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A Comparison of Three Techniques Developed for Sampling and Analysis of Gunshot Residue by Scanning Electron Microscopy/Energy-Dispersive X-Ray Analysis (SEM-EDX)

ABSTRACT: Three gunshot residue (GSR) collection methods from hand samples by scanning electron microscopy/energy-dispersive X-ray analysis (SEM-EDX) were compared: the tape lift, glue lift, and concentration techniques. Efficiency of particle collection was examined based on the number of rounds fired, the temperature, and the shelf life. The tape lift surface demonstrated excellent particle collection ability, and it remained stable for all conditions tested. Glue lift was less efficient under all conditions tested. Collection followed by concentration gave highly variable results. A table of advantages and disadvantages of each technique was developed.

KEYWORDS: criminalistics, ballistics, sampling, gunshot residues, glue lift, tape lift, concentration technique, scanning electron microscopy, energy dispersive X-ray analysis

Scanning electron microscopy with energy dispersive X-ray analysis (SEM-EDX) has several advantages over other current techniques in the analysis of gunshot residue (GSR). Unlike neutron activation analysis (NAA) or flameless atomic absorption spectroscopy (FAAS), which are destructive bulk elemental techniques, SEM-EDX is a nondestructive technique which can be applied to a specific particle of GSR. Consequently, the technique is not subject to spurious results from background or contaminating levels of lead, barium, and antimony, because these elements do not combine to form spheroid particles in the environment [1]. In addition, GSR analysis by SEM-EDX is more sensitive than bulk analysis techniques. This is again due to its ability to analyze individual particles of gunshot residue, thus permitting detection even in cases where very few particles remain on the hand.

Although several methods have been developed for collection of GSR, no thorough investigation has been made comparing several of the most common methods for GSR collection and analysis by SEM-EDX. The results of such an investigation should allow

crime laboratories to select a collection technique for GSR. Currently, each laboratory must decide somewhat arbitrarily which collection method it will use for GSR analysis by SEM-EDX. By determining the strengths and weaknesses of each technique, the study will enable crime laboratories to make more informed choices.

While SEM-EDX is conclusive in describing the morphology and elemental composition of the particles in question, it suffers from the excessive analysis time (up to several hours) required [2–4]. An ever-increasing caseload in the laboratory requires a collection technique that is rapid, dependable, and efficient.

Analysis time can be reduced by the following:

1. Increasing the sample concentration—Such a technique was developed by D. C. Ward [4] for concentrating GSR from a Vistanex adhesive-coated surface by repetitive centrifugal concentration through a high-density liquid.

2. Increasing the efficiency of particle lifting from the hand—The tape-lift method is said to have excellent particle lifting ability and has been employed by many researchers [1,3,5–8]. An alternative to tape is the glue lift method developed by Basu and Ferriss [2,9,10].

3. Decreasing the area to be scanned—Ward scanned a 2-mm area [4], and Basu and Ferriss scanned several 1.5-mm areas [2]. A general reduction in stub size from 25 to 5 mm has also been employed.

This paper is a comparative study of the concentration technique, described by Ward [4] with modifications by Sugarman [the technique was described at the American Academy of Forensic Sciences (AAFS) meeting, in San Diego, CA, in February 1987]; the glue lift technique, described by Basu and Ferriss [2]; and the tape lift technique, described by Nesbitt et al. [3].

Materials and Methods

Firing and Collection

The shooter washed and dried his hands prior to firing. The weapon was cleaned prior to firing. The same weapon was used for all firings [a Smith and Wesson Model 10-8 revolver, with a 2-in. (5.1-cm) barrel]. The ammunition used was Federal .38 Special caliber, jacketed soft point lead, 125 grain for law enforcement use. The firing distance was 32 in. (81.3 cm) from the barrel to the target face (cotton box). After firing, the shooter left the firing room and his hand was sampled by the researcher. The researcher wore latex gloves and sampled the thumb, web, and index finger of the firing hand by successive dabbings of the hand with the collection stub. In the case of the glue lift technique, five gentle dabs of the hand were made [2]. The collection stub was then numbered and covered with its protective cap. The collection stub was removed from its support piece (rubber stopper) and carbon coated in the vacuum evaporator or processed through the concentration technique as described by Sugarman (AAFS meeting, February 1987).

Controls

Before analysis on the electron microscope, all samples were assigned a number using a table of random numbers. The analyst knew what type of collection device was being analyzed by the surface characteristics of the device but had no idea what treatment the collection device had received or whether it was a control collector. For every treatment, five repetitions of firing and GSR collection were made, and a control was run. The

control consisted of a "hand control," where the subject who had been firing the revolver washed his hands and was then sampled.

Collection Devices

The tape-lift and glue-lift collection devices consisted of an inverted No. 3 stopper that had a 15-mm diameter aluminum stub mounted on top of it. The surface of the aluminum stub was coated with either a 3M brand 465 adhesive transfer tape [3] or a 15-mm-diameter polished-carbon planchet coated with rubber cement diluted 1:4 with toluene [2,9,10]

Each collection device was covered by a protective cap, consisting of a No. 20 plastic test tube closure. Unless otherwise noted, the devices were stored at room temperature and used within 24 h after preparation.

The collection devices for the concentration technique consisted of a 15-mm-diameter Mylar surface coated with Vistanex adhesive diluted to 15% with hexane.

Electron Microscopy

The JEOL 35C scanning electron microscope was operated at an accelerating voltage of 25 kV. The lithium-drifted silicon crystal of the X-ray detector was kept in liquid nitrogen at a distance of 55 cm from the center of the column. The working distance for the specimen was 39 mm. A brass specimen holder with a 25-mm diameter was lined with an aluminum adapter to accommodate the 15-mm-diameter stub being analyzed. An objective aperture setting of 600 μm was used to increase the signal to the solid-state backscatter detector. A known sample of gunshot residue collected on 3M brand 465 adhesive transfer tape (three rounds fired) was inserted into the microscope as a standard for fine adjustment of the backscatter image. The image was focused at ×300 in the secondary electron image (SEI) mode. The backscatter image, which is sensitive to increasing atomic number, was collected using the slow scanning option while adjusting the gain and contrast to give a dark background, with GSR appearing as bright circular white spots with a circular halo around them.

The surface of all the collection devices was searched manually by a single operator at ×300 magnification in the backscatter mode. A particle was selected, and the magnification was increased so that the particle image filled the majority of the screen. The image was refocused in the SEI mode, and an X-ray spectrum was accumulated for 70 s at a beam current of 550 pA. The particle was confirmed as gunshot residue if it fell into one of the following four categories:

(a) lead (Pb), barium (Ba), and antimony (Sb);
(b) Ba and Sb;
(c) Ba, calcium (Ca), and silicon (Si), with a trace of sulfur (S); and
(d) Ba, Ca, and Si, with a trace of Pb, provided that no zinc (Zn) was present [residue from stud guns has been found to contain Ba, Ca, Si, Pb, copper (Cu), and Zn] [1].

Dependent Variables

Efficiency of Collection—In this study, efficiency of collection is defined as the number of GSR particles found in 1 h of searching at ×300 magnification in the backscatter mode in the SEM. If five GSR particles were found in <1 h, the time taken to find the particles was recorded, and the number of particles found in 1 h was extrapolated according to the equation

$$\frac{60 \text{ min}}{\text{search time (min)}} \times \text{particles found} = \text{particles found/h}$$

This analysis assumes that GSR was distributed randomly on the stub [11].

Analysis Time—Analysis time is defined as the time required from insertion of the sample into the electron microscope (time zero) to the moment that five particles whose spectra and morphology characterize them as GSR have been found and saved to disk. An upper limit of 1 h was set for the analysis time per sample.

Concentration Time—This refers only to the collection technique using the Vistanex adhesive. Concentration time is defined as the time from removal of the Mylar surface to the moment that the 0.45-μm filter was dry and ready to be carbon coated.

Independent Variables

Number of Rounds Fired—Either one or three rounds were fired. Sampling was conducted using a collection device prepared 24 h in advance and stored at room temperature.

Temperature—Holding the number of rounds fired constant at three, each collection device was prepared 24 h in advance, keeping the devices at a temperature of 56°C or −4°C for 12 h prior to sampling.

Time—Holding the number of rounds fired constant at three and the temperature constant at 22°C (room temperature), collection devices were prepared and stored for three or six weeks prior to sampling.

Statistics

Three different methods of collecting GSR were examined. They included the tape lift, the glue lift, and a Vistanex glue lift, followed by concentration via centrifugation.

For each collection method, six different treatments were examined. The treatments included: (*a*) three rounds fired, (*b*) one round fired, (*c*) the collection device stored for 12 h at −4°C, (*d*) the collection devices stored for 12 h at 56°C, (*e*) the collection devices stored for three weeks prior to use, and (*f*) the collection devices stored for six weeks prior to use. Unless otherwise stated, the collection devices were prepared 24 h in advance and the residue was collected from three rounds fired.

In addition to the five repetitions of firing and GSR collection for each treatment, a "hand control" sample was collected from the hand (see the section for the firing and hand sampling procedure). Washing of the hands is presumed to be sufficient to remove gunshot residue; however, depending on how thoroughly the hands are washed, some GSR may remain [12].

GSR collections were made one method at a time, beginning with the tape lift method. A single treatment consisting of five repetitions and a "hand" control was completed on a given day. After all the GSR collections had been made for the tape lift method and the glue lift method, the collection devices were renumbered, using a table of random numbers, and analyzed in a random fashion on the electron microscope.

Assuming a random distribution of particles on the collection surface [11], the particle count data were normalized to time. For ease of statistical manipulation, the number of particles per 1-h search time was determined. The search time was equal to the time taken to find five GSR particles, or 60 min if less than five GSR particles were found.

The ln $(x + 1)$ transformation was required to fulfill the homogeneity of variance assumption of ANOVA (Analysis of variance using general linear models—Procedure GLM, SAS, 1983 [14]). Homogeneity of variance within method and between treatments was examined with Barlett's test [13]. Means from ln $(x + 1)$ transformed data were separated via the SNK (Student Newman Keuls) procedure or Student's *t*-test following ANOVA.

Results

Comparison Between Collection Methods

An indication of the collection device particle-lifting efficiency was reflected by the mean number of particles found per hour. Obviously, the more particles found per unit of time, the greater the efficiency of collection. The data in Table 1 indicate that the ln $(x + 1)$ particle per hour means were significantly different from each other at the 0.05 alpha level for the three collection methods tested.

If one examines the particles per hour mean for the tape lift method, the mean particles per hour were more than five. In other words, on the average, it took less than 1 h to find five particles of GSR on the tape lift surfaces (Table 1).

The particle per hour means for both the concentration technique and the glue lift were much lower than that in the tape lift method.

Comparisons Within Collection Methods

The data in Table 2 suggest that the collection efficiency of the 3M-type 465 adhesive transfer tape was stable under all conditions tested. The SNK test showed a significant

TABLE 1—*Statistical analysis of the ln (x + 1) of particles, per hour means between collection methods.*[a]

Method	SNK Grouping[b]	Mean of ln $(x + 1)$ of Particles per Hour	N	Mean of Particles per Hour
Tape lift	A	1.973	30	6.192
Concentration technique	B	1.081	30	1.948
Glue lift	C	0.538	30	0.713

[a]Calculated by SNK procedure using analysis of variance.
[b]Means sharing the same letter are not significantly different from each other at alpha = 0.05 level.

TABLE 2—*Statistical analysis of the ln (x + 1) of particles, per hour means for tape lifts.*[a]

Treatment	SNK Grouping[b]	Mean of ln $(x + 1)$ of Particles per Hour	N	Mean of Particles per Hour
1 Round fired	A	2.233	5	9.380
3 Rounds fired	A	2.188	5	8.100
−4°C	A	2.105	5	8.320
3 Weeks old	A	1.938	5	7.540
56°C	A	1.694	5	5.240
6 Weeks old	A	1.678	5	6.128
Hand control	B	0.384	5	2.567

[a]Calculated by SNK procedure using general linear models.
[b]Means sharing the same letter are not significantly different from each other at alpha = 0.05 level.

difference in means [ln $(x + 1)$ of particles per hour] at an alpha level of 0.05 for the hand control data compared with the other treatments, as would be expected.

The concentration technique gave highly variable results between treatments tested. A comparison of the ln $(x + 1)$ of particles per hour means for the treatments in the concentration method by SNK resulted in means that were significantly different at the 0.05 alpha level (Table 3). Specifically, one-round-fired samples had the highest mean and were significantly different from the six-week-old collection samples with three rounds fired. The potential source of the decreased particle counts found in the concentration method will be examined in the discussion section. In the present study, statistical analysis by SNK of ln $(x + 1)$ of particles per hour means for the glue lift method showed no significant differences between hand controls and any of the treatments examined (Table 4). Too few particles per stub surface were found. This is indicative of an inefficient particle-lifting surface. Note the mean number of particles found per hour for the glue lift technique in Table 4.

In order to test the efficiency of the glue lift technique, an experiment was designed as described in the procedure for three rounds fired, in which the hand was sampled by

TABLE 3—*Statistical analysis of the ln (x + 1) of particles per hour means for concentration method.*[a]

Treatment	SNK Grouping[b]		Mean of ln (x + 1) of Particles per Hour	N	Mean of Particles per Hour
1 Round fired	A		1.828	5	6.080
3 Weeks old	A	B	1.520	5	4.160
56°C	A	B	1.430	5	4.320
Hand control	A	B	0.855	5	1.600
−4°C	A	B	0.748	5	1.800
6 Weeks old		B	0.599	5	1.200
3 Rounds fired		B	0.358	5	0.600

[a]Calculated by SNK procedure using general linear models.
[b]Means sharing the same letter are not significantly different from each other at alpha = 0.05 level.

TABLE 4—*Statistical analysis of the ln (x + 1) of particles per hour means for glue lifts.*[a]

Treatment	SNK Grouping[b]	Mean of ln (x + 1) of Particles per Hour	N	Mean of Particles per Hour
1 Round fired	A	1.011	5	2.540
3 Rounds fired	A	0.832	5	1.800
3 Weeks old	A	0.555	5	0.800
6 Weeks old	A	0.416	5	0.600
56°C	A	0.277	5	0.400
−4°C	A	0.139	5	0.200
Hand control	A	0.000	5	0.000

[a]Calculated by SNK procedure using general linear models.
[b]Means sharing the same letter are not significantly different from each other at alpha = 0.05 level.

TABLE 5—*Statistical analysis of ln (x + 1) of particles per hour means for glue lifts followed by tape lifts.*[a]

Method	T Grouping[b]	Mean of (ln $(x + 1)$ of Particles per Hour	N	Mean of Particles per Hour
Tape lift	A	1.788	5	6.040
Glue lift	B	0.8832	5	1.400

[a]Calculated by Student's *t*-test (least significant difference [LSD]).
[b]Means sharing the same letter are not significantly different from each other at alpha = 0.05 level.

twelve dabs with the glue lift, and the same area of the hand was then sampled with a tape lift collector. The procedure was repeated five times, with gun cleaning and hand sampling as described previously. Tape lift stubs were carbon coated and all stubs were assigned random numbers and analyzed on the electron microscope.

An ANOVA by Student's *t*-test indicated a significant difference in the ln $(x + 1)$ of particles per hour means for the glue lift and tape lift collectors, (alpha = 0.05). These results demonstrated the less efficient particle-lifting surface of the glue lift devices (Table 5). Decreased collection efficiency was reflected not only in the particles per hour means being much lower in the glue lift than in the tape lift method, but also in the fact that particles collected on the tape lift surface represent GSR that was left on the hand after initially sampling the hand with a glue lift device.

Discussion

The tape lift collection devices in this study proved to be the most efficient particle-lifting devices examined. The concentration technique gave highly variable results between treatments but still had a higher ln $(x + 1)$ of particles per hour mean than the glue lift technique (Table 1).

The tape lift surfaces were found to be stable under all treatments tested. They have a shelf life of at least six weeks and were not effected by 12-h exposure to temperatures which might be encountered by collection devices stored in a crime scene vehicle.

The concentration technique, on the other hand, gave highly variable results between treatments (Table 3). The concentration technique is actually a combination of two techniques: collection and concentration. Whether a decreased number of particles found per hour was due to the Vistanex surface being less efficient in collecting the particles or due to loss of particles during the concentration procedure cannot be determined from the present data.

There are at least three possible areas where GSR may be lost in the concentration method: (1) particles lost from the nonadhesive 0.45-μm Nucleopore filter when it was teased away from subfilter; (2) actual particles not counted because of Ba, Ca, and Si contamination of the subfilter; and (3) particles trapped in debris and either aspirated out of the concentrator or pelleted onto the filter surface.

The data, in fact, suggest that a problem may exist in the concentration procedure. If one examines the particles per hour means of three rounds fired versus one round fired in the concentration method, the means are 0.60 and 6.1, respectively (Table 3). This was exactly the opposite of the results one might expect. Going back to the actual concentration procedure employed, it was noted at the time that in six out of the seven concentrators, the 0.45-μm Nucleopore filters could not be peeled away from the underlying Nucleopore D-79 subfilter. The filters were mounted together on an aluminum
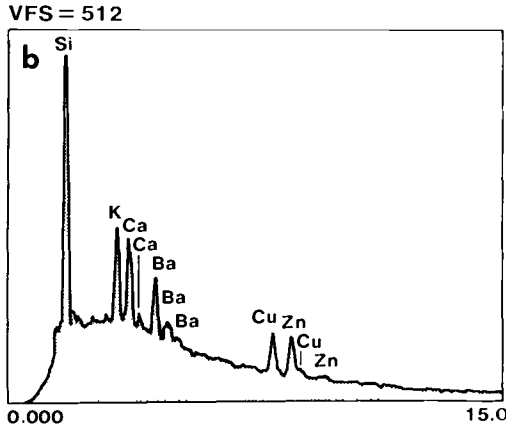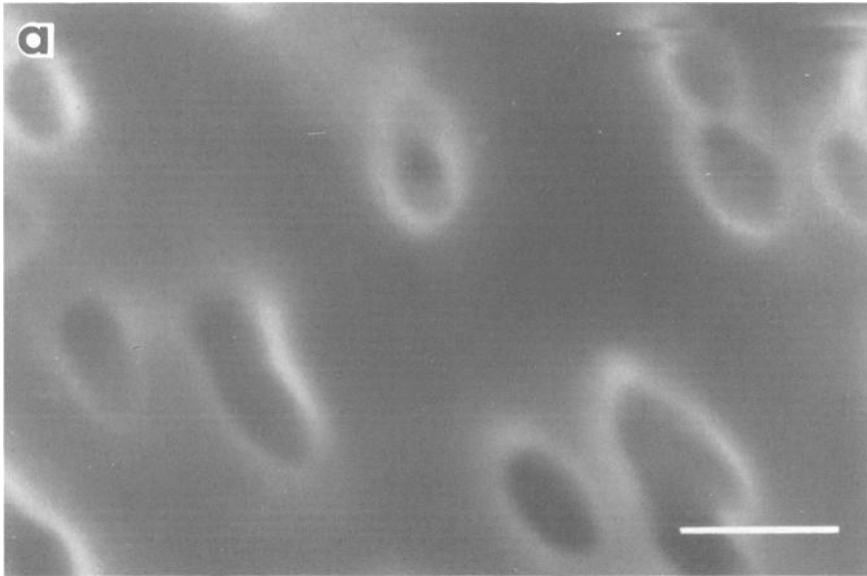
FIG. 1—*Identification of contaminating elements on a filter surface used in the concentration technique: (a) secondary electron image of a 0.45μm Nucleopore filter adhered to a D-79 subfilter after carbon coating (magnification = ×16 000; bar = 1 μm); (b) energy-dispersive X-ray spectrum of the filter sandwich in (a). Vertical full scale = 512 counts; x-axis is from 0.0 to 15.0 KeV.*

stub, carbon coated, and viewed. Analysis in the electron microscope gave high background levels of Ba, Ca, Si, and potassium (K) for the 0.45-μm Nucleopore filters adhering to the subfilters (Fig. 1). The surface itself tended to pucker and charge to a degree that a reliable backscatter image was unattainable. At this point, the 0.45-μm filter was dissected away from the subfilter with a razor blade and remounted on an aluminum stub. This required manipulation of the 0.45-μm filter, which has no adhesive nature of its own, and it is quite likely that GSR particles were lost during this manipulation.

In contrast, with the concentrators used in the one-round-fired experiment, five out of seven Nucleopore filters were easily removed from the subfilter surface, with the remaining two picking up only slight subfilter contamination.

After barium and calcium contamination was observed, a subfilter was mounted on an aluminum stub and carbon coated to determine what elements were present in the subfilter. Results of this analysis showed the presence of silicon, potassium, zinc, calcium, and barium (Fig. 2). This contamination compounds the problem of GSR analysis. One of the forms of GSR recognized in the study by the Aerospace Corp., Segundo, California,
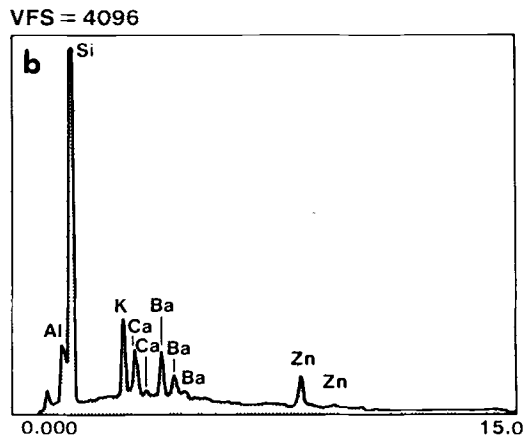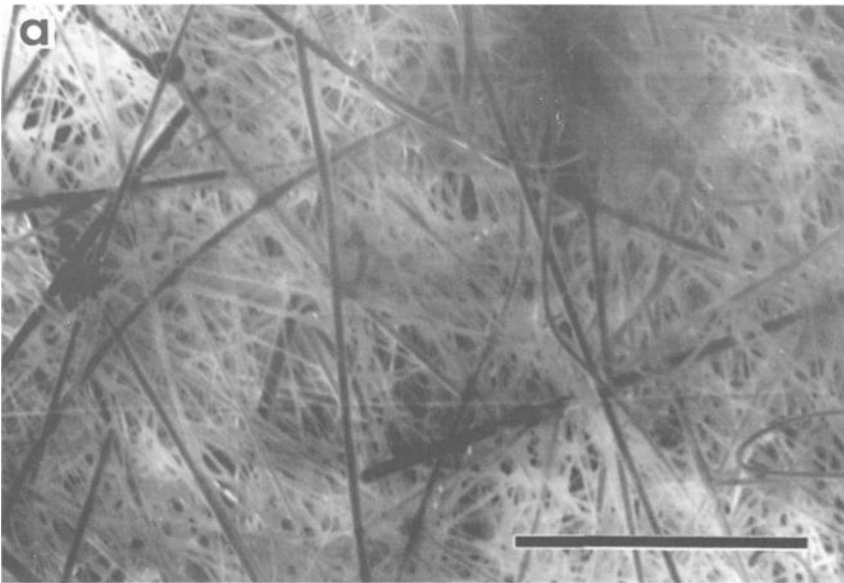

FIG. 2—*Identification of contaminating elements in the subfilter used in the concentration technique:* (a) *secondary electron image of carbon-coated D-79 subfilter (magnification* = ×*300; bar* = *100* μm); (b) *energy-dispersive X-ray spectrum of subfilter surface in* (a). *Vertical full scale* = *4096 counts; x-axis is from 0.0 to 15.0 KeV.*

[*11*] was spheroid particles containing the elements lead or sulfur, silicon, calcium, and barium. On a 0.45-μm Nucleopore filter adhering to a subfilter, such a particle would be difficult to distinguish from a pure lead or sulfur particle. Therefore, all GSR reported in the concentration method consisted of particles composed of lead, barium, and antimony, unless the filter surface showed no background element contamination from adhering subfilter material.

Other investigators, using a different type of concentrator, noticed lead and barium contamination of the 50-μm porous polyethylene filter in their concentration devices. Washing the filter with 20% hydrochloric acid was found to remove the contamination in this case [*16*].

A third factor, which could influence the number of particles found after concentration, is the amount of debris deposited on the 0.45-μm Nucleopore filter. One of the main reasons originally proposed for using the concentration method was its reduction of the amount of epidermal cells and other debris picked up by the collection device. Such debris may cover the GSR particles present, making them undetectable in the electron microscope [*4*]. Dennis Ward at the FBI crime laboratory has suggested that centrifugal force may be a critical factor, depending on the amount and type of debris present on the Vistanex surface (personal communication, 1987). Too low a *g* force results in material floating on the bromoform surface, which may have trapped GSR in it. Too high of *g* force may pellet debris onto the filter, obscuring the GSR particles.

Varying amounts of debris were found on the filter surface in the concentration technique (Fig. 3). While the present study was being conducted, an experiment comparing tape lifts with the concentration technique was performed by Zeichner et al. [*16*]. These researchers concluded that the buildup of debris on the filter was such a problem that direct observation of a tape or glue lift surface was preferable to concentration.

The concentration technique used in the current study was a modification of Ward's technique [*4*] developed by Loren Sugarman at the Forensic Science Laboratory of the Orange County Sheriff–Coroner Department in Santa Ana, California. Sugarman has been able to circumvent some of the problems in the present study (personal communication, 1988). He has not noticed any contamination of the D-79 subfilters obtained from Nucleopore, which suggests that the contamination of the subfilters in the current study may be a batch defect. To reduce adhesion between the 0.45-μm filter and subfilter, Sugarman recommended placing the subfilter with the cross-hatched surface facing up and removing the 0.45-μm Nucleopore filter immediately after centrifugation. He also advised washing the filter with methanol thoroughly after the bromoform step to remove any traces of bromine on the filter surface, which would give interfering backscatter signals.

The glue lift technique was found to have an inefficient particle-lifting surface (Table 1). These findings were not in agreement with the observations published by the developers of the glue lift technique [*2*].

The glue lift technique was developed by Dr. Samarendra Basu and Dr. Stark Ferriss [*2*]. It was designed to be less sticky than the tape lift surface. The reasoning was that the decreased stickiness of the glue lift surface would not collect so much interfering epidermal cells and other debris.

In their original paper on the development of the glue lift technique, several advantages of the glue lift surface were demonstrated in comparison with the tape lift surface [*2*]. The advantages included (*a*) ten or more particles found per area searched (1.5-mm-diameter circle), (*b*) no electron-beam damage to glue lift surface as opposed to melting of the tape lift surface, (*c*) smoother surface of the carbon planchet, and (*d*) no carbon coating required.

At this point it is worth examining some differences between the two studies. The carbon planchets used in the study by the developers of the glue lift were obtained from
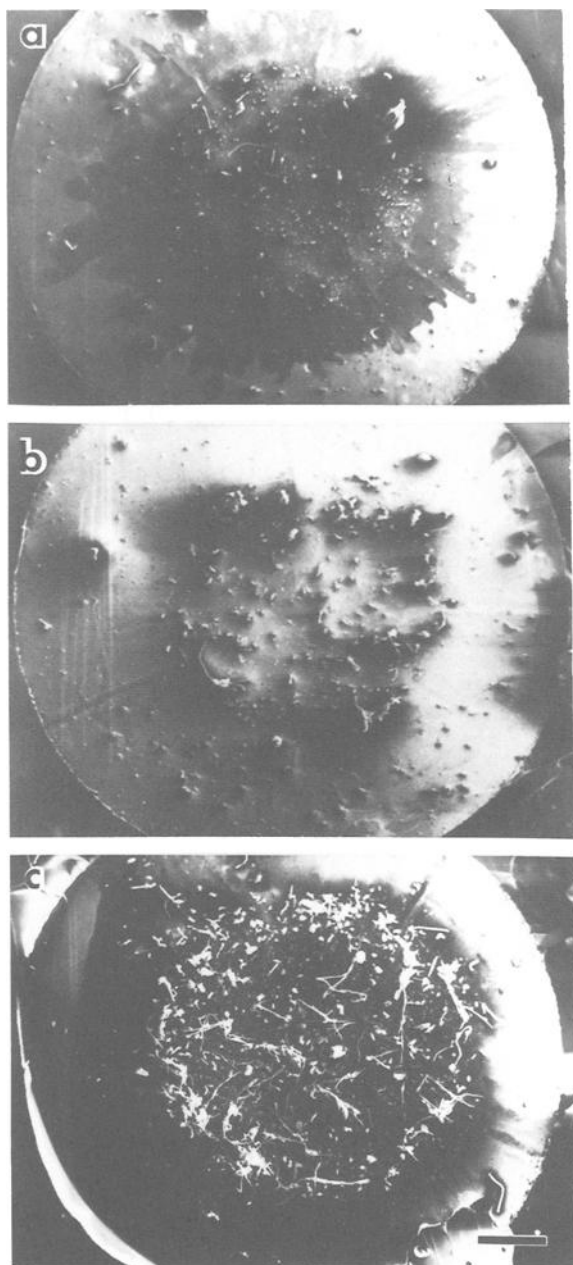
FIG. 3—*Varying amounts of debris found on filter surface after the concentration technique:* (a) *secondary electron image of the surface of a 0.45-µm Nucleopore filter after GSR concentration (a small amount of debris can be observed scattered across the center of the filter);* (b) *secondary electron image of the surface of a 0.45-µm Nucleopore filter after GSR concentration (many pieces of debris can be seen across the center of the filter);* (c) *secondary electron image of the surface of a 0.45-µm Nucleopore filter after GSR concentration. The filter surface is almost totally obstructed by debris. All micrographs are at a magnification of ×10; bar = 1000 µm.*
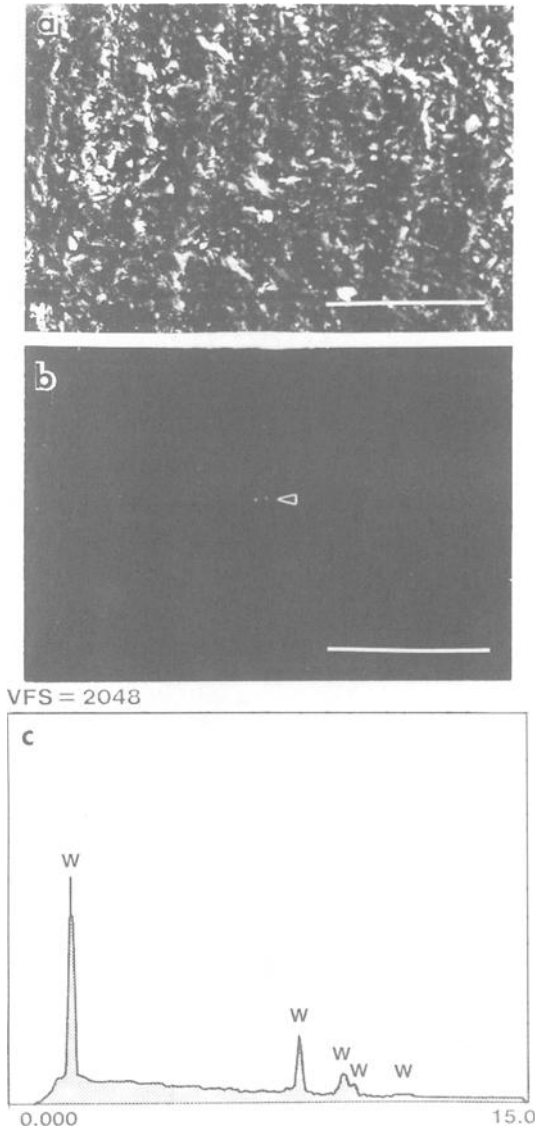
FIG. 4—*Identification of contaminating tungsten particles on a carbon planchet:* (a) *secondary electron image of the surface of a carbon planchet obtained from Ladd Industries Inc. (magnification = ×300; bar = 100 μm);* (b) *backscatter electron image of the same field as in* (a) *(arrowheads point to contaminating tungsten particles; magnification = ×300; bar = 100 μm);* (c) *energy-dispersive X-ray spectrum of a single tungsten particle. Vertical full scale = 2048 counts; x-axis is from 0.0 to 15.0 KeV.*

Ernest F. Fullam Inc. (Schenectady, New York). They were described as the "clean, polished carbon planchets [disk thickness ⅛ in. (3 mm), diameter ½ in. (12.7 mm)]." In the present study, carbon planchets were obtained from Ladd Research Industries, Inc. (Burlington, Vermont); these were specially smoothed surface carbon specimen mounts (disk thickness 5 mm, diameter 15 mm). At high magnification, the surface of these carbon planchets appeared somewhat irregular (Fig. 4). Basu (Albany Crime Laboratory,

Albany, New York) suggests that lack of a smooth regular surface may impair particle-lifting ability (personal communication, 1987). In addition, these carbon planchets were contaminated with tungsten particles, generally of about 0.5 to 2.0 μm in diameter (Fig. 4). Spraying the planchets with compressed air prior to applying the glue surface proved insufficient to remove all of the tungsten particles. The tungsten was presumably material left behind during the manufacturing process. An average of nine contaminating tungsten particles per glue lift was encountered. These particles mimic GSR in the backscatter mode and lengthen the analysis time.

The authors of the glue lift technique examined a minimum of four 1.5-mm circles on the glue lift surface, finding an average of 58 particles per circle [2].

Typically, 15% of the total surface area of the 15-mm-diameter carbon planchet could be covered during 1 h of searching. This was equivalent to examining 2.85 of the 1.5-mm circles described by the developers of the glue lift [2].

The ammunition used in the two studies was also different. The ammunition used in the present study was Federal .38 Special caliber, jacketed soft point lead, 125 grain for law enforcement use. Ammunition from the same lot number (12 A) was used throughout the study. This ammunition was chosen because it gave consistently fewer gunshot residue particles. It was felt that this resembled actual casework conditions in a more realistic fashion than an ammunition that produces hundreds to thousands of particles on the hand. The developers of the glue lift used either standard Winchester or Remington ammunition [2] for the pistol loads. In the present study, Remington .38 caliber, 158-grain lead ammunition was test fired from the same revolver used in this work and found to produce hundreds of particles, mainly lead in composition. The glue lift developers' findings of 30 to 116 residues per 1.5-mm-diameter circle [2] on the glue lift surface was consistent with the Remington ammunition tested. However, the finding that a typical ½-in. (12.7 mm)-diameter tape lift disk may contain from 2 to 10 observable GSR [2], with one round fired, was not consistent with the Remington ammunition tested. In support of the finding of few GSR on a tape lift surface, the authors quote a table in the work by Sild and Pausak [17], where it was mentioned that after firing two shots with a .38 caliber revolver, recovered slightly more than 8 GSR and 20 lead particles were recovered from a 1-in. (2.54-cm)-diameter tape-lift disk [2]. If one examines Table 1 in this work by Sild and Pausak [17], one can also find a test firing of two shots with a .38 caliber revolver where a single sweep (magnification × 1000) shows more than 20 particles of lead and more than 20 particles of GSR (Pb + Sb + Ba) found on the whole stub.

In the original study of the glue lift technique, Basu and Ferriss cite the beam damage that occurs on the tape lift surface. They demonstrated this with a micrograph (Fig. 2f in Ref 2) depicting "a lead particle disappearing into a cavity on transfer tape, created by the bombarding electrons." The authors went on to discuss particles imbedding themselves and disappearing into the melted tape surface.

In the present study, where hundreds of particles of various compositions were observed and spectra were obtained, no particle was ever seen to "disappear into the melted surface of the tape." Electron-beam damage to the tape surface did occur and usually appeared as a crater with surrounding folds around the particle (Figs. 5a and 6a). The only time the tape lift surface was seen to crack or melt severely was when it was not coated with enough carbon initially (Fig. 7). This problem was easily remedied by applying another carbon coat. The thickness of a single carbon coat was typically in the range of 35 to 40 nm.

Another way to induce electron-beam damage is by using excessive beam current. In the work by Basu and Ferriss [2], the beam or specimen current used was not mentioned. The emission current was listed as 100 μA, but this gave no information as to the current which the specimen was encountering. In the present study, a beam current of 550 pA
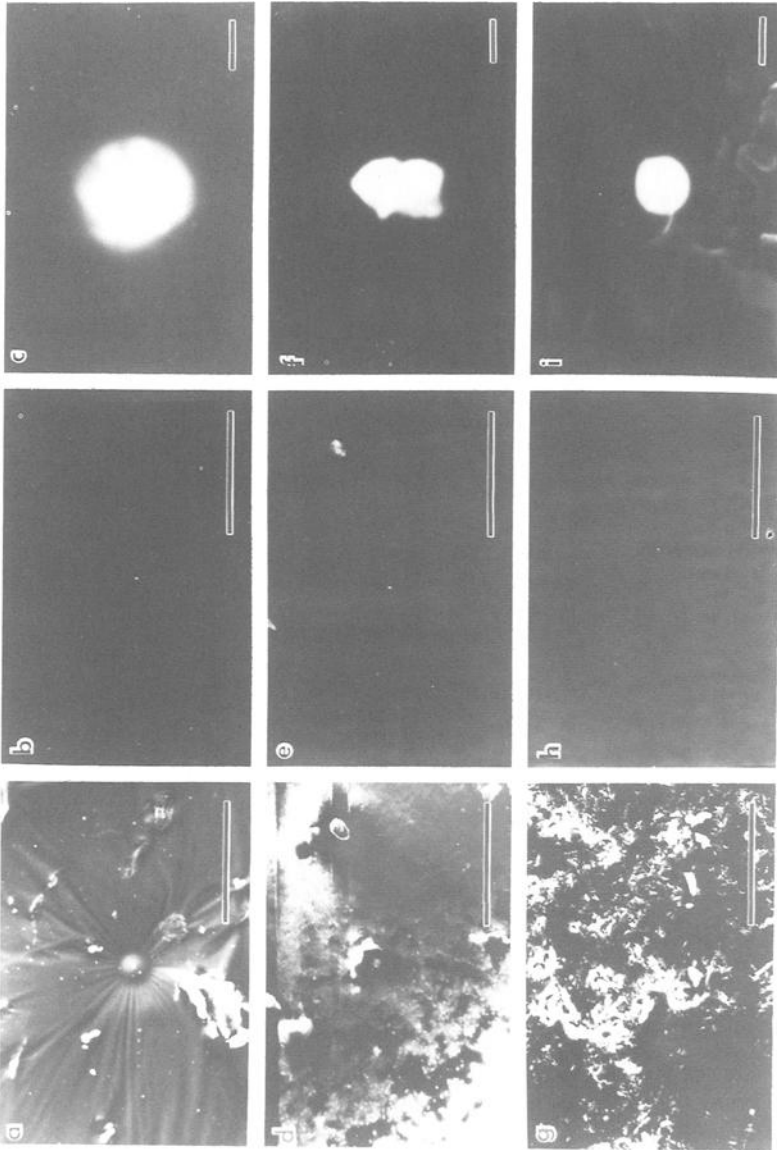
FIG. 5—Electron micrographs of small GSR particles on various surfaces: (a) secondary electron image of GSR on a tape lift surface; (b) backscatter electron image of GSR particle in (a); (c) enlargement of GSR particle in (a); (d) secondary electron image of GSR on a 0.45-μm Nucleopore filter surface; (e) backscatter electron image of GSR particle in (d); (f) enlargement of GSR particle in (d); (g) secondary electron image of GSR on a glue lift surface; (h) backscatter electron image of GSR particle in (g); (i) enlargement of a GSR particle in (g). Magnification for micrographs (a), (b), (d), (e), (g), and (h) = ×300; bar = 100 μm. Magnification of (c), (f), and (i) = ×12 000; bar = 1 μm.
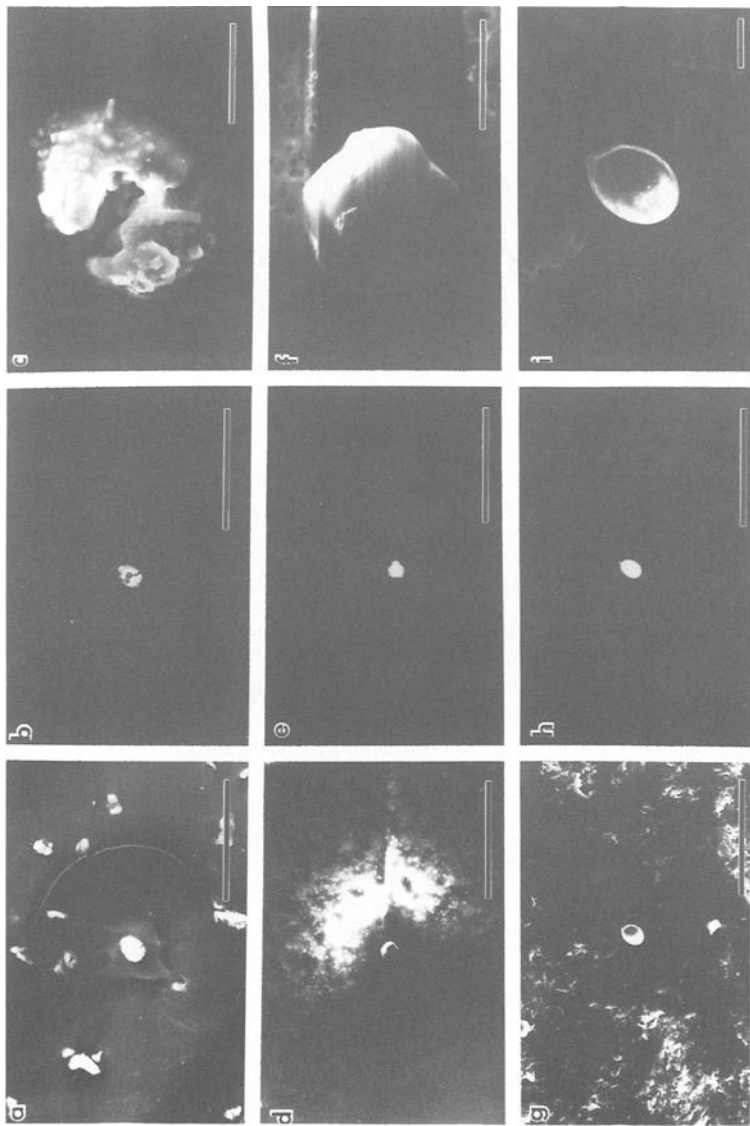
FIG. 6—*Electron micrographs of large GSR particles on various surfaces: (a) secondary electron image of GSR on a tape lift surface; (b) backscatter electron image of GSR particle in (a); (c) enlargement of GSR particle in (a); (d) secondary electron image of GSR on a 0.45-μm Nucleopore filter surface; (e) backscatter electron image of GSR particle in (d); (f) enlargement of GSR particle in (d); (g) secondary electron image of GSR on a glue lift surface; (h) backscatter electron image of GSR particle in (g); (i) enlargement of GSR particle in (g). Magnification for micrographs (a), (b), (d), (e), (g), and (h) = ×300; bar = 100 μm. Magnification in (c) = ×2400; bar = 10 μm. Magnification in (f) = ×2700; bar = 10 μm. Magnification in (i) = ×1200; bar = 10 μm.*
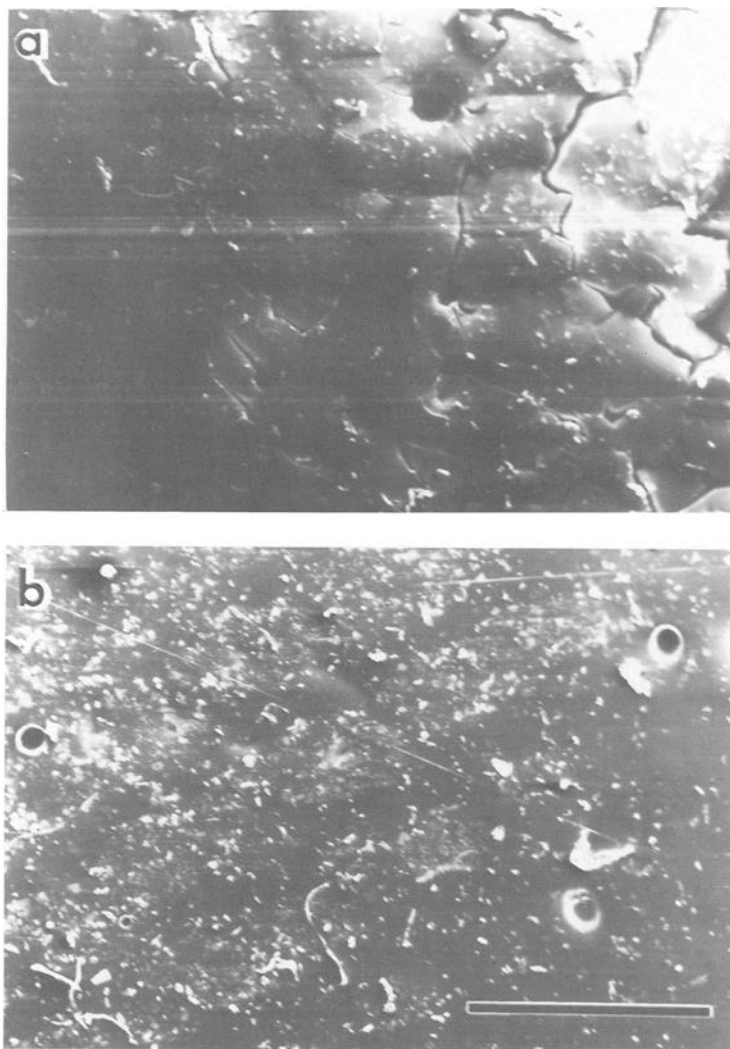
FIG. 7—*Effect of insufficient carbon coating on a tape lift surface*: (a) *secondary electron image of tape lift surface cracking and charging due to beam damage from insufficient carbon coating;* (b) *secondary electron image to the same tape lift stub after a second carbon coating was applied. Magnification of both micrographs* = ×30; *bar* = 1000 μm.

was used. This was measured using a Faraday cup inserted after the final aperture. The tape lift surface was found to be stable under these conditions.

Older energy-dispersive X-ray analysis systems may require higher beam currents when accumulating EDX spectra. An example of this was observed during the present study, where the Tracor Northern TN2000 EDX system required a beam current of approximately 1000 pA to accumulate a spectrum with a 30% dead time for a given GSR particle. Keeping the same detector but changing the hardware to a newer TN5500 EDX system resulted in the accumulation of an EDX spectrum at 500 pA, with a 30% dead time on the same GSR particle.

Perhaps the developers of the glue lift technique were using an older instrument requiring a high-beam current or had insufficient carbon coating on their tape lift samples. This would explain the observed melting of the tape surface and the overall diminished particle counts, as particles "disappeared" from view into the melted surface.

The procedure for sampling the hand with the glue lift disk was different from the hand sampling procedure using the tape lift disk. The developers of the glue lift maintain that the hand should be touched only five times along the thumb, web, and forefinger for sampling the "back" of the shooter's hand [2]. Authors using the tape lift method recommend touching the entire area of thumb, web, and forefinger (about twelve touches) or until the stickiness of the tape is lost [1,3,7,11]. In the present study, perhaps the tape lift picked up more particles because a greater surface area of the hand was sampled.

This possibility was examined by collecting GSR from the hand using a glue lift surface first and dabbing the hand twelve times along the thumb, web, and forefinger. This collection was then followed by a tape lift collection along the same area. The data in Table 5 indicate that, when the surface area sampled was held constant, the glue lift remained a less efficient particle-lifting device in comparison with the tape lift.

Summary and Conclusions

Observed Advantages and Disadvantages of Each Collection Method

Besides examining the collector particle-lifting efficiency for a variety of conditions, such as the number of rounds fired, the temperature, and the storage time, a table of observed advantages and disadvantages for each collection method was developed (Table 6).

The tape lift method for GSR collection has the primary advantage of having an efficient particle-lifting surface, as previously discussed. The tape itself is inexpensive and the collection devices are simple to construct. The adhesive surface was found to be stable, that is, the particle-lifting ability was not decreased significantly under all conditions tested. The tape lift surface gave a good secondary electron image, which is important for photographing particles (Figs. 5a, 5c, 6a, and 6c), especially when using an instrument that has a backscatter detector which does not operate at the normal TV scanning rate (not recommended).

The disadvantages of the tape lift method are few. The surface requires a carbon coat. Depending on the size of the stub used, there is a relatively large surface area to be scanned. Debris collected from the hand may hide GSR particles beneath it.

The chief advantage of the glue lift is that the carbon-coating step may be skipped. The devices are also quite simple to construct. The secondary image was not optimal in the present case because of the roughness of the carbon planchets obtained (Figs. 5g, 5i, 6g, and 6i). Their theoretical advantage is that the surface is less sticky and therefore picks up less debris from the hand.

Unfortunately, the glue lift surface did not pick up much GSR either. It was found to be an inefficient particle-lifting surface. The stability of the glue lift surface to temperature and storage could not be determined because of the minimal number of GSR found on the glue lifts for all treatments tested. The carbon planchets themselves were moderately expensive and were found to be contaminated with interfering tungsten particles. The problem of picking up debris from the hand was reduced with the glue lift surface but not entirely eliminated. The surface area to be searched is the same as in the tape lift, which is relatively large. Typically, 1 h of search time at ×300 magnification resulted in searching approximately 15% of the total surface area of the 15-mm-diameter carbon planchet.

TABLE 6—*Observed advantages and disadvantages of GSR collection methods tested.*

Method	Advantages	Disadvantages
Tape lift	efficient particle lifting surface inexpensive simple to prepare temperature stable stable shelf life at least six weeks good secondary image	requires carbon coat large surface area skin debris may hide particles
Glue lift	requires no carbon coat easy to prepare picks up less debris fair secondary image	inefficient particle lifting surface contaminated with tungsten particles carbon planchets expensive large surface area skin debris may hide particles
Concentration	separate debris from GSR small surface area pre-made collectors and concentrators can be purchased	requires carbon coat collection efficiency variable expensive 2-h processing time subfilter contamination filters stick together poor secondary image

The concentration technique has the potential of separating GSR from debris. Pre-made collection devices can be purchased from Kinderprint Co. Inc. (Martinez, California) or made by the investigator at minimal cost. The total surface area to be searched is reduced to the point where a manual search of the entire filter is possible in less than 1 h, provided there are not a lot of interfering particles of high atomic number.

In the present study, the concentration method was found to give highly variable results. The particle-lifting efficiency and stability of collection surface could not be determined because of several factors contributing to particle loss. As listed in Table 6, those factors included (a) contamination of the subfilter with barium, calcium, and silicon; (b) particle loss due to manipulation of the filter surface; and (c) particle loss due to aspiration of particles trapped in debris or particles trapped in debris on the filter surface.

In addition to the above factors, the concentrators themselves were relatively expensive. In order to achieve a decent secondary electron image, the 0.45-μm Nucleopore filter had to be carbon coated rather heavily. Even under these conditions, distinguishing GSR particles from the background in order to obtain a photograph was difficult at best (Figs. 5d, 5f, 6d, and 6f). An extensive methanol wash must also be used to remove bromoform from the filter to reduce interference in the backscatter mode. Finally, one must consider the additional time required to concentrate the samples. This was approximately 2 h to prepare six samples.

Theoretical Optimal Collection Device

All of the methods examined had their advantages and disadvantages (Table 6). Perhaps the optimal collection device would be one that combines the advantages of all three collection techniques. One would like to have a surface that does not require carbon coating, as in the glue lift technique. That surface would ideally be polished smooth and flat, with no contaminating elements of high atomic number. The surface should be

coated with a substance that has the stickiness of 3M 465 adhesive transfer tape. If the collector was viewed directly in the electron microscope and found to have too much debris on its surface, then one would like to be able to take that same surface and apply the concentration technique to it. The subfilter of the concentrator should be free of contaminating elements of high atomic number. The final filter surface should give a better secondary image than is currently obtained on 0.45-μm Nucleopore filters. The collection device should be relatively inexpensive and stable to conditions of temperature and time (that is, should have a long shelf life).

The proposed collection device could use a polished graphite circular 15-mm wafer as the sample surface. This wafer could be coated with diluted Vistanex adhesive, which is stickier than rubber cement. If, after viewing the sample initially, concentration was deemed appropriate, a concentration device with a larger bore diameter to accommodate the 15-mm wafer could be used. The entire concentration device would then be sonicated to remove and solubilize the Vistanex surface, at which point the wafer could be removed and concentration could proceed as usual. A final filter of the same diameter but of a wider pore spacing, which is more conducive to carbon coating, could be used, as has been suggested by Wallace and Keeley [15].

At the moment, cost is the initial stumbling block. Highly polished graphite planchets are available but are extraordinarily expensive. It would be interesting to obtain some of these highly polished planchets and see to what degree particle-lifting efficiency could be improved over that in the current collection devices.

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