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INDUSTRIAL CHEMICALS

Study of the Silicic Acid Procedure of Armour and Burke for the Separation of Polychlorinated Biphenyls from DDT and Its Analogs

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The silicic acid procedure of Armour and Burke for the separation of polychlorinated biphenyls from DDT and its analogs produces an adequate separation of Aroclor 1260. However, the procedure cannot completely separate *p,p'*-DDE from Aroclors 1221, 1232, 1242, and 1254. Factors causing variable column properties were examined. The 7 hr minimum period for silicic acid activation was found to be inadequate for reproducible water-silicic acid preparations. A 24 hr minimum period is suggested. The use of Celite in column preparation often results in improperly prepared columns and should be eliminated. Nonreproducible columns might also be due to a heterogeneous water-silicic acid adsorbent which is a result of an irregular distribution of water molecules onto the silicic acid particles.

Pesticide analyses of animal tissue are complicated by the presence of polychlorinated biphenyls (PCB). The many compounds in a PCB mixture (a possible 210 different isomers) often interfere in the gas-liquid chromatographic (GLC) identification and quantitation of pesticides. This necessitates a separation prior to GLC analysis. Armour and Burke (1) have presented a procedure utilizing water-deactivated silicic acid for separating PCB from DDT and its analogs. The PCB components are first eluted from a silicic acid column with petroleum ether. The pesticides are then eluted with a more polar solvent (acetonitrile-hexane-methylene chloride). Application of the separation procedure to a fish sample yielded a PCB fraction that somewhat resembled Aroclor® 1254. Initially it was assumed that the variations observed for the relative amounts of each peak, when compared to Aroclor 1254, were due to "weathering." However, when a recovery of Aroclor 1254 was made, the same "weathering" pattern was observed. Examination of the second eluate (acetonitrile-hexane-methylene chloride), which would have contained the pesticides, revealed PCB isomers that could possibly account for the observed "weathering" pattern in the first eluate (petroleum ether). The proportion of PCB found in the pesticide eluate amounted to approximately 20%.

An investigation of some of the factors affecting the Armour and Burke procedure was undertaken, since the separation of PCB from DDT and its analogs did not occur as reported. Factors considered were room humidity, silicic acid activation and deactivation, and column preparation.

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Experimental

Reagents and Apparatus

The reagents (except the deactivated silicic acid and the individual chlorinated biphenyls) and apparatus used in this study were those specified by Armour and Burke (1).

(a) *Silicic acid.*—100 mesh analytical reagent (Mallinckrodt No. 2847). A 1" layer of silicic acid was placed in a beaker and heated >7 hr at 130°C. After heating, the hot silicic acid was quickly transferred into a previously tared screw-cap bottle and the bottle was closed, using a cap with a Teflon liner. After the silicic acid cooled, 3% water was added with a pipet. The bottle was capped tightly and shaken vigorously until a homogeneous smooth flowing mixture was obtained.

(b) *2-, 3-, and 4-Chlorobiphenyl and 2,2'- and 4,4'-dichlorobiphenyl.*—Obtained from Chemical Procurement Laboratories Inc., College Point, N.Y. 11356, and used without further purification.

Procedure

The silicic acid columns were prepared as described by Armour and Burke (1). A mixture of 10–23 μg *p,p'*-DDE and 43–54 μg Aroclor 1254 was added to the column and eluted with petroleum ether only. Ten ml fractions were collected and concentrated to 1.0 ml. Five μl of each fraction was injected into a gas chromatograph to obtain response

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vs. ml plots for the various peaks. Because of the difficulty in determining the true peak height of unresolved peaks, the absolute peak height was always plotted.

A similar procedure was used to determine the elution volumes of selected individual chlorinated biphenyls that were easily available commercially. One mg amounts of the compounds were eluted through a 7 g silicic acid column and the elution volumes obtained were calculated on the basis of a 20 g silicic acid column.

Results and Discussion

Gas chromatograms obtained by the Armour-Burke procedure for a fish sample and a recovery of Aroclor 1254 are shown in Fig. 1. The so-called "weathering" pattern obtained for both the fish sample and recovery of Aroclor 1254 is clearly due to the incomplete elution of PCB from the silicic acid column. Incomplete separations can complicate both the identification and quantitation of the various available commercial PCB mixtures. Weathering, however, might still be possible, due to metabolism of specific isomers in the PCB fraction.

Reasons for the Difference in Separations

Aromatic hydrocarbon impurities present in some commercial pesticide-grade hexanes have been reported by Zitko (2) to significantly affect the elution patterns of PCB and organochlorine pesticides from a silicic acid column. Values of 0.21 and 0.79 ml aromatic compounds/L were reported for 2 commercial hexanes. Examination of Zitko's data, however, indicates that the values reported should be 2.1 and 7.9 ml/L. The solvent used in this study was that specified by Armour and Burke, distilled-in-glass petroleum ether (Burdick & Jackson Laboratories, Inc.). Examination of 2 lots of Burdick and Jackson's petroleum ether revealed that only about 0.005 ml aromatic compounds (estimated as benzene)/L was present. The eluting solvent, therefore,

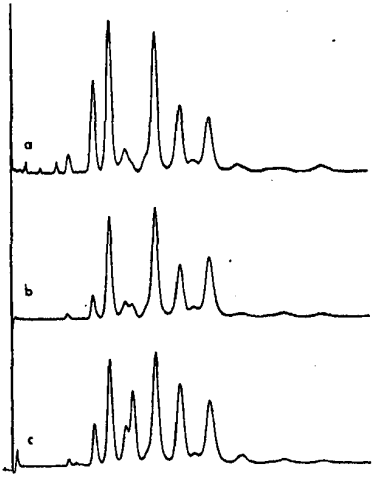

FIG. 1—Gas chromatograms of PCB components: a, petroleum ether eluate from silicic acid chromatography of a fish sample; b, petroleum ether eluate from silicic acid chromatography of Aroclor 1254; c, Aroclor 1254 (not eluted through silicic acid).

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should not be a factor in the difference in separation.

To further minimize the variables, only the "adsorbent of choice" was used in this study, Mallinckrodt's analytical reagent silicic acid No. 2847. Silicic acid of known quality (Mallinckrodt) obtained from Armour yielded the same incomplete separation, indicating that the differences in separation cannot be due to variations in silicic acid lots.

During the investigation it became apparent that some batches of 3% water-silicic acid exhibited greater activity than others. Since the silicic acid used had been activated at 130°C for periods of 7 hr to one week it is believed that the variation in silicic acid activity is related to the different activation periods used. Silicic acid activation involves the removal of water adsorbed at active sites. Complete activation at 130°C is not possible, as temperatures of 150–200°C are necessary for maximum removal of adsorbed water molecules (3). However, activation at 130°C should be acceptable if the silicic acid so obtained is of a reproducible water-silicic acid composition, i.e., activity. Table 1 shows the variation of weight with time for silicic acid heated at 130°C. No significant weight changes were observed after 24 hr. For simplicity, silicic acid activated at 130°C for more than 24 hr may be defined as not having any adsorbed water. Note that the weight loss between 7 and 24 hr is about 0.5%. Thus, water-silicic acid prepared from silicic acid activated for 7 hr and containing 3% added water will yield a 3.5% water-silicic acid adsorbent. For reproducible adsorbent preparation the minimum activation period should be changed from 7 to 24 hr. All of the work reported in this paper has been done with silicic acid activated at 130°C for more than 24 hr.

Figure 2 shows that a complete separation of Aroclor 1254 and *p,p'*-DDE is not obtained on the 3% water-silicic acid column. An attempt to obtain a better separation by using a 3.5% water-silicic acid column seemed to work for a standard mixture of 10 μg *p,p'*-DDE and 43 μg Aroclor 1254. Better recoveries of the various PCB peaks were obtained with no apparent *p,p'*-DDE interference. However, when a similar column was used for a fish sample containing 260 μg *p,p'*-DDE and 40 μg Aroclor 1254, an appreciable amount of *p,p'*-DDE was found in the PCB fraction (petroleum ether). While it may appear that the separations obtained for the standard mixture and the fish sample were different, the results can be explained in terms of similar separations. The elution profiles of the various PCB components and *p,p'*-DDE (Fig. 2) may be considered, as is commonly done in chromatography, as being distribution curves. Assume, for example, that 1% of the *p,p'*-DDE present elutes within 250 ml of the petroleum ether elution. While it may not be possible to detect 1% of the 10 μg *p,p'*-DDE in the standard mixture in the PCB fraction (petroleum ether), it is possible to detect 1% of 260 μg *p,p'*-DDE in the fish sample.

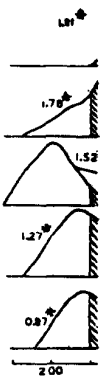
Table 1. Variation of weight with time for silicic acid heated at 130°C

Heated, hr	Wt Silicic Acid, g
0	114.37
1	110.64
3	106.42
4	105.96
7	105.62
8	105.42
24[a]	105.07

[a] No significant weight changes were observed after 24 hr.

Another explanation that may be advanced for the separation obtained for the fish sample described above is that the silicic acid column was overloaded. Linear elution in adsorption chromatography occurs only at low sample concentrations. At higher sample concentrations the elution volume generally decreases (3). Armour and Burke (1), as well as Bellman and Barry (4), emphasize the necessity of limiting the size of the sample placed on the silicic acid column. The work in this study was done with standards which were in the same concentration range used by Armour and Burke. With actual fish samples, however, the range of concentration is not controllable. With the fish sample previously described, while the PCB placed on the column compared favorably to the amount used by Armour and Burke, the *p,p'*-DDE present was more than 100 times the amount they used.

At this point it is possible to postulate on the reasons for the differences between the results obtained in this study and those of Armour and Burke. The bulk of the work by Armour and Burke was probably done with silicic acid that had been activated at 130°C for only 7 hr, their minimum specified period. A 7 hr activation period begun at the start of a working day conveniently allows the water-deactivated silicic acid



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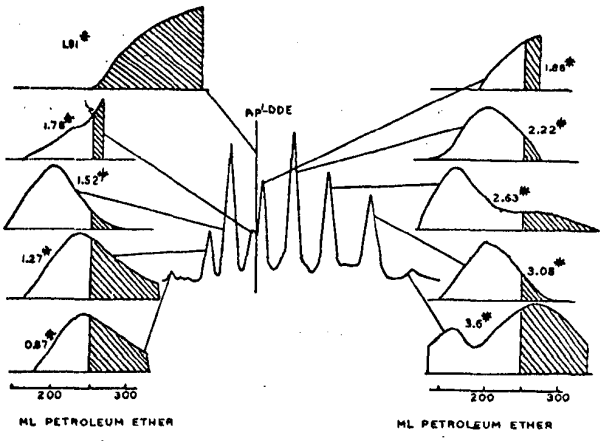

FIG. 2—Petroleum ether elution pattern of p,p'-DDE and Aroclor 1254 from a 3% water-silicic acid column: solid area, portion eluting before 250 ml; lined area, portion eluting after 250 ml.

to be equilibrated overnight for the specified 15 hr. The separations that they obtained for their 3% water-silicic acid columns were comparable to that of a 3.5% water-silicic acid column, due to the incomplete removal of 0.5% of the physically bound water. The lower activity of the column resulted in a more quantitative elution of Aroclor 1254. The low p,p'-DDE:Aroclor 1254 ratios used by Armour and Burke in their recoveries, less than 0.2 in 2 spiked samples, prevented traces of p,p'-DDE present in the PCB eluate (petroleum ether) from being detected. The net effect was an impression that Aroclor 1254 and p,p'-DDE were separated completely.

Limitations of the Separation Procedure

Separations were made of other Aroclors (1221, 1232, 1242, and 1260) that Armour and Burke reportedly separated. Decreasing amounts of PCB were recovered in the "PCB eluate" (petroleum ether) as the chlorine content decreased. Aroclor 1260 was eluted mainly in the first eluate. Some peaks were found in the second eluate (acetonitrile-hexane-methylene chloride) which corresponded to the Aroclor 1254 peaks found in that eluate. However, since these peaks represent only a small fraction of the compounds of Aroclor 1260, the PCB peaks found in the second eluate amounted to only a few per cent. This establishes a reasonable method for Aroclor 1260 with a recovery of approximately 98%. Aroclor 1221 (Fig. 3), on the other hand, was eluted in the second eluate (acetonitrile-hexane-methylene chloride). Decreasing amounts of PCB were found in the second eluate for Aroclors 1232 (Fig. 4) and 1242 (Fig. 5), respectively. The separation technique can be applied to Aroclor 1254 if a less active adsorbent, such as the 3.5% water-silicic acid, is used, although Fig. 2 illustrates that a complete separation of Aroclor 1254 from p,p'-DDE cannot be obtained. However, p,p'-DDE interference can be tolerated in this case, so that a better estimate of Aroclor 1254 may be obtained. DDE quantitation should not be a problem as an electron capture detection system is more sensitive to p,p'-DDE than it is to Aroclor 1254. Thus the effect of the PCB peaks appearing near p,p'-DDE will be diluted out during its quantitation.

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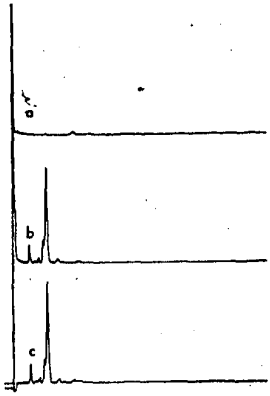

FIG. 3—Gas chromatograms showing the separation of Aroclor 1221 from a silicic acid column: a, petroleum ether eluate; b, comparable volume of acetonitrile-hexane-methylene chloride eluate; c, Aroclor.

Stalling and Huckins (5) have shown that Aroclor 1221 is a mixture of biphenyls containing 1–3 chlorines per molecule. Table 2 gives the volumes observed for elution of a number of selected mono- and dichlorinated biphenyls from a 3% water-silicic acid column. These values, when compared with the elution volumes required for Aroclor 1254 (Fig. 2), a mixture of biphenyls containing 4–7 chlorines per molecule (5), represent an extremely wide range. In essence the volume of petroleum ether would have to be increased to ensure complete elution of all PCB. Such a drastic change could also allow the facile elution of pesticides for which the separation procedure was initially designed.

Snyder (3) maintains that the adsorption energy of 2-methyl biphenyl is lower than that of 3- or 4-methyl biphenyl due to steric hinderance by the *ortho* substituent. Substituents in the *ortho* positions of biphenyl prevent a planar configuration, which is postulated to be preferred for adsorption. It would be expected that 2-chlorobiphenyl will elute before 3- or 4-chlorobiphenyl and that 2,2'-dichlorobiphenyl would be the first to elute as it is the most nonplanar compound studied. In practice, however, both 2-chlorobiphenyl and 2,2'-

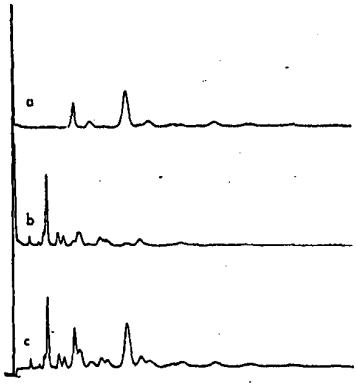

FIG. 4—Gas chromatograms showing the separation of Aroclor 1232 from a silicic acid column; see Fig. 3 for identification.



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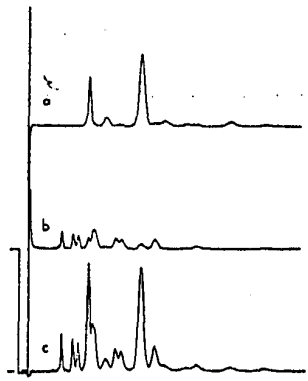

FIG. 5—Gas chromatograms showing the separation of Aroclor 1242 from a silicic acid column; see Fig. 3 for identification.

dichlorobiphenyl have retention volumes much greater (660 and 630 ml, respectively) than their respective planar counterparts (490 ml for 3-chlorobiphenyl, 540 ml for 4-chlorobiphenyl, and 370 ml for 4,4'-dichlorobiphenyl). The chemical and stereochemical properties of the individual chlorinated biphenyls were confirmed by ultraviolet, infrared, and microcoulometric chlorine analysis and by comparing their relative GLC retention times with those given by Zitko *et al.* (6). Apparently the effect of chlorine (chlorine substitution) on biphenyl adsorption is more complex than originally postulated. Further studies are being contemplated with a wider range of chlorinated biphenyls in order to understand the adsorption process more fully.

Factors in Reproducible Column Separation

During this study, inconsistent separations were obtained from columns prepared from the same batch of water-silicic acid. Armour and Burke limit the usefulness of water-silicic acid to a period of about 5 days. It seemed that the inconsistent separations might be due to changes in the activity of the water-deactivated silicic acid relative to time. Figure 6 gives the elution patterns for 4 columns prepared on different days from the same batch of water-silicic acid. Similar

Table 2. Amounts of petroleum ether required for elution of individual chlorinated biphenyls from silicic acid column[a]

Compound	Retention Vol., ml
4-Chlorobiphenyl	540
3-Chlorobiphenyl	490
2-Chlorobiphenyl	660
2,2'-Dichlorobiphenyl	630
4,4'-Dichlorobiphenyl	370 (360[b])
Biphenyl	890

[a] 7 g silicic acid column used; results were calculated on the basis of a 20 g column.
[b] Value obtained from a 20 g silicic acid column.

elutions for columns prepared on Days 1 and 6 discount an age-to-activity correlation. However, Fig. 6 gives an indication of the reproducibility of the silicic acid separation. Since a good reproducible column is necessary for routine pesticide analyses, factors responsible for the large variation of the Day 5 column, as well as the small variation of the Day 4 column, need to be better understood. The following factors were considered: (*1*) room humidity, (*2*) heterogeneous deactivated silicic acid, and (*3*) improper column preparation.

Changes in room humidity probably do not explain the large variation observed for Day 5. Elution profiles of a 3.5% water-silicic acid column indicate that a difference of 0.5% water will produce an elution volume change of about 30 ml. To obtain the 30 ml variation between Day 5 and Days 1 and 6, the 20 g 3% water-silicic acid would have to adsorb the moisture from 10 L air at 50% relative humidity in addition to the moisture it normally adsorbs. Since the work was done in an air-conditioned laboratory, large humidity variations were not a factor.

The homogeneity of the silicic acid in the chromatographic column may also be an important factor in reproducibility. Because variations had been observed in previous work, the water-silicic acid mixture used in Fig. 6 was shaken for 1 hr immediately after the addition of water. The water-silicic acid adsorbent was probably free of gross differences in composition. However, the following similarities and differences between Day 4 and Days 1 and 6 (Fig. 6) suggest that minor variations in column properties are still present:

(*1*) The appearance of *p,p'*-DDE occurs at approximately the same elution volume for the 3 runs.

(*2*) The elution pattern for the peak at 1.27 is similar for all 3 days.

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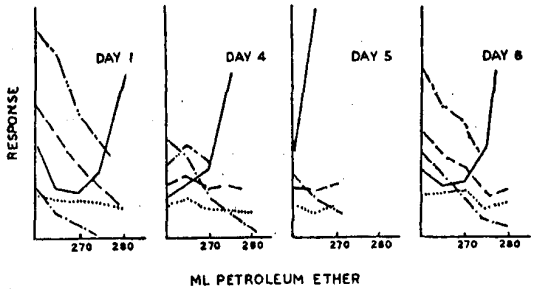

FIG. 6—Elution profiles of 4 silicic acid columns prepared on different days from the same batch of 3% water-silicic acid: . . ., peak at 1.27; ——, peak at 1.8 (Arecior peak at 1.78 and p,p'-DDE at 1.81); — . . —, peak at 1.88; — . —, peak at 1.22; — —, peak at 2.63. Retention times obtained on DC-200 column, relative to aldrin.

(5) The elution patterns of the peaks at 1.88 and 2.63 of Day 4 are different from those of Days 1 and 6.

It is difficult to attribute these similarities and differences to a column error, such as channeling, since all of the peaks should be affected similarly.

When water is initially added to activated silicic acid, lumps of water-silicic acid are immediately formed. The mixture is shaken until the lumps are broken up and the mixture is fluid and even in texture. Snyder (7) postulates that silicic acid has a range of sites with different activities and that the added water is rapidly equilibrated to the most active sites. However, rapid equilibration may be true only for high water-silicic acid ratios. At lower water-silicic acid ratios, water-silicic acid bonds at moderately active sites may prevent the transfer of water molecules to more active sites.

To support the argument that the added water is not distributed preferentially to the active sites, 3% water-silicic acid was heated 3 days in a tightly capped bottle at 130°C in an attempt to obtain better uniformity. Elution profiles obtained from this heat-treated silicic acid exhibited a definite decrease in activity (Fig. 7). All peaks appeared about 100 ml sooner. For example, p,p'-DDE was detected at 270 ml when eluted through unheated 3% water-silicic acid, while it was detected at 170 ml when eluted through heat-treated 3% water-silicic acid. Active sites responsible for the chromatography of PCB and pesticides have been deactivated by water molecules that had been adsorbed initially by less active sites. Water-silicic acid adsorbent is a heterogeneous mixture containing a wide range of active sites, some of which might well have been rendered completely inert by contact with liquid water and some of which are completely active. The relative amounts of each species as well as the intermediate forms in a water-silicic acid mixture should be subject to the techniques used in its preparation. The author believes that if the mixture is shaken vigorously about 1 hr immediately after the addition of water, a nearly uniform and reproducible mixture will be obtained. In addition, the 5 day limit on the usability of the water-silicic acid is an unnecessary precaution.

Ries (8) has reported that steam treatment of silica results in an adsorbent with reduced surface area. It may be argued that the lower activity of the heated 3% water-silicic acid is due to reduced surface area. The temperatures used by Ries, sometimes more than 500°C, were more severe than the 130°C used in this present study. Reactions causing particle-particle fusion and permanent loss of surface area occur at temperatures greater than 400°C, while adsorbed water is lost at 150–200°C (3). Reduced activity of the heated 3% water-silicic acid was probably not due to a change in the surface structure.

Stalling and Huckins (5) have modified the Armour and Burke procedure by adding water to

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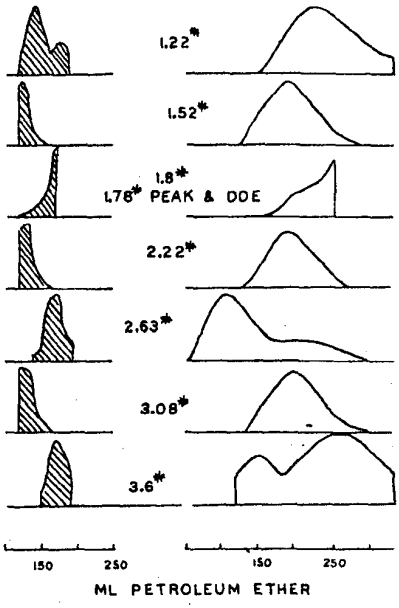

FIG. 7.—Comparison of elution patterns of *p,p'*-DDE and Aroclor 1254 on: solid area, heat-treated 3% water-silicic acid, and lined area, unheated 3% water-silicic acid.

silicic acid previously mixed with Celite. The resulting adsorbent exhibited reduced activity, yielding 41% of the DDE within 250 ml petroleum ether elution. Apparently Celite aids in the regular distribution of water which results in a greater number of active sites being deactivated.

Although irregular distribution of water may be responsible for the large variation of Day 4 (Fig. 6), it is unlikely that it is responsible for the large variation of Day 5. Another possibility is that the columns were prepared improperly. The use of Celite presents a problem in homogeneous column preparation. Some columns were discarded because they contained pockets of Celite that formed when the silicic acid-Celite slurry was not stirred immediately after its transfer to the chromatographic tube. A completely homogeneous silicic acid-Celite column is difficult, if not impossible, to prepare. An elution profile for a silicic acid column prepared without Celite showed that Celite does not take a direct role in the chromatography of Aroclor 1254 and *p,p'*-DDE. The Celite is used as a filter aid for easier column flow and, since air pressure is used to maintain column flow, its use should not be necessary. No difficulties were encountered when columns were prepared without Celite. Figure 8 illustrates the elution patterns obtained from 5

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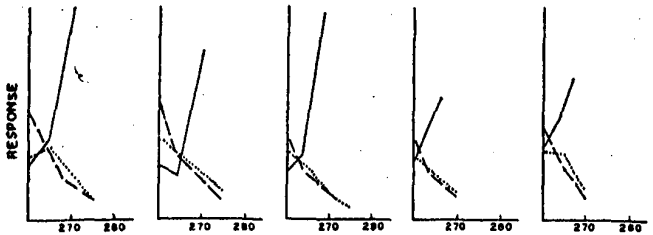

FIG. 8—Elution profiles of 5 silicic acid columns prepared without the use of Celite; see Fig. 6 for identification.

different columns that were prepared on the same day without Celite. The similarities of all 5 patterns suggest that the use of Celite is responsible for the large variation of Day 5 in Fig. 6.

Conclusions

Current analytical procedures group all of the possible 210 polychlorinated biphenyl isomers as one "entity." Their chemical behavior on elution with petroleum ether through a water-silicic acid column indicates that much larger volumes than previously employed must be collected to ensure total PCB recovery. Separations based on PCB components containing 4 or more chlorine atoms per molecule will not work for PCB containing less chlorine. The silicic acid separation of Armour and Burke is specific only for Aroclor 1260. The procedure may be applied to Aroclor 1254 if a less active water-silicic acid adsorbent is used, although this will result in *p,p'*-DDE interference in the PCB eluate.

For reproducible silicic acid columns it is suggested that the use of Celite be eliminated and that the minimum activation period for the silicic acid be increased to 24 hr. The water-silicic acid mixture should be adequately mixed to minimize heterogeneous water molecule distribution which can cause gross variations in column properties. Although the effects of room humidity are probably minimal, contact of silicic acid with air should be kept at a minimum during column preparation.

Acknowledgments

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