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AN ACTUAL CASE OF GLASS DEBRIS IDENTIFICATION BY NAA IN AN AUTOMOBILE ACCIDENT

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The origin of glass fragments, found during an investigation of an accident between two cars, was established by neutron activation analysis. It was possible to ascribe responsibility for the accident based on that identification together with the other general evidence usually considered in such cases. Beside γ-spectrometry, specific activities were also used as criteria for the examination of samples.

In a collision between a truck and a jeep, the left part of the truck carriage hit the jeep, breaking the glass windscreen of the jeep. Glass debris was scattered and some found its way on to carriage of the truck. The driver of the jeep was killed in the accident and the truck was driven away. Two days later the truck was found but the driver denied having been involved in the accident, neither himself nor any other person driving the truck.

The truck driver apparently was unaware that glass debris had reached his vehicle; or, if he was, he did not make a thorogh job of removing the debris and some pieces were found in the truck carriage.

The debris, and that collected from the broken glass windscreen of the jeep were submitted to activation analysis. From each sample, fifteen pieces each of about 30 milligrams were

taken, cleaned with a hot 1:1 nitric acid solution, demineralized water and irradiated for ten seconds in a thermal neutron flux of 5×10^{12} $n.cm^{-2}$ sec^{-1}. Two other sets of ten pieces from each glass sample were irradiated for five days, cleaned with hot 1:1 nitric acid solution and demineralized water.

γ-spectra of the two sets of samples /long and short irradiation periods/ were obtained using a 400 channel analyzer coupled to a NaI/Tl/ 3"x3" well type scintillator. The samples irradiated for a short period were counted at a cooling time of 8 minutes and the samples irradiated for a long period at a cooling time of 10 days. Total specific γ-activities were also determined for the twenty samples irradiated for a long period. Specific β-activities could not be used owing to large variations in specimens of the same sample; such variations of specific activities among the specimens taken from the same sample were due to β self-absorption effects since the various pieces were not of the same size although they had approximately the same mass. Also, a reproducible β counting geometry was difficult to achieve owing to differences in shape of the replicate samples.

The various γ-spectra were compared for the two sets of samples /samples with the same irradiation and cooling times/ by the peak-ratio technique /areas/ taking the 1.37 MeV ^{24}Na peak as a reference or normalizing peak. A statistical treatment was given in order to obtain a two-sided significance t-test for comparison of means at the 0.05 significance level.

This same peak-ratio technique has been applied by others in the case of comparison of hair, paints and also of glass. Jervis[1] had already called attention to the fact that comparisons

of a number of intensity ratios for resolved photopeaks in γ-ray spectra can be made, and possibly without even deducing in every case the exact identity of the several nuclides contributing to the γ-spectrum. Bryan et al.[2] made comparisons via the pulse--height spectra obtained from identical irradiations, decay times, and counting conditions without calculation of amounts of concentrations or use of external standards. Comparison of automobile glasses has been made by Jervis[3] who used itensity ratios of γ-rays at the same energies for the various samples or, alternatively, intensity ratios taking the same reference peak in

TABLE 1

Mean values for the ratios X of listed peak-areas to the ^{24}Na reference peak for the spectra of samples from short irradiation

	Glass				
	1		2		
Peak MeV	$\bar{x}_1$ *	V_1 **	$\bar{x}_2$ *	V_2 **	t_{exp}
0.20	0.486	0.000071	0.481	0.000044	1.803
0.84	0.494	0.000094	0.495	0.000035	0.341
1.00	0.373	0.000069	0.370	0.000049	1.069
1.78	0.561	0.000212	0.565	0.000108	0.865

* Number of samples taken from each glass, n = 15.
Degrees of freedom D.F. = 2n - 2 = 28

$$t_{exp} = \frac{\bar{x}_1 - \bar{x}_2}{\left[2\left(V_1 + V_2\right)\right]^{1/2}} \; /2n/^{1/2} \qquad \text{/ref. 4/}$$

t = 2.048 /Fisher and Yates' Tables/

** : Variances

each sample to be compared, as an internal reference standard, as is done in this paper.

The results X /mean values/ of the ratios for the area of four peaks in the spectra of samples from short irradiations, to the 1.37 MeV ^{24}Na peak are presented in Table 1. Fifteen replicate samples from each glass were irradiated to obtain the mean values and variances shown in this Table. Table 2 shows the results for the spectra of samples from long irradiations, in which a comparison was made among the ratios of eight long-lived peaks to the reference peak of ^{24}Na; ten replicate samples from each glass, were used to give the data in Table 2.

TABLE 2

Mean values for the ratios X of listed peak-area to the ^{24}Na reference peak for the spectra of samples from long irradiation

	Glass				
	1		2		
Peak MeV	$\bar{x}_1$ *	V_1	$\bar{x}_2$ *	V_2	t_{exp}
0.15	7.051	0.238638	7.110	0.228194	0.417
0.30	1.701	0.007358	1.706	0.007089	0.132
0.35	1.147	0.003364	1.140	0.002464	0.290
0.55	5.000	0.036224	5.042	0.027601	0.803
0.85	0.860	0.001265	0.859	0.001160	0.090
1.10	0.664	0.001105	0.660	0.000884	0.283
1.25	0.682	0.001126	0.678	0.001204	0.262
2.00	0.1823	0.000031	0.1820	0.000026	0.081

* Number of samples for each glass: 10
D.F. = 18
t /0.05/ = 2.101

In both tables it can be seen that t_{exp} is always smaller than t values taken from Fisher and Yates' Tables[5] at the 0.05 significance level /two-sided criterion/. For this level Fisher and Yates' values for t are 2.048 for 28 degrees of freedom and 2.101 for 18 degrees of freedom, which are larger than the respective t_{exp} values reported in Table 1 and 2 giving a non-significant difference between the means for glass 1 and 2; this would indicate a common origin for both glasses on the basis of examination of four plus eight, i.e. a total of twelve peak-ratio comparisons.

Samples taken from four other cars were also compared in the same way /t-test/ with glasses 1 and 2 and in all these cases the differences were highly significant.

Specific activities were also used as a criterion for comparison of samples. A significance F-test and Scheffés' test[6] were used. Ten samples taken from each glass, 1 and 2, and ten samples taken from four other cars were irradiated for five days, allowed to cool for ten days, and then total specific γ--activities were determined. Flux monitors were used to correct for flux variations since it was not possible to irradiate the sixty samples /about 30 mg each/ at the same time. Table 3 presents the specific activities for each of the ten samples taken from the six cars. Table 4 presents the data required for the F-test. An F value of 2487 is highly significant /as compared with the F value from Fisher and Yates' Tables for 5 and 54 degrees of freedom at the level of 0.05, i.e. $F_{5.54} = 2.37$/ and so the null hypothesis of equality of the six glass samples must be rejected.

TABLE 3

Specific activities /cpm/mg/ for samples taken from six cars

	Glass					
	1	2	3	4	5	6
	1173	1181	667	1441	784	595
	1197	1174	675	1420	779	604
	1187	1239	683	1372	776	583
	1170	1157	684	1374	759	593
	1135	1194	664	1398	766	610
	1172	1150	692	1376	771	603
	1134	1153	678	1341	747	580
	1166	1197	677	1369	758	607
	1153	1132	653	1348	772	602
	1186	1141	690	1374	771	597
Mean	1167.3	1171.8	676.3	1381.3	768.3	597.6

TABLE 4

Data for calculation of F for Table 3 values

Source of variation	Sum of squares	Degrees of freedom	Mean square	F
Between columns	5138935	5	1027787	$\frac{1027787}{413.26}$ =
Residual	22316	54	413.26	
Total	5161251	59		=2487.02

However, the F-test shows only that among the six samples at least one of them is different from the others but it does not indicate in which pair of the six samples the values are the same. A Scheffes' test can establish that and for this, the Δ value, defined as follows[6]

$$\Delta = \bar{x}_i - \bar{x}_j = \left[MS_r\left(1/n_1 + 1/n_2\right)\left(n_c - 1\right)F_{5.54}\right]^{1/2}$$

must be calculated. The meaning of the new symbols are: MS_r, the mean square of residual; n_c, number of columns /Table 3/; n_1, number of samples 1; n_2, number of samples 2; $F_{5.54}$, F-value for 5 degrees of freedom /columns/ and 54 degrees of freedom /residual/. Taking $n_c = 6$, $n_1 = n_2 = 10$, MS_r /from Table 4/ = 413.26 and $F_{5.54}$ /Fisher and Yates' Tables/ at the 0.05 level = 2.37 one gets $\Delta = 31$. This signifies that all mean values whose differences are smaller than or equal to 31 establish that the samples are the same at the 0.05 significance level. For the values reported in Table 3 this is valid only for glasses 1 and 2.

DISCUSSION

Perkons[7], Bate and Dyer[8] have already called attention to an argument against the use of peak-ratio as a criterion for individualization of materials /hairs, specifically/ in that a peak-ratio value of 1/2 would still be the same whether the individual peak values were, for instance, 10 and 20 or 100 and 200.

However, in the case of hair individualization, specific activities cannot be precisely determined especially in the case of a single hair. There is no doubt that in the case of glass,

or any other material, the same peak-ratio can be found for individual peak values of 10 and 20, and of 100 and 200. This, however, would require an increase in all peaks, and in the concentration of all elements responsible for those peaks by the same factor of 10, which is not likely to happen. Even if it does happen, that is, a 10-times increase in the concentration of all elements in one sample, specific activities of the two samples would then be different indicating that the samples are, indeed, different.

Another criticism of the peak-ratio technique is that a comparison of, for instance, ten peaks might actually be a comparison of less than ten common elements, since one single element may be responsible for more than one peak in the same spectrum. However, if the peak-ratio technique, which is an internal standard technique, is applied using various irradiation and cooling times and if the additional information given by the determination of specific activities, is also used, and if a large number of samples is available for examination /as in the present case/, these will compensate for the smaller amount of information the peak-ratio method may give compared with an actual analysis of elements.

No radiochemical separations were carried out attempting to identify all radioisotopes responsible for the various peaks in the γ-spectra. However, a combination of irradiation-time, photopeak-energy and decay-rate information supported assignments of the peaks in the spectra of samples from short and from long irradiation periods /peaks at 0.20, 0.84, 1.00 and 1.78 MeV for the spectra of samples from short irradiation; and 0.15, 0.30,

0.35, 0.55, 0.85, 1.10, 1.25 and 2.00 MeV for the spectra of samples from long irradiation/ to the following nuclides: ^{27}Mg, ^{180m}Hf, ^{56}Mn, ^{28}Al, ^{181}Hf, ^{59}Fe, ^{51}Cr, ^{76}As, ^{134}Cs, ^{46}Sc, ^{65}Zn, ^{24}Na. No definite conclusions regarding the presence of other nuclides such as ^{66}Cu and ^{182}Ta could be reached although indications exist that they may also be present. It is interesting to note that many of these resolved γ-photopeaks as well as the assigned nuclides for the spectra of glass samples from long irradiation, are the same as those reported by Jervis when making comparisons of automobile glasses[3].

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SUMÁRIO

A origem de fragmentos de vidros, encontrados quando da investigação de acidente automobilístico que resultou em morte de uma pessoa, foi estabelecida pela técnica de ativação por neutrons. As amostras irradiadas foram comparadas atraves dos espectros de raios gama, obtidos em analisadores multicanais, bem como pela medida das atividades específicas das amostras.