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An Improved Density Gradient Technique and Its Application to Paper and Cloth Ash

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AND ITS APPLICATION TO PAPER AND CLOTH ASH*

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The density gradient has been shown to be a valuable adjunct to
the identification of solid materials (1) in that it allows rapid and
sensitive comparison of the density of materials and particularly the
distribution of density of the individual particles in a heterogenous
system. It was applied to identification of glass (2), soil (3), mis-
cellaneous mineral materials (4), and hair (5). It has been found
most effective in the solution of criminal cases involving the types of
evidence indicated. It has not been applied to solution of cases involving
fires in which organic material is destroyed and only mineral constituents
remain in the form of ash. Clearly, these should be susceptible to the
same type of investigation method that is applicable to other mineral
materials.

Paper is one of those items that is very frequently burned in order to
destroy evidence in the form of writing on it. Sometimes the charred
paper is recovered, and by application of various procedures the writ-
ing or other marking may be deciphered. When this is possible, it yields
better evidence than the ash as such could be expected to do. When the
destruction has progressed to the point that only ash remains, it may
still be desirable to determine if possible whether a particular kind of
paper was burned. This should be possible if samples of the paper in
question can be recovered and ashed.

Similar considerations apply to cloth that is burned, though in this
instance, the value of the evidence might well be less than that of
burned paper. In at least two cases known to the authors, burned
cloth was involved and required identification as to the source of the
cloth material. That cloth has a certain degree of individuality follows
from the fact that the mineral constituents of the raw materials are far
from constant, and particularly that a variety of treatments are utilized
which may alter and often greatly increase the quantity of certain min-

eral materials. This is even more true of paper than of cloth since clay, titania, barium sulfate, starch, glue, rosin, and various other substances are employed for filling, opaquing, and sizing. Dyeing, bleaching, and other chemical treatments as well as raw material variations also produce alterations in both cloth and paper which in some instances will be large.

This communication reports the results of a study of the application of the density gradient tube method to the identification of cloth and paper ash. Similar studies of wood, tobacco, and other ashes are being pursued and will be reported separately. An improved procedure for establishing gradients is also included.

EXPERIMENTAL

The use of comparison gradient tubes, each of which contains one of the samples being compared, requires that the gradients be established so as to be as nearly identical as possible. This necessitates that each of the various liquid levels be added to heights which are very precisely identical. This is made difficult by drainage of liquid from the walls after adjusting each meniscus. Also, the addition of each of the liquid increments to each tube by pipet is slow and causes inhalation of vapors which is disagreeable and possibly detrimental to health.

Gradient tubes of 6mm. outside diameter were prepared as described previously. The open tops of the tubes were fire-polished but not flared. A number of tubes were laid on a board[1] equipped with side and bottom strips, and carrying parallel lines ruled at ½ inch intervals as shown in Figure 1. This board served to place the exact bottom tips of all tubes on precisely the same level. At the proper intervals for each level added, a strip of Scotch tape was placed across all tubes over one of the parallel lines so that the tape marked every tube at the same distance from the bottom. A series of such tapes served to label all levels. With a razor blade the tape was cut between tubes, leaving on each a section of tape marker. In some instances the cuts were made between every other tube and the longer piece of tape obtained was wrapped around alternate tubes to give a marking around the entire tube. This avoided parallax in adjusting the meniscuses. The intervening tubes were not marked with this procedure.

The marked tubes were placed in a rack[1] constructed of plastic as shown in Fig. 1 in which the bottoms of the tubes were aligned exactly and the tubes were held in a perfectly parallel alignment. Liquids were prepared in suitable densities as described earlier (1). Each was

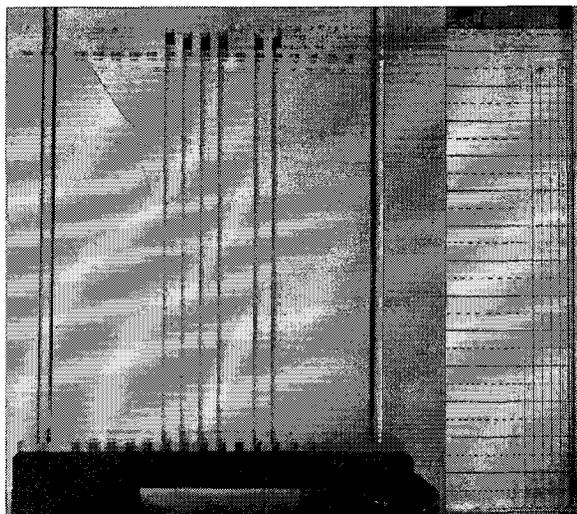1. Obtainable from Microchemical Specialties Co., Berkeley, California.

Figure 1.
Plastic Rack and Marking Board for Gradient Tubes

charged into a 50 ml. microwash bottle[1] as shown in Figure 2. Addition
from this was rapid, simple, and involved no inhalation of vapor except
that which escaped into the air.

Liquids were added in six or seven layers as described above and
previously (1). Occasional ash samples were so heavy that they re-
quired methylene iodide as the bottom layer. In some series, bromo-
form or a mixture of bromoform with methylene iodide was heavy
enough to support all of the ash samples. The top layer was usually
bromobenzene, though again this liquid was lighter than necessary for
all but a few samples. Intermediate liquids were mixed from the heavy
and the light liquid in suitable proportions. After standing 24 hours,
the gradients were sufficiently uniform to receive the samples.

Each layer was added rapidly to the various tubes, stopping the ad-
dition when the layer reached the tape mark on the tube. By the time
the last tube had received a given liquid, the first tube of the series had
drained fully and the liquid level was more or less higher than the mark.
At this point the first tube and the others successively were pushed up
over an elongated small glass tube attached to a suction vessel as shown
in Figure 2. The small amount of excess liquid could be readily re-
moved and the meniscus adjusted very precisely to the mark, where it
remained subsequently.

In early experiments different samples of the same untreated ash did
not reproduce their heights or distribution in the gradients. This was
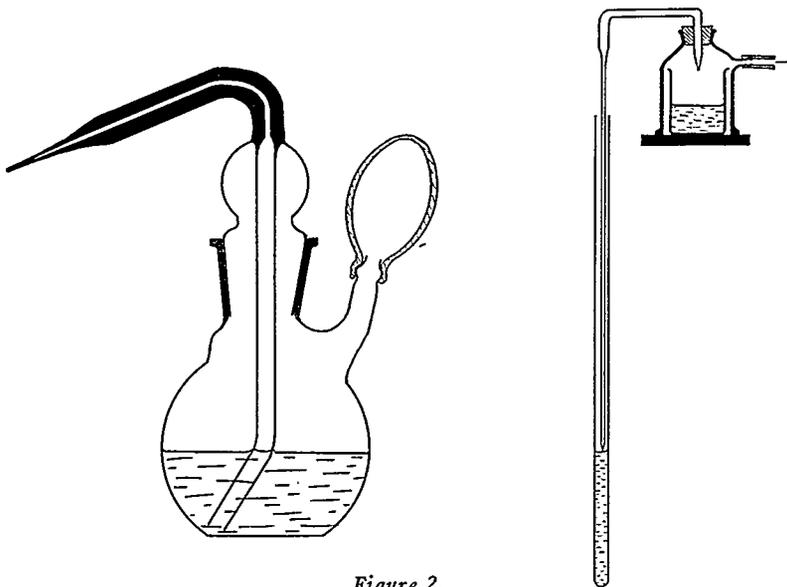found to be caused by incomplete combustion with resultant lack of

Figure 2.
Left: Micro Wash bottle, 50 ml., for charging gradient tubes.
Right: Suction device for adjusting liquid levels.

homogeneity from partially combusted material. In many samples some obvious charred material remained, and in most instances the ash had not reached its final color from heating. In order to overcome this effect, it was found necessary to ignite all samples uniformly and strongly to give the final products of the ashing process. A micromuffle furnace[1] was employed at a temperature of 1200° F. for a period usually of 15 minutes. Some ash samples became very white, while others retained more or less yellow, brown, or other color caused by iron and other non-volatile contaminents. After this ashing, all samples showed complete reproducibility when different samples of the same ash were placed in identical gradients.

In this study, the samples were not weighed as has been customary with soil and other heterogenous samples. An alternative which was essentially as satisfactory and much more rapid was to construct a small scoop which could be loaded quite reproducibly. Into each gradient tube was placed one scoopful of the sample, thus approximating a uniform volume rather than a uniform weight of the various samples. It was found quite simple to reproduce the appearance of duplicate samples of the same ash, while the differences between different ashes was in no way affected.

RESULTS

The technique described was first applied to samples of paper ash
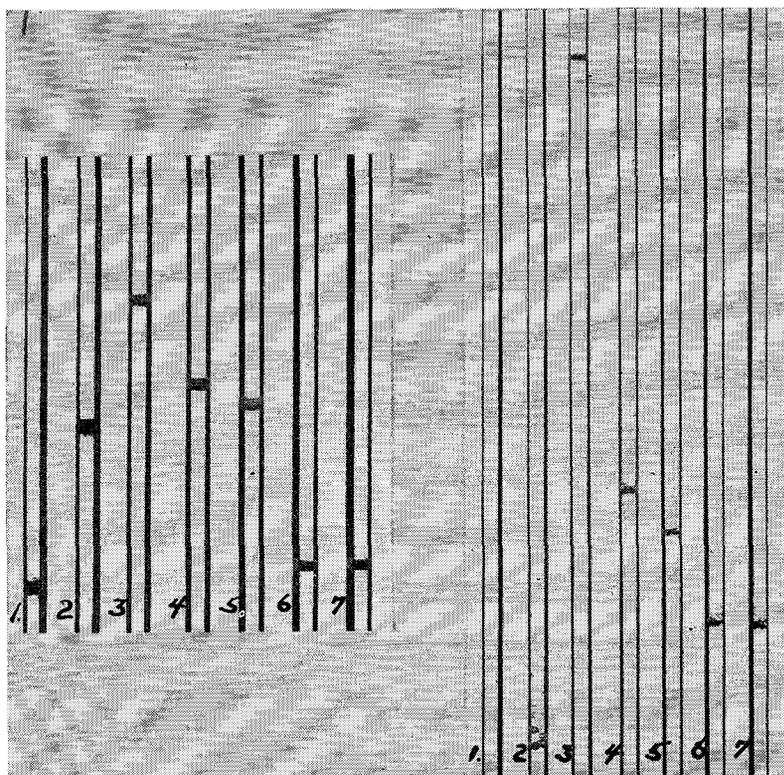
A.   *Figure 3.*   B.

A. Samples of various types of paper—left to right: 1. Newspaper, 2. Brown wrapping paper, 3. Notebook paper, 4. Bond paper, 5. Green letter paper, 6. White letter paper, and 7. Same as 6 (control).

B. Bond paper ash samples—Left to right: 1. Bankers Security Bond, 2. Gilbert Randiage (75% cotton), 3. Easton Bond, 4. Blackston Bond (opaque), 5. Highway Bond (opaque, rag content), 6. Parson's Heritage (opaque, cotton filler), and 7. Same as 6 (control).

from several types of paper to determine if there were considerable differences between various types. The results are shown in Figure 3A. They are inconclusive as regards the differences between widely different kinds of paper, for newspaper is closest to white bond, and notebook paper diverges most widely from the other types. Since bond paper might be expected to be most common in recording important documents worthy of destruction to conceal evidence, six different bond papers were compared as shown in Figure 3B. Despite the use of methylene iodide as the heavy layer, one sample of ash sank to the bottom of the tube and is not shown in the figure. All of the bond papers tested showed marked or extreme differences, greater than were shown by the different types of paper studied first. It is apparent that wide differences in the mineral constituents and content of bond papers is to be expected.

Wrapping paper also might well be destroyed because of the contents wrapped and the frequent presence on the paper of writing and of fingerprints. Nine samples, three paper bags and six of wrapping paper, were burned and ashed as described and placed in gradients. The heights of the ash of the various samples in the tubes was much more uniform than those of bond paper, only two samples being markedly different from the others. The three paper bags were distinguishable, one being quite different from the other two. Two of the wrapping papers were not clearly distinguishable, the others all being so. One was very markedly different from the others. One paper bag was also not clearly distinguishable from one sample of wrapping paper by its height, but the color of the ash was not identical.

A similar study was made of seven samples of ash from very similar appearing ruled notebook papers. Six of the samples were quite similar to each other with all but two being distinguishable by the level in the tube. These were slightly different in color and appearance and could be distinguished with some difficulty on this basis. The seventh was so heavy that methylene iodide at the bottom of the tube would not float it. The results showed clearly that wider differences may exist between samples of similar paper than will be shown between most samples of different types of paper. The identification value of the method for individual paper is good because only an occasional paper ash sample will be found that floats at the same level as a given sample, and differences in color of the ash is quite often sufficient to distinguish between samples without relying on the density of the ash. This is not the case unless the ashing is completed to eliminate carbonaceous material that normally persists in the sample that has merely been burned in a fire.

Since cloth also is subject to destruction by burning, some study was made of the possibility of identifying the ash by the gradient tube. Eight cloths were picked more or less at random, burned, and ashed as described for paper. Three cotton cloths, two wool, one silk, one acetate rayon, and one marquisette (nylon-viscose rayon) were included. Wool, silk, and nylon cloth particularly were more difficult to ash completely than either paper or the other types of cloth tested. The samples required that they be spread thinly on the crucible bottom or the charred material on the inside did not heat sufficiently. In some instances, the ash was respread after one 15 minute period in the muffle and replaced for an additional 15 minutes. Very considerable differences in the color of the ash were noted. One ash was nearly black, while others were grey or·
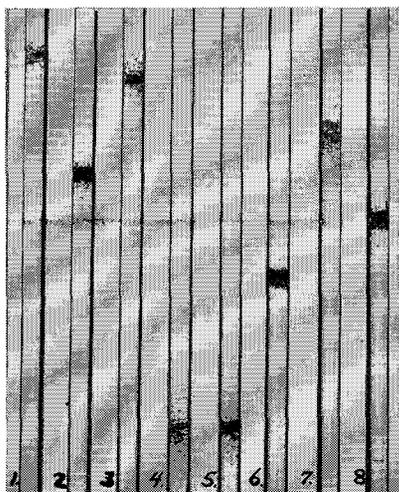
Figure 4.

Cloth ash samples—Left to right: 1. Cotton towel, 2. Silk scarf, 3. Marquisette (nylon and viscose rayon), 4. Rayon slip, 5. Same as 4. (control), 6. Cotton scarf, 7. Wool (rose color), and 8. Cotton (deep green dress).

reddish. This feature alone is sufficient to establish non-identity when the color of the ashed material is significantly different.

The results of suspending the cloth ash samples are shown in Figure 4. The rayon ash was notable as the heaviest ash, floating on methylene iodide but not on bromoform. One wool sample yielded an ash so light that it floated on the lightest liquid used ($\frac{3}{4}$ bromobenzene, $\frac{1}{4}$ bromoform), and the other wool also occurred in the lighter region but much nearer the average. Two cottons were below average density and very different from each other, the third being next to the lightest ash. In no instance were the samples from different cloth near enough to be confused under any circumstances. While no further study was made of variations in similar cloths, it can be stated here, similarly to paper, that cloths made of the same material may show greater differences of ash density than do cloths from different materials. It can further be stated that the variations in density of cloth ash are so great as to make the method of definite value in cloth ash identification. This, added to the ease of procedure should recommend it for general use in the study of identity of ash samples.

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