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The Forensic Significance of Glass Composition and Refractive Index Measurements*

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ABSTRACT: The frequency distributions of refractive index and the concentrations of ten elements in 204 glass specimens received as evidence in casework were evaluated. These data were used to estimate the probability of randomly selecting, from a general population, a glass specimen that is indistinguishable in all measured characteristics from a given glass specimen. The probability of two unrelated glass specimens having indistinguishable elemental compositions and refractive indices is calculated and lies between the extremes of 10^{-5} and 10^{-13}. For each of the 20,706 pairwise comparisons of the 204 specimens in this study, the two specimens are analytically distinguishable. The use of highly discriminating analytical methods for the comparison of trace evidence and the corresponding low probability of two unrelated glass specimens being indistinguishable eliminates the need to collect extensive databases for the purpose of making exact probability calculations. The approach used here can be applied to other highly discriminating parameters and trace evidence sample types.

KEYWORDS: forensic science, criminalistics, trace evidence, glass, elemental analysis, refractive index, statistics

A recent topic of discussion in the forensic science community concerns approaches to assigning a level of significance when an item of trace evidence is compared to an object of known origin and the two are found to be indistinguishable. There have been requests by the trier of fact and also some scientists, particularly proponents of a Bayesian approach to evidence interpretation, for the forensic scientist to calculate a quantitative significance measure regarding an opinion as to a common source of compared items. While estimating the likelihood of occurrence of a set of characteristics has been applied to DNA-typing of evidence, the same statistical approaches may not be appropriate for non-biological trace evidence.

One difficulty encountered in the application of statistically-based significance testing to trace evidence derives from the reliance of frequency of occurrence calculations upon databases containing results for the measured parameters. For items of trace evidence, particularly those which are manufactured, the distribution of measured parameters in any location are controlled by the manufacturers' production and shipping practices, as well as the use, destruction, and replacement of the items. Since the databases must be representative of the sample populations in the environments of the crime scene and the subject, and these are unique to each situation, the appropriate databases must be obtained separately for each particular crime scene. In general, this is not practical and may, in fact, be impossible (for example, when specimens from the crime scene cannot be obtained). The difficulty in obtaining an appropriate database is compounded when very discriminating multivariate analytical schemes are used for the examination of items of trace evidence because the database must be large enough to reflect accurately the population distributions of all measured parameters. The large number of samples required to predict accurately probabilities for events with low frequencies of occurrence can neither be representatively acquired nor accurately modeled.

Most prior studies concerning statistical evaluation of trace evidence have used glass evidence as a model, primarily because substantial refractive index data with a high degree of analytical precision have been accumulated over many years, most individual glass objects exhibit reasonably good homogeneity compared to the range across sources, and glass is recovered as transfer evidence following a variety of criminal activities. Initially, significance testing of this data was done using a two stage approach for the comparison of source and transfer evidence consisting of first assessing the analytical parameters used to determine two specimens to be indistinguishable and then estimating the significance of any resulting association (1,2). Most recent publications describe a Bayesian approach that combines the two stages to establish a single measure of likelihood. The literature addressing these questions has recently been summarized in two reviews (3,4). Because refractive index data were readily available, these statistical studies have focused almost entirely upon use of that single parameter. However, anecdotal evidence suggests that improvements in quality control in glass manufacturing have resulted in a decrease in the range of indices observed among glass products (e.g., see [5]). Thus, particularly when considering float glass of recent origin, the value of refractive index for source discrimination may be less than it has been historically. As a result, there is renewed interest in the use of other parameters, particularly elemental concentrations for comparison of glass (5,6).

The concentrations of specific elements have long been used to classify glass as to its production source, relying on the fact that patterns of composition vary from one manufacturer to another depending upon manufacturing processes and raw materials. There have been several studies reported in the scientific literature using elemental composition to address questions of discrimination, that is, distinguishing among similar sources or manufacturing production runs of the same type of glass (7–12). The data reported in these studies have not been considered for calculation of frequency of occurrence statistics for a general glass population because the

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methods of specimen selection did not provide appropriately randomized specimens.

The authors suggest that, in most instances, an exact statistical analysis to assess the significance of nonbiological trace evidence should be avoided, as it must necessarily be based on poorly supported assumptions about each individual crime scene and subject. In place of statistically-based significance testing, emphasis must be placed upon the use of more discriminating analytical methods to enhance the significance of findings of indistinguishability among items of trace evidence. A study of the refractive index and elemental concentrations of glass fragments received as evidence in the FBI Laboratory was made in order to assess the value of significance testing of data from discriminating analytical methods. The variables considered in this study are refractive index (n_D) and the concentrations of aluminum (Al), barium (Ba), calcium (Ca), iron (Fe), magnesium (Mg), manganese (Mn), sodium (Na), strontium (Sr), titanium (Ti), and zirconium (Zr) in glass, but the approach presented here can be used for any quantitative or categorical variables and for any type of associative evidence.

Details

Sample Description

Most of the glass specimens used for this study were received as evidence by the FBI Laboratory in cases submitted during the period 1990 to 1996. A few additional specimens were acquired during manufacturing plant visits, limited product surveys, or as comparison samples in individual cases. According to the protocol for glass examination in the FBI Laboratory, elemental concentrations are determined for those specimens that are indistinguishable by color, refractive index, dispersion, and meet other criteria, such as minimum fragment size and legal permission to consume the sample. In some instances, two or more specimens from the same case having differing values of n_D may be analyzed for elemental content. Three fragments from each specimen are analyzed separately to provide a measure of variability for each analytical parameter. Where possible, the triplicate samples are selected from separate fragments taken from the broken object, but because of the nature of physical evidence, in some instances triplicate samples must be taken from a single fragment. It is the FBI Laboratory's practice to compile the results of analysis as part of their experience base. At the time this study was undertaken, the compositional database consisted of 1545 evidentiary samples and approximately 600 samples of standard reference glasses, which were analyzed as internal quality checks with each case.

In order to remove sampling bias in this study, the full database was reduced in size to make a test database according to the following procedure. First, the standard reference glasses were deleted from the full database. Next, those specimens for which data had not been obtained for three separate fragments were withdrawn from the test population. For each case, a specimen was selected and its data placed into the test database. The results for every other specimen in that case were compared to the selected data. If a second specimen was analytically distinguishable in one or more parameters, it was considered to represent a second source of glass, and it was also selected for the test population. This process was continued for all specimens, comparing each one with all previously selected specimens from that case, until a group was selected representing all of the distinguishable sources of glass received in each particular case. This process was repeated for each case in the original database and all selected data were compiled together into the test database. Since the test database does not include data from two indistinguishable specimens from the same case, it represents an unbiased sample, at least in the sense that each glass specimen is represented only one time. The resulting database used for this study consists of 204 specimens, each analyzed in triplicate, or 612 analytical samples.

Methods of Analysis

All samples in the test database were analyzed using well-established methods in use in the FBI Laboratory for examination of evidentiary specimens. Refractive index was determined by the Emmons double variation method (13). The concentrations of the ten elements, Al, Ba, Ca, Fe, Mg, Mn, Na, Sr, Ti, and Zr, were determined by the method of inductively coupled plasma-atomic emission spectrometry (ICP-AES). Details of the cleaning, dissolution, and analysis procedures are presented elsewhere (11,12). One or more well-characterized standard reference glasses (NIST, Gaithersburg, MD) were prepared and analyzed as samples with each case. The accuracy of the results for these reference glasses is verified for each element before sample results for that case are added to the FBI Laboratory's database. This long-term check on the accuracy of the results allows the database to be used for studying the distribution of the measured parameters in the glass population represented by the database.

Discussion of Results

Analytical Precision

The precision associated with any quantitative analytical measurement is limited by the random analytical uncertainties in weighing the sample, dissolution, and instrumental variations, and by the heterogeneity of the glass specimen. The total precision is a limiting factor in the ability of an analytical method to discriminate among specimens. For the concentration of each element in each specimen in this study, a precision was calculated based on the standard deviation of the triplicate determinations. These results, expressed as percent relative standard deviation are shown as functions of concentration in Fig. 1. Most elements occur in ranges of concentrations from close to the limit of detection to levels where the analytical precision is better than a few percent. In general, the precision of the analytical results improves (decreases) with increasing concentration up to a point where it becomes constant. In most glasses, Ca and Na and, in some glasses, Fe and Mg are controlled by the manufacturers at readily measured concentrations. Therefore, the majority of the measured precisions for these elements are at the flatter portion of the precision distribution curves and are nearly constant across the concentration range. Since the total precision limits the discrimination capability, the degree of difference that must exist between two glass specimens to distinguish them from each other is a function of concentration for each element according to the relationships shown in Fig. 1.

Element Distributions

Histograms with bins whose widths vary as functions of analytical precision were constructed to display the frequency distributions of element concentrations. This was done as follows. For each element, the individual data points shown in Fig. 1 were plotted into a smooth curve. At appropriate concentration increments of each element, histogram bins were made using the relative standard deviations from the smoothed curve, which were converted to standard deviations by multiplying each by the concentration at the respective bin center. The width of each bin was set at 12 times the

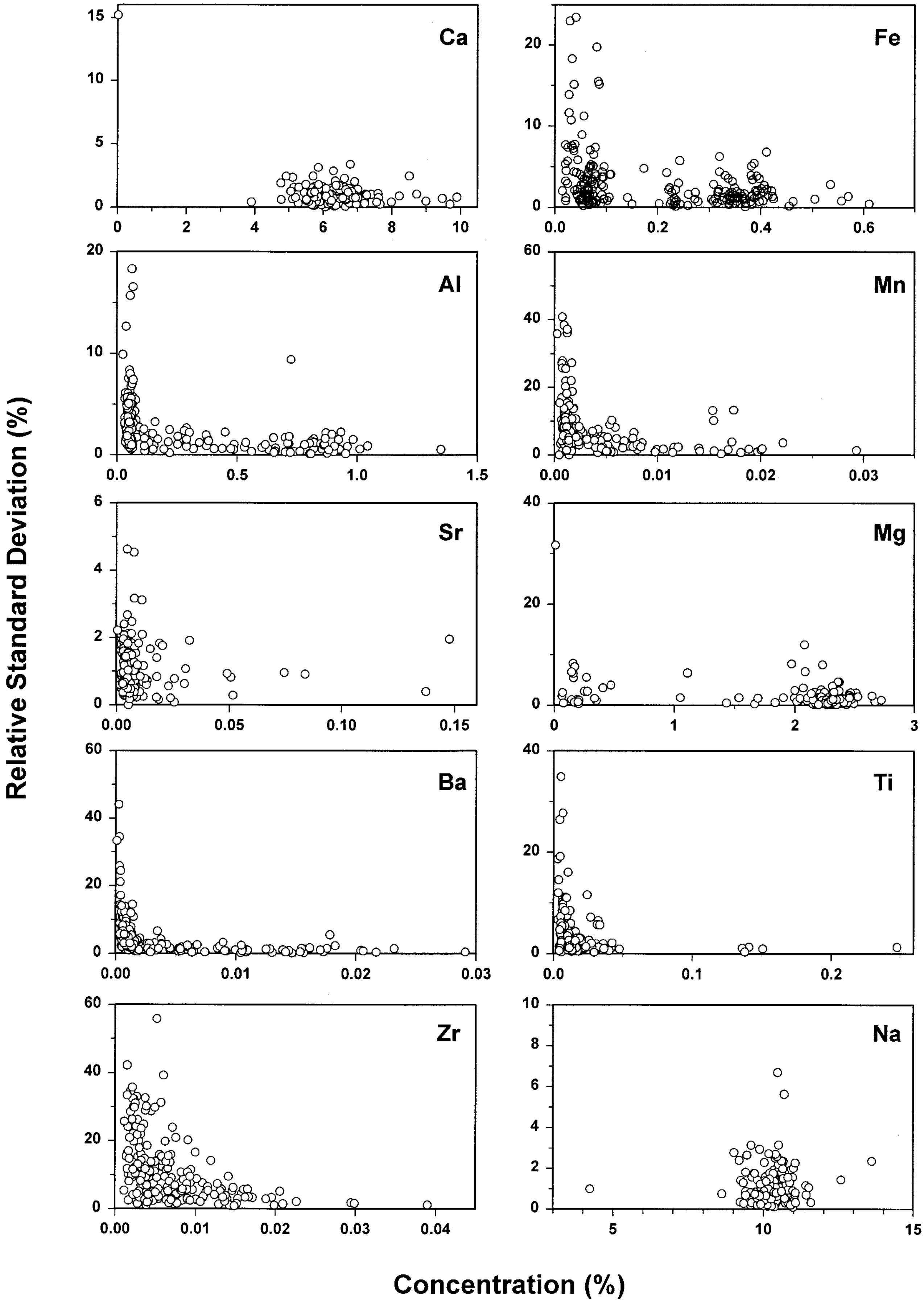

FIG. 1—*Relative standard deviations (RSDs) of triplicate samples of each glass specimen as a function of element concentrations. Concentrations are in element percent by weight; RSDs are in percent.*

standard deviation (12σ). Starting from the lowest measured concentrations, bins were formed side-by-side over the observed concentration ranges. The distribution of specimens among the histogram bins was then tabulated. Specimens whose concentrations of a particular element are below the detection limit are included in the lowest concentration bin for that element. Histograms of specimen frequencies as a function of concentration constructed using these 12σ bin widths are shown for each element in Fig. 2. Note that bin widths are shown in standard deviation units and, therefore, are narrower at the lower end of the concentration range for

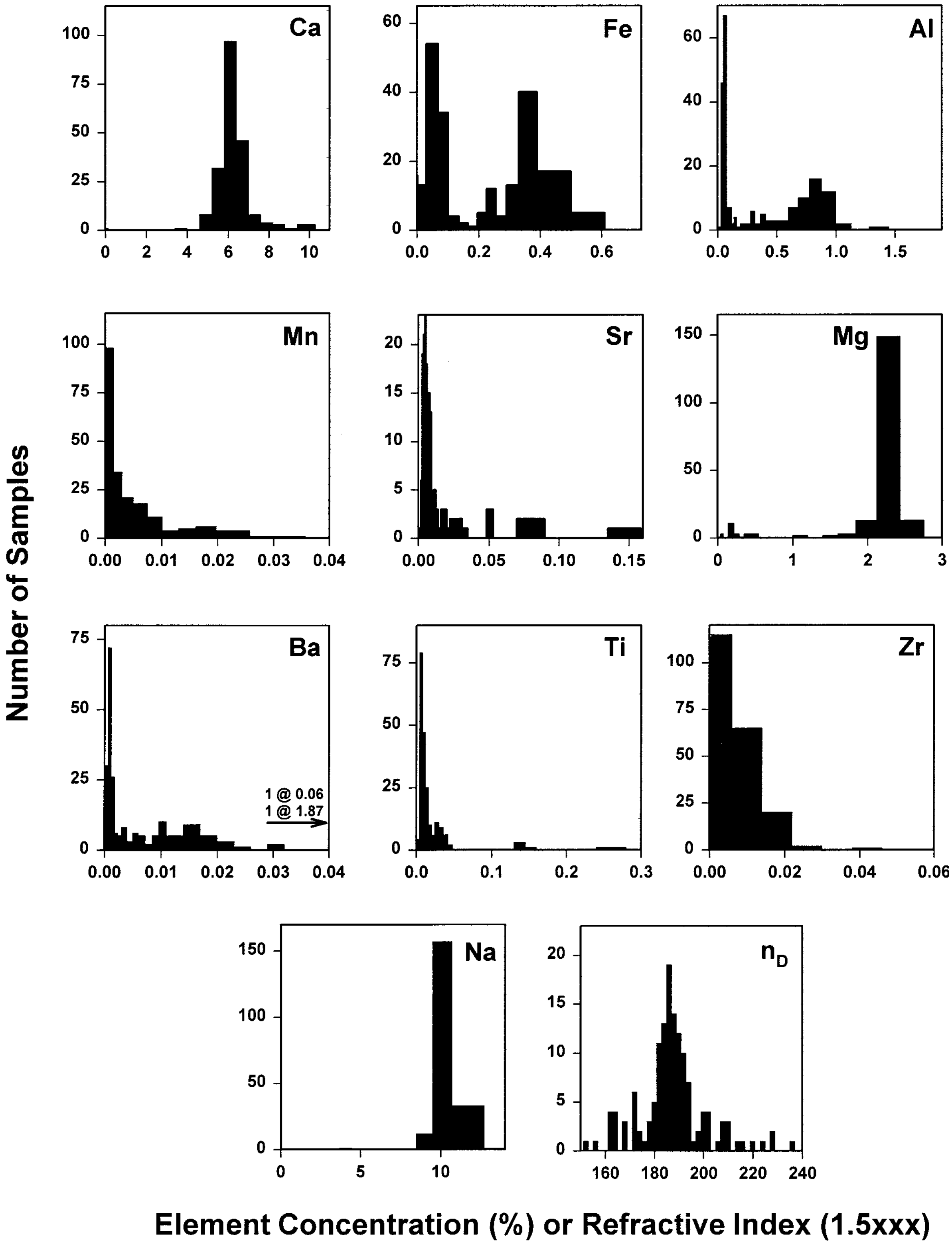

FIG. 2—*Frequency of occurrence distributions of glass specimens among bins for 10 elements and refractive index.*

each element. Had Fig. 2 been drawn using bin widths based on relative standard deviations, then bin widths would have appeared wider at the lower end of the concentration ranges, reflecting the improvement in analytical precision as concentrations increase into good analytical working ranges.

The widths of the histogram bins shown in Fig. 2 were selected as 12σ in order to be conservative when assessing the discrimination capability of the analytical method. The use of wide bins results in an underestimation of the discrimination capability, a desirable result as it prevents overstating the value of an analytical method. In case situations, where results for individual items of evidence are compared to each other, narrower overlap criteria would be used to decrease the likelihood of making false associations.

A histogram of refractive index distributions is also shown in Fig. 2. For this variable, the bin width was not based upon standard deviations of triplicate measurements. Rather, the bin widths for refractive index are a constant 0.0002 units, a cut-off value commonly used in forensic laboratories for discriminating between two sources of flat glass (15). The refractive index and element concentration ranges for all specimens are summarized in Table 1. Also included in Table 1 are the number of specimens, number of bins, and number of specimens in the most populated bin for each measured parameter. Refractive index data are included for only 141 of the glasses used in this study, because the measured refractive index values for the other samples were not recorded during compilation of the FBI Laboratory's glass compositional database. All of the elements except Mn and Zr were determined in each of the 612 samples. The Mn data for two specimens and the Zr data for one specimen are not included in the database summary, because this data was not included in the FBI Laboratory's glass compositional database from which the test database was derived.

Information Content of the Analytical Method

Information content is a term used in this report as a measure of the discrimination capability of the total analytical method. The information content is defined as the total number of possible analytically distinguishable results or, in this case, the total number of bins in eleven-dimensional space. In this study, in order to provide a conservative estimate of the information content, it is calculated using only the ranges of compositions that are approximately continuously filled. For example, for Mg there are seven bins in the $0.01 - 0.5\%$ range and seven bins in the $1.0 - 2.8\%$ range, making a total of 14 bins. Any unfilled bins that could occur at Mg concentrations between 0.5% and 1.0% are omitted from the calculation of information content. The seven elements with discontinuous bin distributions are so indicated in Table 1. The information content of the method is calculated by multiplying together the number of bins (n_x) for each measured parameter:

$$\textit{Information Content} = n_{n_D} \times n_{Ca} \times n_{Fe} \times n_{Al} \times n_{Mn} \times n_{Sr} \times n_{Mg} \times n_{Ba} \times n_{Ti} \times n_{Zr} \times n_{Na}$$

Substituting the values from Table 1,

$$\textit{Information Content} = 43 \times 12 \times 13 \times 23 \times 10 \times 29 \times 14 \times 20 \times 13 \times 6 \times 5 = 4.89 \times 10^{12}$$

At 4.89×10^{12}, the information content or total number of distinguishable bin combinations is extremely large. As a point of reference, the two dimensional land area of the earth is approximately 1.5×10^{14} m^2. Therefore, the capability of refractive index and ICP-AES data for discriminating among different sources of glass is so great that if the earth's land surface could be covered by sheets of glass each 30 m^2 in area, it would be possible to have each one represented by a bin combination that is analytically distinguishable from all of the others. Of course, all of these compositions do not actually exist; this illustration merely shows that the analytical methods used here offer the potential for excellent source-discrimination capability. Based only on analytical considerations and omitting glass product distributions for the moment, the probability of a given subject specimen randomly falling into a given bin combination (such as the one defined by a broken object at a crime scene) is the inverse of the information content, or 2.05×10^{-13}.

It is again noted that the number of bins selected here is quite conservative. Bin widths of less than 12σ could be used to construct the histograms. For example, if the information content were calculated using bin widths of 4σ ($\pm 2\sigma$) for each element, then it would be increased by a factor of 3^{10} to a value of 2.9×10^{17}. Another factor that could increase the information content for any given analysis of evidentiary samples is that the precision may be better than the mean precisions shown in Fig. 1. Finally, samples of other compositions of glass such as headlamps, tempered cookware, and optical glass may not be represented in this database. Inclusion of these products would greatly expand the ranges for refractive index and several elements and correspondingly increase the number of bins. Increasing the range of compositions or decreasing the bin widths, or both, would result in significant reductions in the already low calculated probability of a randomly selected glass specimen falling into a given bin combination.

The very large number of possible bin combinations is the result of multiplication of eleven factors together and demonstrates the

TABLE 1—*Summary of binning results. Range of values in percent by mass, except for n_D, which is unitless.*

Variable	Number of Measurements	Range of Values	Number of Bins	Maximum Bin Occupancy
n_D	141	1.5152–1.5237	43	19
Ca	204	0.0079–9.90	12(d)	97
Fe	204	0.015–0.611	13	54
Al	204	0.024–1.35	23	67
Mn	202	0.00026–0.0293	10	98
Sr	204	0.00045–0.148	29(d)	24
Mg	204	0.0098–2.72	14(d)	149
Ba	204	0.00003–1.87	20(d)	72
Ti	204	0.003–0.248	13(d)	79
Zr	203	0.00001–0.0390	6(d)	115
Na	204	4.23–13.61	5(d)	157

(d) indicates discontinuous bin distributions, i.e., skips in concentrations between bins.

TABLE 2—*Pearson coefficient of linear correlations (r) between pairs of variables for 204 glass specimens.*

	n_D	Ca	Fe	Al	Mn	Sr	Mg	Ba	Ti	Zr
Ca	0.439									
Fe	0.150	−0.175								
Al	−0.269	−0.111	−0.341							
Mn	−0.347	−0.017	−0.138	0.601						
Sr	−0.054	0.032	−0.231	0.138	−0.072					
Mg	−0.064	−0.405	0.359	−0.457	−0.135	−0.194				
Ba	−0.054	−0.192	−0.086	0.263	0.042	0.076	0.006			
Ti	0.115	−0.023	0.257	0.129	0.241	−0.027	−0.005	0.002		
Zr	0.147	−0.147	0.045	0.012	−0.111	0.054	−0.084	0.197	0.184	
Na	0.035	0.299	−0.036	−0.246	−0.227	0.010	−0.003	0.117	−0.090	0.032

superb discriminating power when many analytical variables are measured. The information content is a useful measure of the potential capability of an analytical method. Information content can be applied to any variable used for evidence comparison, whether it is continuous or categorical. However, a limitation to the information content is that it does not give any indication of how actual glass specimens are distributed among the bin combinations or into forensically meaningful groups. In order for the true probability of selecting any given composition profile from a random population to be the same as the probability calculated from the information content approach, two conditions must be met. These conditions are that the measured variables are mutually independent and that all bin combinations are equally filled. Each of these conditions will be considered separately.

Independence of Variables

The Pearson product moment correlation statistics for linear correlation of all combinations of pairs of variables for the 204 specimens are shown in Table 2. Using $r > 0.5$ as a criterion, the sole pair of elements that exhibit a significant linear correlation is Al and Mn. The concentrations of all other element pairs in this study can be considered to be independent. In fact, the magnitudes of the correlation coefficients shown in Table 2 are extremely low, most less than 0.2. Although the correlation coefficient between Al and Mn concentrations is 0.601, this correlation is weak, as shown in Fig. 3. The distribution of data points in both dimensions indicates that the combination of Al and Mn provides more discrimination than either one alone.

The calculation of information content does not require the variables to be independent, as it is merely the total number of distinguishable element combinations. However, it is more meaningful forensically to calculate the probability of two randomly selected glass specimens being indistinguishable based on the frequency of occurrence of specific compositions. Such probability calculations do require independence of variables. For the calculations that follow, it will be assumed that all variables, including Al and Mn, are independent. The weak correlation between Al and Mn concentrations is balanced by the conservative selection of bin widths.

Most Common Composition Calculation

The information content can be considered the maximum discrimination capability that the analytical method will allow. At the opposite extreme, the most common composition is defined to assess the probability of randomly selecting a specimen whose composition is the most likely to occur. The most frequently occurring glass composition is the one for a sample having that value for each variable that corresponds to the bin in Fig. 2 with the greatest occupancy. Since all variables are independent, the probability for the most common composition is the product of the fraction of total specimens in the most filled bin for each element:

$$Probability = f^{n_D} \times f_{Ca} \times f_{Fe} \times f_{Al} \times f_{Mn} \times f_{Sr} \times f_{Mg} \times f_{Ba} \times f_{Ti} \times f_{Zr} \times f_{Na}$$

where f_x = the fraction of specimens in the most filled bin for variable x.

Substituting the values from Table 1,

$$Probability = \frac{19}{141} \times \frac{97}{204} \times \frac{54}{204} \times \frac{67}{204} \times \frac{98}{202} \times \frac{24}{204} \times \frac{149}{204} \times \frac{72}{204} \times \frac{79}{204} \times \frac{115}{203} \times \frac{157}{204} = 1.38 \times 10^{-5}$$

This probability value of 1.38×10^{-5} is quite small, although it is much larger than the probability value of 2.05×10^{-13} calculated using the information content approach. If the values for Mn are deleted from the calculation to eliminate any effect due to the correlation between Mn and Al, the probability increases to 2.84×10^{-5}. It is interesting to note that none of the 204 sources of glass (specimens) in this study has a composition that falls into the most common bins for all of the measured variables. The lack of a specimen having the most common composition in a subpopulation containing 204 members is not sufficient to prove the accuracy of a probability figure of 10^{-5}. However, it does at least support the idea that the likelihood of selecting a glass fragment having even the most common composition in a random collection of glass is quite small.

The two probabilities calculated in this study may be thought of as two extremes, in the sense that the probability of obtaining any given composition of glass drawn randomly from a larger popula-

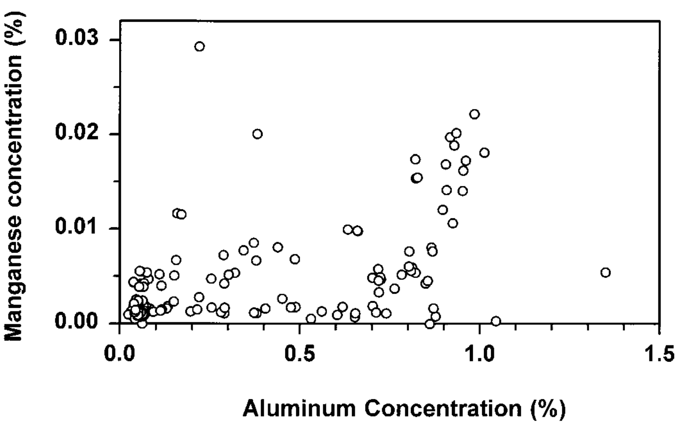

FIG. 3—*Distribution of glass specimens among Al and Mn bins.*

tion represented by this database will lie between them. The results of the analysis of an evidentiary specimen can be compared to the database using a floating-bin approach; that is, bins centered on the specimen means and widths based on the measured standard deviations. Using data for evidentiary specimens received by the FBI Laboratory, the calculated probability of a random match using 12σ bin widths is typically in the range of 10^{-8} to 10^{-10}. The values calculated in this study are, at best, rough approximations, since addition of more samples to the database could increase the information content and either increase or decrease the most common composition probability depending upon their composition.

A useful measure for assessing the likelihood of random matches of glass and to test the reasonableness of the calculated probabilities is to make pairwise comparisons of the specimens in the test database. There are 20,706 pairwise comparisons that can be made among the 204 specimens in the test database. All of these pairs are readily distinguishable by one or more analytical variables using the previously reported discrimination criteria (12). Again, while 20,706 comparisons are not enough to completely validate the very small probabilities calculated in this study, the lack of a pair of indiscriminable specimens supports the belief that the probability of the coincidental occurrence of two or more indistinguishable sources in a randomly selected glass population is extremely low.

Conclusions

The two approaches described here to define the general range for the probability of random occurrence of indistinguishable glass fragments place this value between 10^{-13} and 10^{-5}. These low values indicate that element concentrations and refractive index provide an extremely high source-discrimination capability among glass fragments. In the evaluation of evidentiary glass, in most instances, a high degree of confidence can be placed in a conclusion that two indistinguishable glass fragments came from a common source. However, the forensic scientist must be careful when pressed to assign a significance measure to a conclusion of indistinguishability when dealing with manufacturer-controlled distributions of specimens. When the frequency of occurrence of a particular combination of variables is low, then the calculated probabilities are necessarily low. For most, if not all, nonbiological items of trace evidence, it is not possible to appropriately sample or otherwise determine a frequency distribution for each variable in the population relevant to a given crime scene or subject. Therefore, valid probability calculations cannot be made. The best uses of databases obtained using highly discriminating analytical techniques are to compare methods of analysis and to demonstrate that the chance of random matches is extremely small. Once this is established, there is little imperative for continued collection of databases or in attempting to make precise probability calculations. In fact, to do so would require making assumptions concerning distributions of the specific commodity in the crime scene or subject environments that may be difficult to support. Databases are still required, however, for purposes of continued monitoring of changes in manufacturing technology that could affect the discrimination capabilities of the analytical method and for classification, or placing of a specimen into a product-use class (10,11).

It is interesting to consider the effects of discrimination capability on results calculated using the Bayesian approach, as the probability of the evidence given that the subject is not associated with the crime scene is a term required in calculation of a likelihood ratio. One factor in calculating the value of this probability is the frequency of occurrence in the appropriate sample population of evidence having the given values of measured parameters (3,4). Though this is not calculable in a statistically valid sense for the analytical methods used in this study, it is clearly an extremely small number, between the limits of 10^{-13} to 10^{-5}. Since this value appears in the denominator of the likelihood ratio, a very large likelihood ratio will always result in those cases where elemental analysis of glass fragments is consistent with a common source. The impact of other less extreme factors, such as transfer and persistence considerations, will be minimal in comparison. For example, using a frequency of occurrence value of 0.03 for refractive index alone, Aitken (4) calculates likelihood ratios on the order of $10^2 - 10^6$ for various scenarios. Substituting a typical glass compositional frequency value of 10^{-8} in the denominator in place of 0.03 raises his likelihood ratios to the 10^8 to 10^{12} range. These very high likelihood ratios indicate that glass will nearly always be a significant piece of evidence when compositional measurements are considered. The advantage of using highly discriminating analytical techniques also lessens the importance of the often subjective transfer, persistence, and population distribution assumptions, which are required by the Bayesian model.

The approaches in this paper were applied to glass evidence because robust data are available for that material. However, the same approaches can be used for other types of trace evidence. For example, if measured parameters are such that only five distinguishable categories (bins) can be determined for each of ten measured parameters, then the total number of distinguishable combinations is 5^{10} or nearly 10,000,000. This is a reflection of the power of multiplying frequencies together and can yield good frequency of occurrence statistics when variables are independent, the sample population is distributed evenly across the categories, and the database is a reliable representation of the relevant population.

The forensic scientist should use the most discriminating technique available in the examination of glass or other form of trace evidence because it is the most effective means of both avoiding false associations and excluding two similar, but separate, sources. It is in the best interest of the court for the scientist to use the most discriminating analytical technique even if this means that exact probability figures for a conclusion cannot be calculated. In cases where the analytical discrimination is very good, as in compositional measurements of glass, factors such as manufacturer distribution of products and age and breakage of glass objects in the crime scene and suspect environments are more significant than the probability of two randomly selected sources from a large glass population having coincidentally indistinguishable characteristics. These factors can either be determined by standard investigative techniques or they involve everyday experiences of the nonscientist. As a result, their significance can be readily weighed by the trier of fact without resorting to statistical calculations.

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