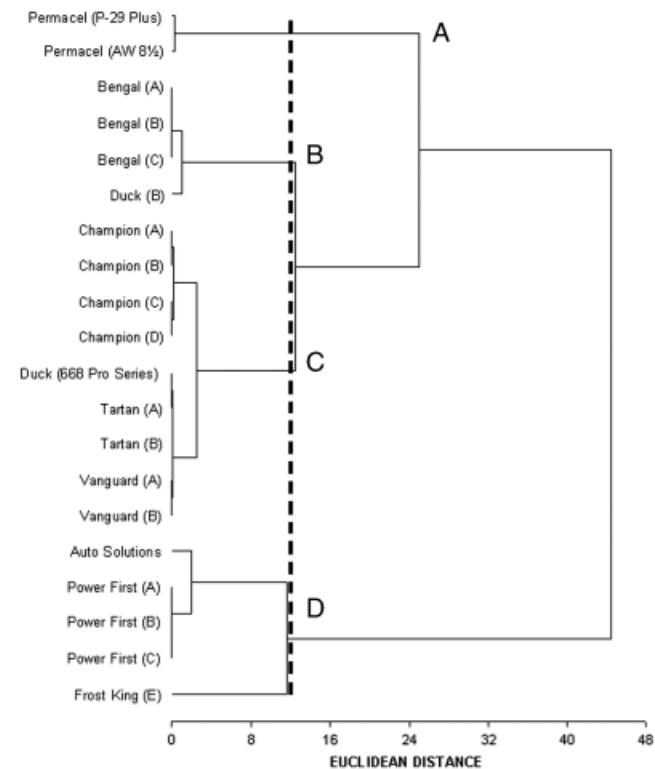


FIG. 7—Agglomerative hierarchical clustering of elemental data for tapes that had clear adhesive but did not contain aluminum.

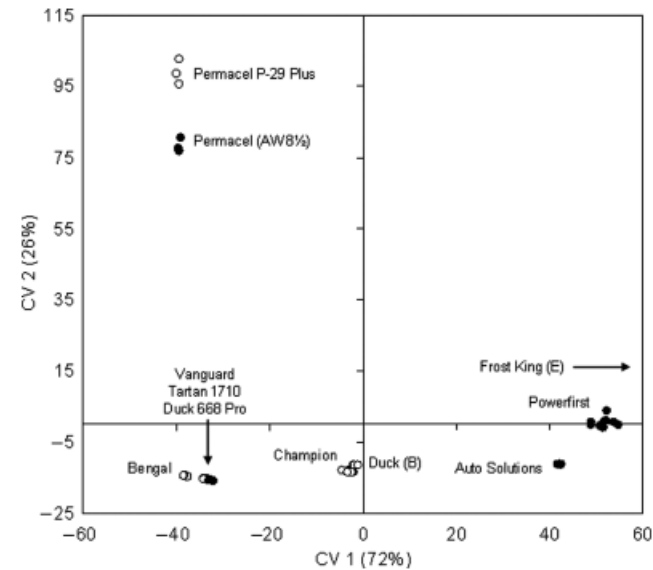


FIG. 8—The observations of tapes that had clear adhesive but did not contain aluminum expressed in terms of the first two canonical variates (CV). The categories used to group the data were the nominal brand with subcategories established for brands in which composition differed qualitatively between rolls. Filled data labels correspond to those observations with negative scores for CV 3, while unfilled data labels correspond to positive scores. Owing to the high level of Ca in Frost King (roll E), its observations appear to the far right of the displayed plot.

TABLE 8—Apparent error rate when classifying tapes by roll.

Brand (Rolls)	n	Reclassified Observations	Apparent Error Rate (%)
Commercial 700 (A, B)	6	0	0
Commercial 700 (C, D)	6	0	0
Scotch 33 (A–D)	12	2	16.7
Super 33+ (A–G, CW)	24	11	45.8
Super 33+ (A2, B2)	6	2	33.3
Super 88 (A–C*)	12	4	33.3
Super 88 (A2, B2)	6	3	50.0
Temflex 1700 (A–C, RS)	12	3	25.0
Apparent error rate = 25/84 = 29.8% (black adhesives)			
Bengal (A–C)	9	5	55.6
Champion (A–D)	12	3	25.0
Powerfirst (A–C)	9	3	33.3
Vanguard (A, B)	6	3	50.0
Apparent error rate = 14/36 = 38.9% (clear adhesives)			

*Roll C was sampled in triplicate on two separate occasions.

differences may exist between rolls of the same brand and formulation.

However, some caution is warranted regarding the tests that can be used to determine if such differences are statistically significant. In particular, the use of traditional univariate statistical tests such as the two sample *t*-test can be problematic if the number of data points is small. For example, at the 95% confidence level and $n = 3$, the true standard deviation can be as high as 6.3 times that of the measured standard deviation (22). As a result, measured precisions may be artificially low and two sample sets may be incorrectly determined to be unequal.

An alternative approach is to use multi-variate techniques to classify data at the level of individual tape rolls. In this way, the hypothesis of roll discrimination was tested for tapes with more than one possibly indistinguishable roll per nominal brand. By using DA with the observations classified according to the roll from which they originated, an apparent error rate could be calculated. Table 8 summarizes the reclassification errors that arose. It is immediately clear that discriminating individual rolls is much more prone to error than classifying samples according to brand. The error rate was highest among the rolls of Super 88 and Super 33+, consistent with the larger number of rolls of these brands and their similar compositions. Error rates for rolls of tape with clear adhesives were similarly high with a significant fraction of the observations being reclassified to another roll of the same brand. While observations from a particular roll were correctly assigned for rolls of the Commercial 700 brand (both new and old), it is likely that the degree of confusion on the part of the DA algorithm, and hence its error rate, would increase as additional exemplar rolls are analyzed.

Single-Blind Study—The best test of a classification scheme is the use of test samples of known origin. Therefore, a single-blind



FIG. 9—An inner tube valve stem with a short length of black electrical tape wrapped around its tip, as recovered postblast.

study was conducted in order to fully validate the discriminant analysis. In this study, seven samples of previously analyzed rolls of tape with black adhesive were presented to a co-author (K. L. A.) without identifying marks. Each sample was analyzed in triplicate and compared with previous data as well as one another. Judgments as to the brand and possible roll were made visually, but statistical analysis was utilized to assess the samples' origin. In this case, the observations for the unknowns were included as supplemental data in the PCA and DA algorithms and not used in the construction of principal components or CV. Therefore, the previously formed principal components and CV were used to calculate the classification of the new observations.

The results of the DA for the unknown samples are shown in Table 9. Classification of the unknowns in terms of their overall brand was generally successful, with all samples except for Unknown G (Super 88, roll A) correctly identified. The fact that a roll of Super 88 would be mistakenly identified as Super 33+ is not surprising given the previous discussion of reclassification errors and the degree to which their chemical composition is indistinguishable. Another key finding is that unknowns A and C, which originated from the same roll of tape, were correctly classified as originating from the same brand.

Attempting to use DA to identify the specific rolls from which the unknowns originated was clearly not as successful, with only three samples (unknowns C, E, and F) correctly identified. Also significant is that a roll of tape sampled on the same day (unknowns A and C), while correctly classified by brand, was incorrectly assigned to two different rolls. This further indicates that the inherent variance of the SEM-EDS method can be larger than that found between rolls of the same brand.

TABLE 9—Brand and roll identification of single-blind samples via discriminant analysis (incorrect assignments are in *italics*).

Unknown	Brand (Roll)	DA Classification			
		Brand	P _{GROUP}	Roll	P _{ROLL}
A	Super 33+ (A)	Super 33+ (2003–2005)	0.99	<i>Super 33+ (CW)</i>	0.50
B	Commercial 700 (B)	Commercial 700 (pre-2002)	1.0	<i>Commercial 700 (A)</i>	0.76
C	Super 33+ (A)	Super 33+ (2003–2005)	0.82	Super 33+ (A)	0.49
D	Super 33+ (B)	Super 33+ (2003–2005)	0.92	<i>Super 88 (A)</i>	0.29
E	Temflex 1700 (B)	Temflex 1700 (2003–2005)	1.0	Temflex 1700 (B)	0.84
F	Scotch 33 (C)	Scotch 33 (1984)	1.0	Scotch 33 (C)	0.54
G	Super 88 (A)	<i>Super 33+ (2003–2005)</i>	0.93	<i>Super 33+ (E)</i>	0.84
Error Rate = 1/7 = 14.3%				Error Rate = 4/7 = 57.1%	

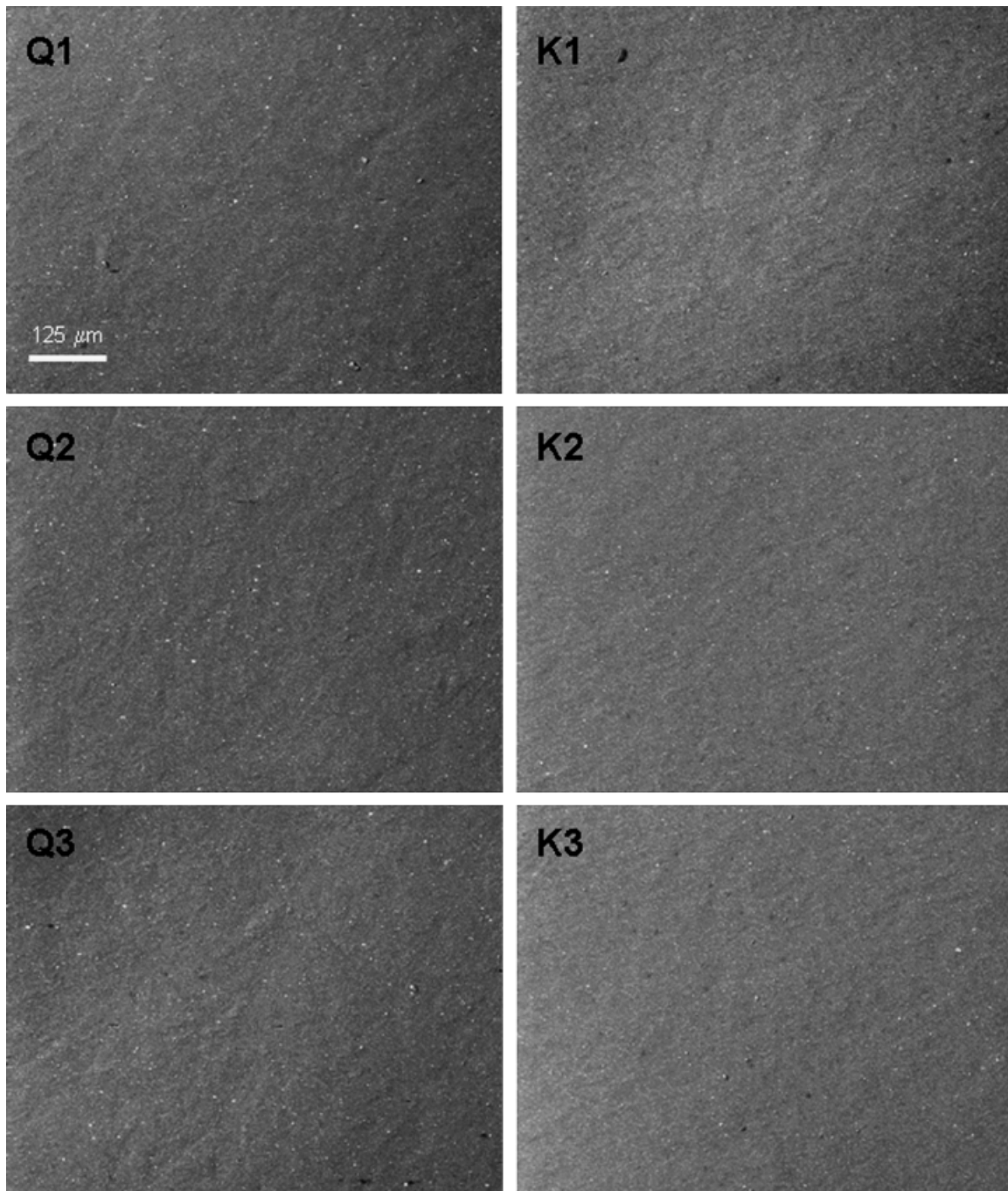


FIG. 10—Scanning electron micrographs of three locations from a questioned sample recovered from the valve stem in Fig. 9 (Q1, Q2, and Q3) and three locations from a tape roll recovered from a suspect (K1, K2, and K3).

Case Study (Exclusion)—Having established the reproducible and reliable differentiation of electrical tapes via surface texture and elemental composition, two case studies will be discussed that demonstrate the ability of these characteristics to either differentiate or associate a questioned and known sample.

The first case involves a large IED composed of an off-road tire inner tube that was fused through its valve stem. This valve stem was recovered postblast, and a small length of electrical tape was observed wrapped around its tip as shown in Fig. 9. While the recovered tape was not amenable to a thickness measurement, a small section of clean tape backing that lay underneath the outer tape layer was recovered and provided a suitable sample for SEM-

EDS analysis. Before analysis, it was noted that the tape had a nominal width of 3/4 in. and the adhesive was black, indicating that the tape was manufactured by 3M. Two exemplar rolls of tape from two separate suspects were submitted for comparison. One roll, identified as 3M Tartan 1710 on its roll core, was easily eliminated as it had clear adhesive. While samples from this roll were analyzed via SEM-EDS for database purposes, it was not compared with the questioned sample. The other roll had black adhesive, was identified as 3M Super 33+ on its roll core, and was marked with a manufacturing year of 2000. A physical/fracture match was not observed between the end of this tape roll and the fragment recovered from the device. However, these two exhibits

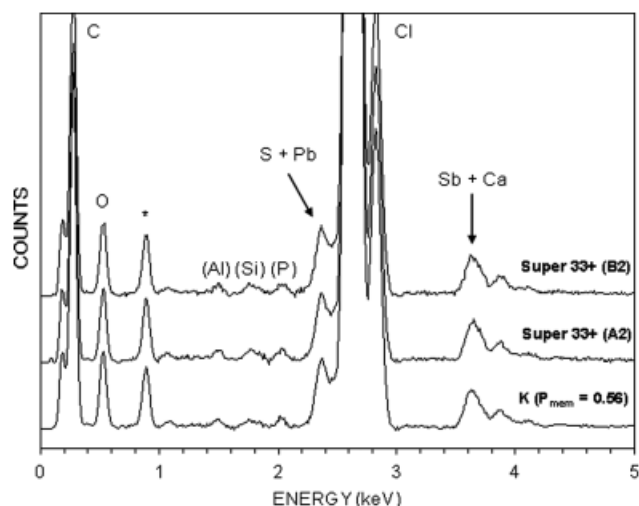


FIG. 11—Energy dispersive spectroscopy spectra of the known tape sample from Fig. 10 (K) as well as two exemplars of the same nominal brand manufactured before 2002 (Super 33+ [A2] and Super 33+ [B2]). The calculated probability that the known tape sample belongs to the same group as the exemplars is 56%. Each spectrum is baseline corrected and represents the average of three locations with trace amounts of Al, Si, and P noted in parentheses. All intensities have been normalized to the principal (K_{α}) emission line of chlorine. The escape peak that results from this emission appears at c.0.9 keV and is noted with an asterisk.

were visually consistent and, therefore, both were analyzed via SEM-EDS.

Figure 10 contains three images of the surface texture for both the questioned (valve stem) and known (Super 33+ [K]) tape samples. As is evident from these micrographs, the two samples had reproducible surface features over the distance sampled (on the order of mm). When examined visually, there was a subtle but distinct difference in the surface finish of the two samples, where the tape recovered from the device was glossier than that from the tape roll. When examined microscopically, the surface textures were quite similar and any clear differences that could lead to a confident exclusion were not present.

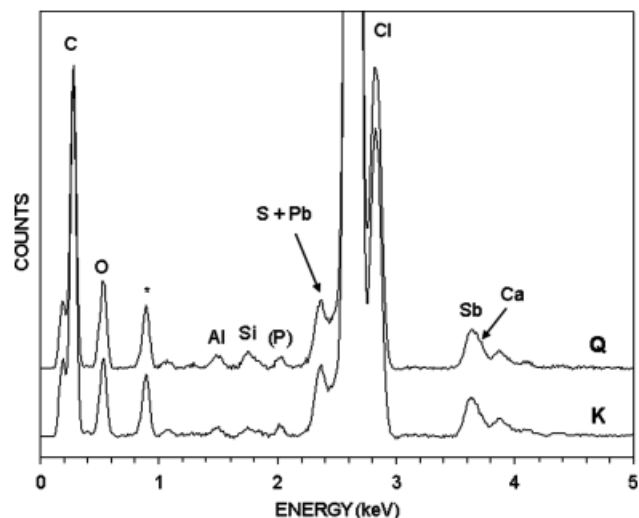


FIG. 12—Energy dispersive spectroscopy spectra of the questioned (Q) and known (K) tape samples from Fig. 10. Each spectrum is baseline corrected and represents the average of three locations. All intensities have been normalized to the principal (K_{α}) emission line of chlorine.

TABLE 10—Comparison of the elemental composition of a questioned tape sample recovered from a valve stem (Fig. 9) and a known tape sample.

Sample	n	Normalized Peak Areas ($\times 100$)				
		Si	S+Pb	Cl	Sb	Ca
Q	Mean	0.46	2.12	99.97	1.08	0.89
	s	0.09	0.05	0.002	0.02	0.02
	% RSD	18.9	2.2	0.002	1.5	2.8
K	Mean	0.22	2.16	99.97	1.16	0.75
	s	0.02	0.03	0.000	0.08	0.04
	% RSD	9.8	1.4	0.000	7.3	5.0
	t-statistic (%)	1.0	35	51	18	0.6

RSD, relative standard deviation.

Before comparing the known and questioned samples based on elemental composition, the known roll served as additional external validation of DA as a means for brand identification. Figure 11 contains the elemental compositions of the known tape sample (K) as well as two exemplars of the same brand (Super 33+ [A2] and Super 33+ [B2]) that were manufactured before 2002. The data is presented as baseline corrected EDS spectra, where each spectrum represents the average of three locations and all intensities have been normalized to the principal (K_{α}) emission line of Cl. The exemplar spectra are highly similar to the known sample, including the presence of weak emission from trace amounts of Al, Si, and P. Note that the P/B ratios for these elements range from 0.1 to 0.2, therefore, they were not included in any subsequent statistical analysis.

DA was utilized to classify the tape from the known roll by constructing a new set of CVs using all exemplar rolls with black adhesive including the single blind samples, now with their cor-



FIG. 13—A propane cylinder with multiple lengths of black electrical tape wrapped around it, as recovered following a render safe procedure.



FIG. 14—Photographic documentation of a physical match between the outermost tape fragment on the propane cylinder and the end of the tape roll.

rect classifications. The replicates for the known roll were included as supplemental data and the centroid of these points was then classified by DA. The observations from this roll were correctly assigned to the group consisting of Scotch Super 33+ tapes manufactured before 2002 (i.e., rolls A2 and B2). The membership probability was modest (56%) with the alternative classification being Super 88 tapes of similar vintage (Super 88 A2 and B2). This result is consistent with the similarity between these two brands and the degree to which these two classes may be confused (see Table 5).

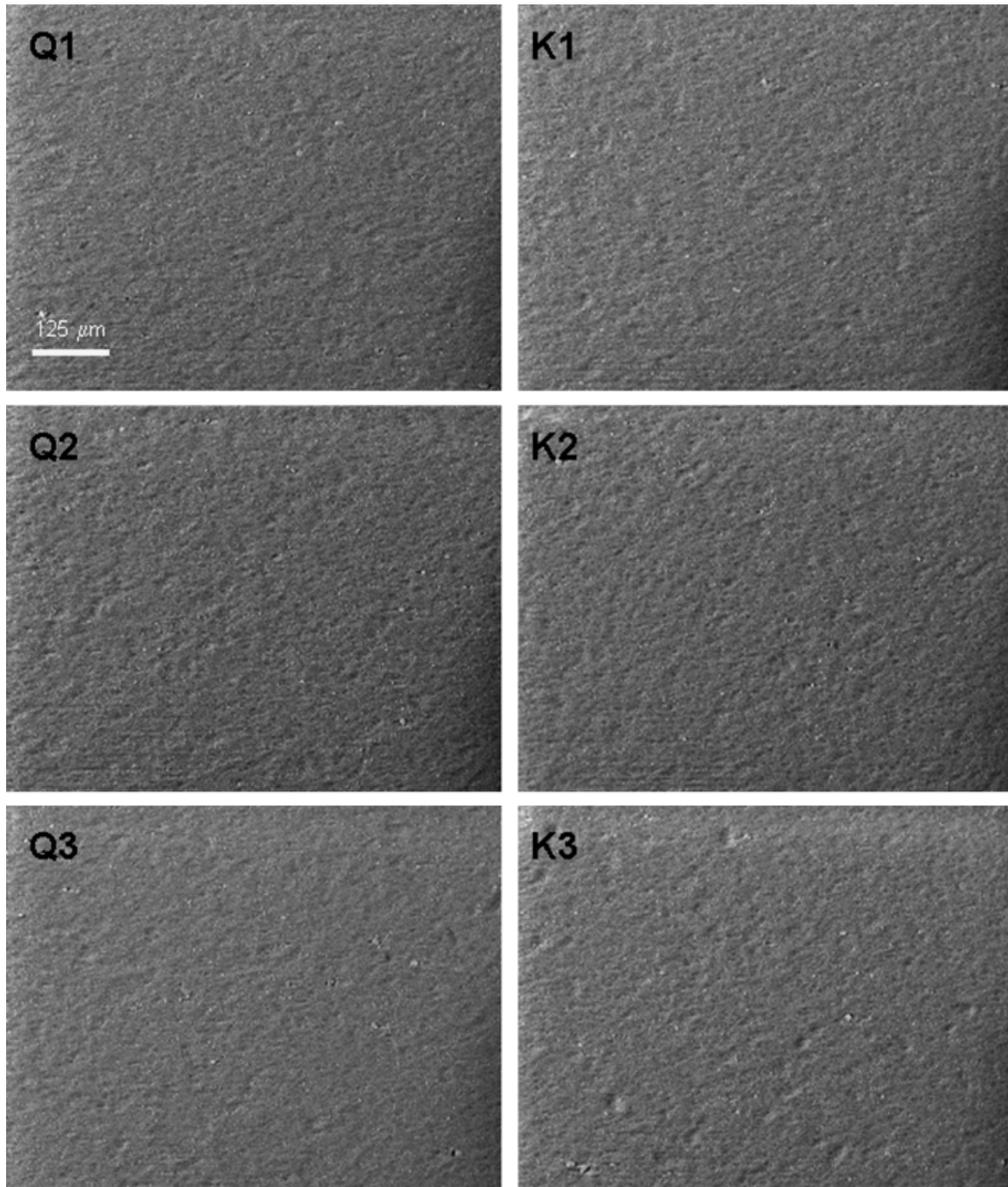


FIG. 15—Scanning electron microscope micrographs of three locations from a questioned sample recovered from the propane cylinder in Fig. 13 (Q1, Q2, and Q3) and three locations from a tape roll recovered from a suspect (K1, K2, and K3).

Figure 12 and Table 10 compare the elemental composition of the known and questioned samples. There are some clear similarities between the samples, including the presence of Pb which, along with the adhesive color, indicates that both tapes were manufactured by 3M before 2002. Qualitatively, both tapes contain the same elements (Al, Si, P, S+Pb, Ca, and Sb). Beyond these similarities, some systematic differences can be noted. One immediate point of differentiation are visibly higher levels of Al (P/B = 0.3) and Si (P/B = 0.3–0.4) in the questioned sample. Furthermore, unlike measurements made of the known and other exemplars, the emission intensities for Si exceeded the defined limit of detection. Based on a perusal of the 3M tapes in the data base, the tape recovered from the device can be seen as a class unto itself—it is unlike any of the 3M exemplars manufactured before 2002. As a result, conducting DA would be a trivial exercise. Interestingly, the roll of Tartan tape that was recovered from a different suspect was also found to have noticeable levels of Al and Si (P/B = 0.2–0.4), in contrast to the two exemplars in the data base. This indicates that a change in formulation may have occurred in Tartan brand tape, establishing a new class of tape with aluminosilicate filler.

The less ambiguous presence of Si in the questioned tape could in and of itself eliminate the known sample as the source for the questioned tape, but traditional statistical testing can further illustrate the differences between the samples. If the trace amount of Si in the known is integrated and compared with the questioned sample, quantitative differences can be seen. Table 10 includes the results of a Student's *t*-test in the form of a calculated *t* statistic. This statistic corresponds to the probability that the means for the two samples are equivalent for the element in question. Probabilities that fall to 1% or below indicate that one can reject the above hypothesis with an extremely high degree (99%) of confidence. Despite the cautions discussed above for samples with low numbers of data points, it cannot be overlooked that the relative amounts of both Si and Ca exhibit statistically significant differences between the known and questioned sample.

Overall, the differences discussed above contributed to the elimination of the known roll of tape as the source of the tape recovered from the device. An important concern when utilizing a surface technique such as SEM-EDS is the possibility that surface contamination of a questioned sample could inadvertently exclude it from the known source, which is generally more pristine. Contamination can arise from differing storage conditions, the effects of render safe procedures or, ultimately, blast effects. Recovering lower-lying tape layers from the questioned sample that are shielded from these effects is crucial and was achieved in this case. Given the sample area of the SEM ($\sim 1.6 \text{ mm} \times 2 \text{ mm}$), a clean section of tape as small as $c. 12 \text{ mm} \times 12 \text{ mm}$ is sufficient to generate at least three replicates with adequate spacing between the sampled areas. Samples of the known roll of Tartan were inherently pristine as a fresh, clean length of tape could be readily acquired.

It is also important to note that the use of SEM-EDS as a sole means for sample differentiation in case work is not recommended unless the samples are immediately differentiable. In this case, further analysis of the tape backing and adhesive by FTIR established additional systematic differences in the organic composition of the questioned and known rolls. The use of statistical techniques for the treatment of this type of data will be the subject of a future manuscript.

Case Study (Association)—The second case involves an IED composed of a small pipe bomb that was affixed to the propane fuel cylinder pictured in Fig. 13. This device was rendered safe,

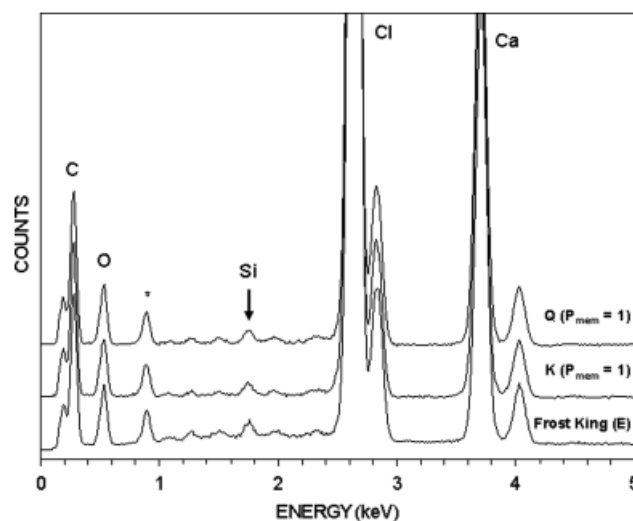


FIG. 16—Energy dispersive spectroscopy spectra of an exemplar of Frost King brand tape (roll E), the known tape sample (K) as well as the questioned sample (Q). The calculated probability that the questioned and known tape samples belong to the same group as the exemplar is 100%. Each spectrum is baseline corrected and represents the average of three locations. All intensities have been normalized to the principal (K_{α}) emission line of chlorine. The escape peak that results from this emission appears at $c. 0.9 \text{ keV}$ and is noted with an asterisk.

and it was noted that multiple lengths of electrical tape were wrapped around the cylinder. Subsequently, an exemplar roll of electrical tape was recovered from a suspect. In this case, the exemplar roll was unlabeled and had a plain white roll core, so its nominal brand was not known *a priori*.

Significantly, a physical match was observed between the outermost tape length on the propane cylinder and the end of the exemplar roll (Fig. 14). Given the flexible nature of electrical tape, a physical match of this sort is significant (23). In some laboratory protocols, in fact, this would obviate the need for further chemical analysis. However, it also is a rare and valuable opportunity to conduct an instrumental and statistical comparison on a “positive control” whereby a conclusive association between the known roll and at least the outermost tape fragment has already been established.

Both the tape on the device and that from the known roll had a measured width of approximately 11/16 in. This is unusual as tape widths are generally closer to the nominal value of 3/4 in. Both tape samples had clear adhesive, indicating they are general-use brands. Finally, significant overlap between the tape lengths on the propane cylinder created areas of clean backing material that were suitable to SEM-EDS analysis. Figure 15 contains three images of the surface texture for both the questioned (propane cylinder) and known (roll) tape samples. The two samples have reproducible surface features over the distance sampled with a noticeably rougher surface than was observed in the previous case example. Microscopically, the surface textures are highly similar.

EDS analysis of the known and unknown rolls showed that they both had rather simple elemental compositions that consisted of C, O, trace Si (S/B = 0.4–0.6), Cl and a relatively large amount of Ca. Trace amounts of other elements (e.g., Mg, Al, and P) were indicated but not reliably quantifiable. As per the previous example, DA was used to classify these tapes by calculating the PCA factor scores for each of the EDS replicates using tapes with clear adhesive that did not contain Al as a learning sample. The factor scores for both tape rolls were unambiguously assigned by DA to

TABLE 11—Comparison of the elemental composition of a questioned tape sample recovered from a propane cylinder (Fig. 13) and a known tape sample.

Sample	n		Normalized Peak Areas ($\times 100$)		
			Si	Cl	Ca
Q	3	Mean	0.75	95.83	28.57
		s	0.05	0.10	0.34
		% RSD	7.2	0.11	1.2
K	3	Mean	0.68	95.85	28.49
		s	0.04	0.02	0.06
		% RSD	5.8	0.02	0.2
		t-statistic (%)	14	80	78

RSD, relative standard deviation.

the group consisting of Frost King brand tape, roll E with a membership probability of 100%.

Figure 16 contains the visibly indistinguishable EDS spectra of the Frost King exemplar as well as the known and unknown sample from this case. Table 11 compares the relative peak areas for each element including a calculated t statistic. In contrast to the previous example, all elements in question are statistically indistinguishable.

It is important to note that the membership probability stated above reflects the fact that only one tape roll has been analyzed to date that has the same chemical composition as the known and questioned sample. While this formulation of Frost King may prove to be unique, another indistinguishable but as yet unknown brand may exist. Definitive brand identification of the known tape roll is not possible given the lack of any manufacturing marks on the roll core. However, it is important to note that of the dozens of tape rolls in the reference collection only recently manufactured rolls of the Frost King brand (rolls C, D, and E) have white roll cores that are devoid of any markings. In addition, the measured width of the Frost King (E) exemplar was approximately 11/16 in. Additional analysis of these tape samples using FTIR and GC/MS was completed and will be the subject of a future manuscript.

Conclusions

Based on the results discussed above, the following prerequisites are proposed for any analytical scheme that addresses the class characteristics of a mass-produced product such as electrical tape:

- (1) understand the product population, including manufacturing and distribution;
- (2) obtain a large, representative collection;
- (3) analyze samples using multiple orthogonal techniques;
- (4) avoid microheterogeneity through appropriate sample sizes;
- (5) assess the heterogeneity in the sample collection with rigorous quantitative methods; and
- (6) monitor changes in the population over time

This study has addressed the above-mentioned issues in a variety of ways. First, as much information as possible was gathered about the manufacturing process and distribution of electrical tape. For example, various large manufacturers of tape and their major brand names have been successfully traced via information such as the UL and CSA listing. At least seven manufacturers from four countries were identified, only one of which (3M) was located in the United States. Conversations with representatives of tape manufacturers confirmed a general trend of overseas production as the profit margins for electrical tape demand lower manu-

facturing costs. The exemplar collection itself was designed to be large, geographically diverse, and representative of various tape products. Overall, 34 nominal brands are represented and 67 exemplar rolls of tape were analyzed. These exemplars were purchased in seven different states from 1984 to present.

An advantage of the SEM-EDS method is that it reveals the physical texture as well as the inorganic content of the tape backing. As assessing the morphology of a tape surface in a quantitative fashion is not readily accomplished, it must be assumed that there is little to no correlation between the surface texture of a tape sample and its chemical composition. Hence, it is reasonable to assume that these techniques are largely orthogonal. The fact that surface texture could discriminate tape rolls of the same brand (e.g., Champion, Scotch 33, and Bengal) or tape rolls of two different brands with indistinguishable elemental compositions (e.g., Tartan and Duck 668 Pro) lends credence to this assumption. Additional analyses have been completed of the organic portion of electrical tape, including infrared spectroscopy of the tape backing and adhesive as well as GC-MS analysis of the plasticizer content. These techniques are equally amenable to statistical analysis, the results of which will be published in a future manuscript.

The issue of microheterogeneity and subsampling is critical when dealing with materials that may not be completely homogeneous, particular on microscopic scales. This concern can be addressed by examining relatively large portions of the tape backing, acquiring replicate measurements and verifying the reproducibility of all results. In this case, the precision of the results for any given element was generally quite good ($< 10\%$ RSD) and results were reproducible over long time periods.

As new or previously unknown products have been found, they will be included in the tape data base. As was demonstrated with the case studies, samples obtained from IEDs and suspects may turn out to be heretofore unknown brands or formulations of established brands. As this historical data is incorporated, the size of the database increases and it will become more representative of the diversity of the product population.

Overall, the quantitative comparison of tapes using AHC, PCA, and DA resulted in the following major findings:

- 3M tapes with black adhesive can be readily classified according to their nominal brand based on the presence or absence of Pb as well as the relative amounts of other fillers. The sole exception is the two premium brands (Super 88 and Super 33+), which are effectively indistinguishable both microscopically and chemically.
- Tapes with clear adhesive fall into two major categories: those with an aluminosilicate filler and those without. Samples containing Al and Si were well discriminated and correctly classified with a 1.7% error rate. For tapes without Al, one pair of brands (3M Tartan 1710 and Duck [668 Pro Series]) were found to be elementally indistinguishable. However, systematic differences in the surface texture of these two tape brands allows for their differentiation.
- Changes in the formulations of some tape brands can serve to further differentiate them and possibly indicate the year of manufacture. For example, 3M tapes with black adhesives and a Pb-based stabilizer were manufactured before 2002. Frost King brand tapes (ET60 and ET60FR) changed their formulations c. 2001 and 2005, respectively. Duck brand tape changed its formulation between 2004 and 2005.
- Overall, the 67 rolls of tape that were analyzed could be assigned to 36 differentiable groups based on surface texture and statistical analysis of elemental composition.

- The results of a single-blind study confirmed the confusion between the two 3M premium brands. Assignment of a tape sample to its correct brand was largely successful, including associating two samples from the same roll of tape to the same brand. Linking tape samples to their original rolls was likely to be erroneous. However, the stability of the SEM-EDS technique is such that tapes sampled days, weeks, or months apart were successfully associated with the same brand.
- Rolls of tape with differing nominal brands and the same listed manufacturer, and therefore possibly indistinguishable characteristics, could not be confirmed.
 - For example, Duck 668 Pro (362K) and Tartan 1710 (9Z53) are chemically very similar and both are listed as being manufactured in Taiwan. However, 3M appears to be co-listed and hence the manufacturing codes for Tartan are assigned to 3M.
 - Electrotuff, Intertape, and Radio Shack (362K) are all attributed to the same product manufactured by AICHEM Technology (Taiwan, China). While they have similar levels of aluminosilicate and calcium carbonate fillers, they differ in the use of a Pb-based stabilizer, Sb-based flame retardant and/or Ti filler. As noted above, frequent changes in formulation for a single tape product have been noted with other manufacturers and may be operative here.
 - Another example is Powerworks and Michigan Industrial Tools (590J), which are attributed to Symbio (Taiwan). In this case, some confusion occurred between the elemental compositions of the two tapes, but it was limited to a single observation. The fact that their chemical composition is qualitatively the same and only differentiable based on subtle quantitative differences may not be coincidental, however.
- The results of two case studies illustrate that careful examination of surface texture and elemental composition can either eliminate or associate electrical tape recovered from an IED and tape recovered from a suspect. In the first case, differing levels of Al, Si, and Ca served to exclude two tape samples. In addition, a potentially new formulation of Tartan brand tape was documented. In the second case, a physical match served as a positive control for the extent to which two tape samples will be microscopically and elementally indistinguishable under realistic conditions. Use of DA for the brand identification of the tape strongly suggests a recently manufactured roll of Frost King brand.
- While only a physical match of tape ends can be considered a definite association, this work demonstrates that two tape samples with surface textures and elemental compositions that are found to be indistinguishable (as discerned by multivariate techniques) may indeed originate from the same roll of tape. With only one exception observed to date (Super 88 and Super 33+), the common source for such samples can be conservatively described as at least the same nominal brand of tape manufactured with the same formulation. However, it must be acknowledged that other rolls of tape of that brand and formulation are likely to be similarly indistinguishable. Lastly, the size and diversity of the database assembled thus far allows for brand identification of unknown samples to include the nominal brand and potential year of manufacture.

Acknowledgments

The authors would like to acknowledge helpful discussions with Edward Bender, Amy Michaud, and Cassandra Burke of the ATF Electrical Tape Working Group. The cooperation of ATF Special Agents Scott Fulkerson and Daniel Fridley for use of data from their casework is also much appreciated. Finally, the authors

would like to thank the following industry representatives for their assistance: Glen Anderson (Pressure Sensitive Tape Council), Kevin Doolan (Plymouth-Bishop), John Houle (3M), John Johnston (Pressure Sensitive Tape Council), Bob McCarley (Intertape), and Jane Matulich (Underwriters' Laboratories).

References

1. Steele CA, Ball MS. Enhancing contrast of fingerprints on plastic tape. *J Forensic Sci* 2003;48:1314–7.
2. Howard S. Basic Fuchsin—A guide to a one-step processing technique for black electrical tape. *J Forensic Sci* 1993;38:1391–403.
3. Menzel ER, Savoy SM, Ulvick SJ, Cheng KH, Murdock RH, Sudduth MR. Photoluminescent semiconductor nanocrystals for fingerprint detection. *J Forensic Sci* 2000;45:545–51.
4. Houck MM, Budowle B. Correlation of microscopic and mitochondrial DNA hair comparisons. *J Forensic Sci* 2002;47:964–7.
5. Bradley MJ, Keagy RL, Lowe PC, Rickenbach MP, Wright DM, LeBeau MA. A validation study for duct tape end matches. *J Forensic Sci* 2006;51:504–8.
6. The Freedonia Group Inc. Pressure sensitive tapes to 2008. Cleveland: The Freedonia Group Inc, 2004.
7. Johnston J. Pressure sensitive adhesive tapes. Northbrook, IL: Pressure Sensitive Tape Council, 2000.
8. Kee TG. The characterization of PVC adhesive tape. Proceedings of the International Symposium on the Analysis and Identification of Polymers; 1984 July 31–August 2; Quantico (VA). Washington, DC: U.S. Department of Justice, Federal Bureau of Investigation, 1984.
9. Keto RO. Forensic characterization of black polyvinyl chloride electrical tape. *Crime Lab Digest* 1984;11:71–4.
10. Pressure Sensitive Tapes Council. Tape products directory. Northbrook, IL: The Council, 2005.
11. Blackledge RD. Tapes with adhesive backing: their characterization in the forensic science laboratory. In: Mitchell J Jr, ed. *Applied Polymer Analysis and Characterization: Recent Developments in Techniques Instrumentation and Problem Solving*. New York: Hanser, 1987;413–21.
12. Merrill RA, Bartick EG. Analysis of pressure sensitive tape. I. Evaluation of infrared ATR accessory advances. *J Forensic Sci* 2000;45:93–8.
13. Williams ER, Munson TO. The comparison of black polyvinyl chloride (PVC) tapes by pyrolysis gas chromatography. *J Forensic Sci* 1988;33:1163–70.
14. Postek MT, Howard KS, Johnson AH, McMichael KL. Scanning electron microscopy. Williston: Ladd Research Industries, 1980.
15. Currie LA. Limits for qualitative detection and quantitative determination: application to radiochemistry. *Anal Chem* 1968;40:586–93.
16. Kramer R. Chemometric techniques for quantitative analysis. New York: Marcel Dekker Inc., 1998.
17. Beebe KR, Pell RJ, Seasholtz MB. Chemometrics: a practical guide. New York: John Wiley & Sons Inc., 1998.
18. Cho LL, Reffner JA, Wetzel DL. Forensic classification of polyester fibers by infrared dichroic ratio pattern recognition. *J Forensic Sci* 1999;44:283–91.
19. Koons RD, Fiedler C, Rawalt RC. Classification and discrimination of sheet and container glasses by inductively coupled plasma-atomic emission spectrometry and pattern recognition. *J Forensic Sci* 1988;33:49–67.
20. Egan WJ, Morgan SL, Bartick EG, Merrill RA, Taylor III JH. Forensic discrimination of photocopy and printer toners. II. Discriminant analysis applied to infrared reflection-absorption spectroscopy. *Anal Bioanal Chem* 2003;376:1279–85.
21. Egan WJ, Galipo RC, Kochanowski BK et al. Forensic discrimination of photocopy and printer toners. III. Multivariate statistics applied to scanning electron microscopy and pyrolysis gas chromatography/mass spectrometry. *Anal Bioanal Chem* 2003;376:1286–97.
22. Limentani GB, Ringo MC, Ye F, Bergquist ML, McSorley EO. Beyond the *t*-test: statistical equivalence testing. *Anal Chem* 2005;77:221A–6A.
23. Agron N, Schecter B. Physical comparisons and some characteristics of electrical tape. *AFTE J* 1986;18:53–9.

Additional information and reprint requests:

John V. Goodpaster, Ph.D.
ATF National Laboratory Center
6000 Ammendale Road
Ammendale
MD 20705-1250
E-mail: john.goodpaster@atf.gov