

The Early Development and Rapid Growth of Gas Chromatographic Instrumentation in the United States

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Abstract

The role of the American instrument companies involved in the development and manufacturing of gas chromatographic (GC) instruments up to about 1962 is reviewed. In addition, the origin of some supply houses is discussed and the role of the instrument companies in the dissemination of technical information is emphasized. Finally, some data are given on the worldwide GC market, and its expected evolution in the near future is outlined.

Introduction

Although the first gas chromatographs (GCs) were self-built, it soon became obvious that the sophisticated systems necessary for their proper operation did not belong to the standard laboratory equipment and the construction of the whole unit was beyond the capabilities of the analyst; the immense potential of the technique could only be fully exploited if proper instrumentation became available. The first commercial GCs were introduced in the mid-1950s and their availability made the rapid expansion of the technique and its use possible.

The need for standardized instruments coincided with the emergence of a new branch of the industry: the scientific instrument industry. This infant industrial branch greatly benefited from the demand of these new types of instruments. In fact, we can observe a symbiosis between GC and the scientific instrument industry; the evolution of the former could not happen without the involvement of the latter. In this process the American scientific instrument companies were particularly active and had the lion's share.

GC was developed in Europe¹, and practically all of the American instrument companies received their basic know-how directly or indirectly from Europe. Some British companies actually started to provide GCs at an early stage of the devel-

opment. Still, however, in general, the impact of the American instrument companies was more significant in the overall development. This followed from the availability of a much larger geographical area without any market restrictions and from the technological advantages of the American companies gained during their involvement in supplying high-precision systems for the Allied military and war efforts. While it took some time to rebuild the European economy, these American companies emerged at the end of World War II with a superior know-how in electronics and optics and in high-precision work; thus, they were eager to turn their energy to new fields, and scientific instruments represented such a new, promising field.

In the first two decades the design and performance of the GCs became continuously more sophisticated, adding new techniques (e.g., temperature programming), new detectors (e.g., the ionization detectors), and new column types (e.g., the capillary columns and new stationary phases) to the originally relatively simple isothermal systems. In this respect it is worthwhile to compare an early instrument, the Model 154 (Figure 1) introduced in 1955, with the Model 900 (both of Perkin-Elmer) introduced in 1967. The Model 900 is probably the best example for the highly sophisticated GC systems; as shown in Figure 2, its front panel had over 30 switches, knobs, and dials. Oven temperature was controlled by separate dials setting the initial and final temperatures (each in steps of 5°C between -75°C and +400°C when using with a subambient temperature control accessory) and the program rate (providing 15 rates between 0 and 32°C/min). Separate dials permitted the setting of the initial and final isothermal periods and even the rate of cooling at the end of the program. During actual programming the large middle dial was slowly turning, indicating the actual temperature. However, even in these more sophisticated systems the basic design essentially remained the same; these were analog systems relying on manual, mechanical controls, the difference being only that more functions could be controlled and more precisely.

In the mid-1970s major changes started to be introduced in the construction of our GCs because of advances in elec-

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¹ Ralph Müller, who in the 1950s published a monthly instrumentation column in the "A" pages of *Analytical Chemistry*, went so far that in one of his contributions dealing with the sudden interest in GC he stated that "modern [gas] chromatography is practically a British science" (1).

tronics; the analog systems reigning for 20 years were to be replaced by microprocessor-controlled, digital systems, and the first such instrument was introduced at the 1977 Pittsburgh Conference. In these instruments, oven temperature and the analytical conditions were set from a central keyboard, and not with the help of the many knobs and dials. Also, the conventional packed columns were replaced by capillary columns in an increasing number of applications. At the same time a new generation of chromatographers started to emerge who did not participate in the early development of GC. Thus, at that time, we considered it important to summarize the information related to the beginnings of the evolution, to



Figure 1. The Model 154 Vapor Fractometer from Perkin-Elmer introduced in May 1955. Behind the door on the left was the U-shaped column mounted on the thermal-conductivity detector block; both were in a constant temperature air thermostat. A potentiometric recorder was housed in a similar cabinet (not shown here).



Figure 2. The Model 900 GC from Perkin-Elmer introduced in March 1967. The column oven opened on the top left; it permitted the installation of two columns (packed or capillary). Two FIDs were in the wire cage to its right; other detectors could also be installed at this place. Behind the detectors were the controls for three gases: the carrier gas, air, and hydrogen for the FIDs. The dual injection port was left of the name plate. The various knobs and dials represent temperature controls and controls for the detectors' electronics.

document it to this new generation, permitting them to compare the systems used by the pioneers with their instruments. This detailed report was published in the March/April 1977 issue of this journal (2). As an introduction to this paper, Dr. Roy A. Keller, the editor of the journal, emphasized the importance of such a compilation. As he stated, instrument developments are usually not reported in scientific papers, thus details of these important contributions are lost, fading into history, and could not be reconstructed anymore without such reports written by a participant of the evolution who "was there".

Today, 25 years after this publication, GC, its instrumentation, and the industry manufacturing and marketing these products are again at a crossroads. By now, GC became a mature technique; the instruments became commodities in which one would expect only very little changes. In fact, while in the first decade of commercial instruments new GCs were introduced almost yearly; the design of today's instruments remained practically unchanged for many years. However, a number of new trends are evident that will definitely change radically the design of the future instruments. Also, a new generation of chromatographers have grown up, for whom the early development of the technique and its instrumentation is more remote than the Model T Ford was for my generation. Therefore, we feel that a recapitulation of the essential part of the 1977 paper (2) may be of interest to present-day chromatographers; reading about the beginnings, the work of the pioneers, and the early design of the instruments may probably help them in the better understanding of their present systems. In this respect, I would like to cite the old saying: those who don't know history will repeat its mistakes!

This is the purpose of this paper: we shall discuss the evolution of GC instrumentation in the first decade and this discussion will be based on the paper published 25 years ago. A special part will deal with the role of the instrument companies in the dissemination of technical information in the early stages of GC development. In addition, in the last part of this paper we shall compare the early times of GC instrumentation with the present, pointing to the meteoric rise in sales, which made GC within two scores of years one of the most widely used laboratory technique, surpassed only by its child: high-performance liquid chromatography.

1955: The Pioneers

The first three American instrument companies involved in GC and introducing instruments in 1955 were Burrell Corp., Perkin-Elmer, and Podbielniak. From these, two—Burrell and Podbielniak—are unknown to present-day chromatographers. It is intuitive to investigate their origin and the circumstances leading to their involvement in GC.

Burrell Corporation

This company was founded in Pittsburgh, PA, after World War I, by Guy Burrell, a mining engineer with the aim to supply gas analysis equipment mainly to the natural gas

industry. At that time the so-called charcoal test was used for the determination of the higher hydrocarbon content of natural gas. In the more advanced form of this test developed by Henry B. Hass, then research director of the Gasoline Recovery Corporation, the sample was passed through a tube containing charcoal; next, the adsorbed compounds were gradually displaced by glycerol and measured volumetrically after collection (4). This work was continued by Nelson A. Turner in cooperation with Burrell Corp., resulting in the so-called Turner-Burrell Adsorption Fractionator introduced in 1943 (5,6). This was a fairly complicated, floor-standing instrument employing a large column filled with charcoal, and desorption was carried out by a combination of a moving heater and mercury vapor; the separated fractions were registered by a thermal-conductivity detector (7).

During World War II, a crash program was carried out in the U.S. to develop a synthetic rubber industry (the sources for natural rubber were now under Japanese occupation). For this, more improved methods were needed to analyze the volatile hydrocarbons used as the raw material. The Podbielniak low-temperature distillation systems used for these measurements (as will be discussed) were very complicated and had some inherent problems; therefore, Burrell tried to further improve the Adsorption Fractionator. The result of this work—carried out under the guidance of Lloyd V. Guild—was the so-called Fracton introduced in 1953 (Figure 3). In this instrument (8,9) mercury vapor was replaced by organic vapor displacement using tetrachloroethylene², and the detector and fraction collection system were thermostatted at an elevated temperature. Also, the time of analysis and the necessary sample size were reduced as compared with the Turner-Burrell system.

² Most likely neither the Adsorption Fractionator nor the Fracton would be permitted today in a laboratory: mercury or tetrachloroethylene vapors certainly do not conform with present-day health regulations!

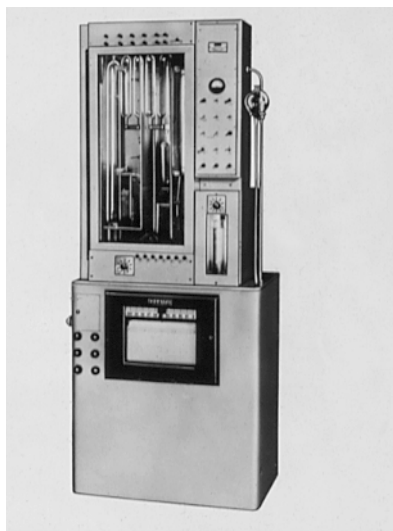


Figure 3. The Fracton from Burrell Corp. introduced in 1953. In this instrument the sample components were retarded on an adsorption column and successively desorbed (displaced) with tetrachloroethylene vapor.

As mentioned, the Fracton was introduced in 1953, less than a year after the seminal paper of James and Martin on gas-liquid partition chromatography (10). At this time the laboratories of the large American petroleum and chemical companies already started to explore the possibilities of GC. Among these was the Tennessee Eastman Co. in Kingsport, TN, and the first paper on GC in the U.S. was presented by Hugh W. Patton of this company at the Symposium on Hydrocarbons and Related Compounds held during the Fall 1954 National Meeting of the American Chemical Society (11). Lloyd Guild of Burrell attended this meeting and immediately realized that the Fracton could be easily modified into a real GC. The development took only a few months and the new instrument—the first American commercial GC—was introduced in March 1955. This was the Burrell Kromo-Tog Model K-1 (Figure 4). The instrument had a 100-cm-long column at room temperature, a flow-through thermal-conductivity detector in a constant temperature air bath, a recorder, and a constant-temperature gas inlet system with 1–20-mL variable volume sample tubes.

Patton's first paper only dealt with gas *adsorption* chromatography, and accordingly, the original Kromo-Tog Model K-1 used only charcoal as the column packing. However, within a year Patton's group also discussed the possibilities of gas *partition* chromatography (12), and then, Burrell also offered partition columns. In the Model K-1, the column was unheated; however, in 1956, Burrell introduced a more advanced model, the Kromo-Tog Model K-2, in which the column was heated with the help of a nichrome wire wound around it (13).

In the subsequent years Burrell Corp. further improved their instruments, extending their range of application. However, the way of column heating remained the weak point of their instruments; instead of an air thermostat the columns were heated by wrapped-around wires. Around the middle of the 1960s, the company discontinued their interest in GC.

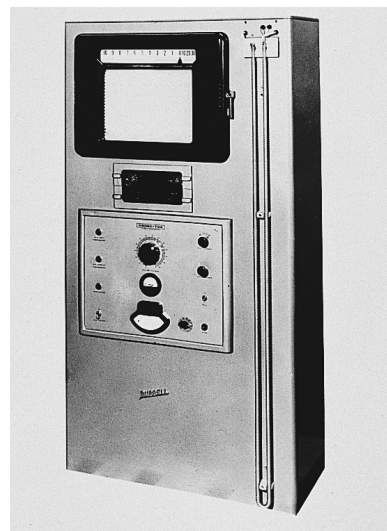


Figure 4. The Kromo-Tog Model K-1 from Burrell Corp. introduced in March 1955. This instrument had a 100-cm-long U-shaped column (cover removed). In later models a heating coil was wound around the column. The potentiometric recorder was built into the instrument.

The Perkin-Elmer Corporation

The second company introducing a commercial GC in the U.S. was The Perkin-Elmer Corporation, then located in Norwalk, CT. The company was founded in 1937 by Richard S. Perkin and Charles Elmer to supply precision optics. They started to produce infrared spectrophotometers in the mid-1940s and by the first part of the 1950s they became one of the leading manufacturers of such instruments. By that time the company had increasing contacts with scientists in Europe, particularly in England. Dr. Van Zandt Williams, then director of instrument development at Perkin-Elmer, heard during one of his visits to England in 1953–54 about GC. He sent Harry H. Hausdorff, then head of the IR Applications Laboratory, to learn more about the technique, and he visited C.S.G. Phillips at Oxford University and A.J.P. Martin and A.T. James at the National Institute for Medical Research in London. Returning from his trip Hausdorff gave an enthusiastic report on the potentialities of GC. At first Perkin-Elmer considered GC mainly as a sampling system for IR (simplifying the analysis of mixtures), but they realized during development the immense potential of the technique in itself. In 1954, a breadboard model was constructed and detailed investigations were carried out by Hausdorff on the influence of the operation parameters on the analytical results (14). In this period, researchers at the American Cyanamid Laboratories in Stamford, CT (with whom Perkin-Elmer had close contact), also started to investigate the possibilities of GC, and Perkin-Elmer cooperated with Dr. E.F. Williams in these investigations³.

The final version of the instrument, the so-called Model 154 Vapor Fractometer, was introduced in May 1955 (15).

It may be interesting to note that during development, A. Savitzky investigated in detail the possibilities of a large number of physical measurements to serve for detection (16). After careful considerations thermal-conductivity detection using thermistor beads (produced by Victory Engineering Co.) was selected. In the detector block the column effluent flew through the sensing side, while the carrier gas line bypassed the reference side and filled it by diffusion.

The Model 154 (Figure 1) employed U-shaped glass or metal columns of 1-m length and 1/4-inch o.d., and two such columns could be installed in series, directly on the detector block, in a thermostatted oven that could be heated up to 150°C. Contrary to Burrell, Perkin-Elmer immediately concentrated on partition columns; at introduction columns with two liquid phases (dodecyl phthalate and 2-ethylhexyl sebacate) were available. The instrument employed a flush vaporizer with a rubber septum and syringe injection for the introduction of liquid samples.

The Model 154 was a very successful instrument (Figure 5), it was simple to operate, with only a few controls, and for some years it was the most widely used commercial GC. Ralph Müller, in one of his monthly instrumentation columns, called it “a splendid example of automatic analysis”, and speaking about the form of chromatograms (obtained by using a potentiometric recorder) he added that “if the reader can tolerate our

exuberance, which we hope is contagious, these recordings are a delight to behold” (17). The first advertising of the instrument (18) showed the analysis of liquefied petroleum gases (Figure 6); up to then such a measurement could only be carried out with the help of low-temperature distillation (using the Podbielniak instrument), a fairly difficult, long, and tedious technique. The chromatogram in the ad demonstrated this analysis in 23 min by GC.

Within a year the original version of the Model 154 underwent a number of changes: W-shaped metal columns of 2-m length became available and the list of standard columns with various stationary phases was extended to over one dozen. In fact, Perkin-Elmer was the first company acting also as a “column supply house.” The temperature control was improved and extended to 225°C, and a very simple and reliable valve for

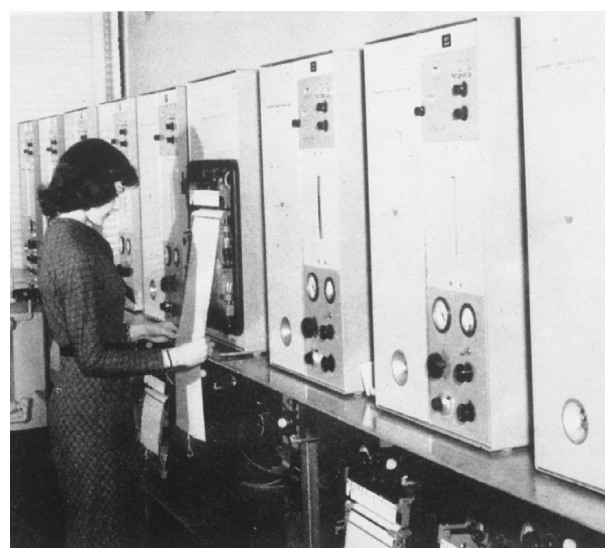


Figure 5. Routine GC laboratory at Esso R&D Co. in early 1958. This room contained at that time eight Model 154 GCs. Except for the unit attended by the operator the potentiometric recorders were placed directly under each chromatograph.

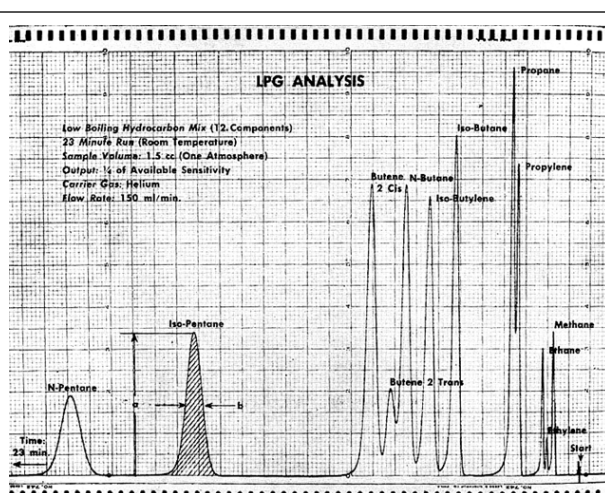


Figure 6. Part of the first advertising of Perkin-Elmer showing the separation of C₁–C₅ hydrocarbons and explaining the way of quantitative calculation.

³ In order to distinguish between Dr. V.Z. Williams, the R&D director of Perkin-Elmer, and Dr E.F. Williams of Cyanamid, the latter was nicknamed “tiny Williams” because of his smaller stature.

the introduction of gaseous samples also became available. This system (19) invented by E.S. Watson—the engineer mostly responsible for the overall design of the Model 154—and D.R. Bresky was a rotary-type valve with variable sample loops; in fact all the present-day multiport valves can be traced back to this design.

The Model 154 was also the basis of a special triple-stage instrument, the Model 188 (Figure 7), introduced in the middle of 1957 (20) and described in detail at the 1958 Pittsburgh Conference (21). Such systems were in use mainly by the petroleum companies before the advent of temperature programming for the analysis of wide boiling range mixtures. In such systems a



Figure 7. The Model 188 triple-stage GC from Perkin-Elmer introduced in the middle of 1957. The three units were connected in series but could also be used independently. Each unit had its own potentiometric recorder.



Figure 8. A publicity photo showing four instruments from Podbielniak Co.: two fractional distillation units and two Chromacon 9400 series GCs (second and fourth unit). The original Chromacon Model 9400 was introduced in December 1955.

number of GC units (three in the Model 188) were connected in series so that the detector outlet of one unit was connected to the inlet of the following unit and each unit was at a different temperature. In this way each unit was more or less optimized for a given segment (boiling range) of the sample. Such systems were very cumbersome to operate and, with the introduction of temperature programming in 1959, these complex instruments soon died out.

In 1957, the utility of the Model 154 was also extended by providing a preparative column (22). Contrary to other systems introduced by others about the same time, the Perkin-Elmer accessory consisted of a number of normal-sized columns used parallel to one another; the carrier gas flow from the injector was divided into parallel flows to the individual columns, and the column effluents were united again to provide a single outlet to fraction collection.

The Model 154 underwent a number of successive changes (e.g., the addition of a flame-ionization detector (FID) and arrangement for capillary columns in 1959) (23). In 1962, Perkin-Elmer also introduced two new instruments: the Model 226 for capillary columns and the Model 800 for dual-column baseline compensation operation⁴. However, the isothermal Model 154 continued to be marketed until 1967.

It should be mentioned that soon after the introduction of the Model 154, Perkin-Elmer also developed an automated process GC analyzer, the Model 184, introduced in 1957. This R&D work was carried out in cooperation with the laboratories of Phillips Petroleum Co. (Bartelsville, OK) (24,25). However, at the beginning of the 1960s this product line was transferred to Mine Safety Appliances Co. in Pittsburgh, PA.

Podbielniak, Inc.

The third company introducing in 1955 GC instruments was Podbielniak, Inc. of Chicago, IL.

A few years ago we have dealt with this company and its involvement in GC (26); thus, we only briefly summarize these questions here. The company was founded in 1929 in Tulsa, OK, by Walter J. Podbielniak and in 1934 the company moved to Chicago, IL. Originally, it was a testing laboratory, and then, a few years later, it started to produce a special low-temperature fractionation distillation (LTFD) apparatus used mainly in the natural gas, petroleum, and petrochemical industries; in fact, they had a virtual monopoly in these fields⁵.

Walter Podbielniak had frequent contact with the various petroleum laboratories in both the U.S. and Europe, and he heard as early as 1952–53 about GC; in fact, he also visited the laboratory of A.J.P.

⁴ The Model 800 was the first GC using $\frac{1}{8}$ -inch-o.d. packed columns. Until then every instrument utilized $\frac{1}{4}$ -inch-o.d. columns.

⁵ Another important product of the company was a centrifugal countercurrent solvent extractor used in the production of penicillin and other antibiotics.

Martin in London. In 1954, S.F. Birch, the head of British Petroleum Company's research laboratory in England, when visiting Podbielniak in Chicago specifically warned him that GC might seriously affect the LTFD market. Dr. Pod (as his unpronounceable name was usually abbreviated) did not take this warning seriously, and even after the introduction of the first commercial GC instruments, he believed that the two techniques were complementary; however, within a short time, LTFD was completely replaced by GC.

Fortunately for Podbielniak, Seaton T. Preston, Jr., his chief chemist, took the warning of Dr. Birch more seriously and—almost clandestinely—kept himself up to date in chromatography; thus, when the GC instruments hit the market, a crash program could be initiated, and in December 1955, Podbielniak Co. was able to introduce their first instrument, the Chromacon Model 9400 (27). This was a large, floor standing instrument with U-shaped columns and a thermal-conductivity detector, using a built-in potentiometric recorder (Figure 8).

Within a year Podbielniak greatly expanded its GC line. By then the Chromacon series consisted of 12 versions, differing in the upper temperature limit (up to 300°C) and the number of columns that could be installed; instruments under reduced pressure were also available (28). The company also offered process GC instruments and large-scale preparative units with 2–4-inch-o.d. columns (29); in addition, some other types of laboratory GCs were also available. In fact, the company overextended itself; they never had the staff to efficiently handle such a large line of instruments plus the management of the Podbielniak Institute (where they were teaching customers the use of their instruments). Some major personal controversies also contributed to the decline of the company, and around 1960, they discontinued their GC product line. In a listing of suppliers of GC instruments published in its July 3, 1961 issue (30), *Chemical & Engineering News* did not mention anymore the company.

The Followers

In 1956, four companies joined the field introducing laboratory GCs: Fisher Scientific, Beckman, Consolidated Electrodynamics Co. (CEC), and Hallaikainen Instruments. Today, none of these are anymore active in GC; still, it is interesting to investigate the circumstances how these companies became involved in our field and their early products.

Fisher Scientific

Fisher Scientific was founded 100 years ago in Pittsburgh, PA, by Chester G. Fisher to supply laboratory equipment (31). The company continuously grew and eventually became the most important supply house in the U.S. Naturally, they had a close contact with various laboratories in the Pittsburgh area, among them the R&D laboratories of Gulf Oil. Scientists at Gulf started to investigate the potentialities of GC soon after the first news from England, and D.M. Lichtenfels already presented a paper on “gas–liquid partition chromatography” at the 1955 Pittsburgh Conference, which was later published in

Analytical Chemistry (32) almost simultaneously with another paper from this lab on the use of GC for the study of catalytic reactions (33). Gulf also developed a GC in cooperation with Fisher, which was then introduced at the 1956 Pittsburgh Conference (34). This was the so-called Fisher-Gulf Partitioner. In the original model the temperature of the air thermostat could be set in six steps between 50°C and 140°C; within a year the range was extended to 300°C and a flash vaporizer was added, which was the Fisher-Gulf Model 300 (Figure 9) (35). At Fisher, B.W. Taylor and A.A. Poli were particularly active in the early GC development work.

A specialty of the Fisher instrument was the so-called micro-dipper for liquid sample introduction⁶. At that time the low-volume syringes did not exist as yet, and the available syringes had a fairly large volume (50–150 µL), making the reproducible introduction of small volumes difficult. The micro-dippers were supposed to solve this problem; however, they were unreliable and cumbersome to use.

Soon after the introduction of the Gulf instrument, Fisher also carried out original development work. The result was the Model 25 Gas Partitioner, designed to replace the Orsat-type gas analysis and introduced in the fall of 1958 (36). This unique instrument utilized two columns in series, having one side of the thermal-conductivity detector between the two columns and the other side at the end of the second column. Because this technique is completely forgotten today, its operation is illustrated in Figure 10. This instrument was also modified into a specialized GC for the analysis of gases in blood.

Fisher ceased to offer the Fisher-Gulf Partitioner around 1960–61, but continued marketing the small gas analyzer for a few more years.

⁶ Micro-dippers were also offered for other GCs. In 1958, when I was in charge of a service laboratory in Germany, with two Perkin-Elmer Model 154s, I also purchased one. However, it never worked properly and particularly the reproducibility of the injected volume was very poor.



Figure 9. The Fisher Gulf Model 300 Partitioner, a slightly modified version of the original model introduced in March 1956. The box on the lower right side of the front is the micro-dipper.

Beckman Instruments

This company was founded in the 1930s by Arnold O. Beckman (37), initially to supply a pH meter of advanced design. In the 1940s, it became involved in building a number of mass spectrometers (MS) for the U.S. Atomic Energy Commission for online monitoring of the separation of ^{235}U from ^{238}U . The success led the company into other fields from which the development of the Model DK ultraviolet spectrophotometer was the most famous. This instrument was developed in cooperation with Wilbur I. Kaye—then associated with Tennessee Eastman Co.—who has also been involved there in the early GC work (11,12). Through this contact Beckman learned about GC. At that time the company was developing a special MS for stream analysis of petrochemical plants, and it became obvious that GC would be better suited for this purpose. Thus, the MS program was redirected to the development of GCs. H.J. Noebels (then the head of Beckman's Applications Engineering Group), Theron Johns, and Don W. Carle were involved in this activity, the result of which was the

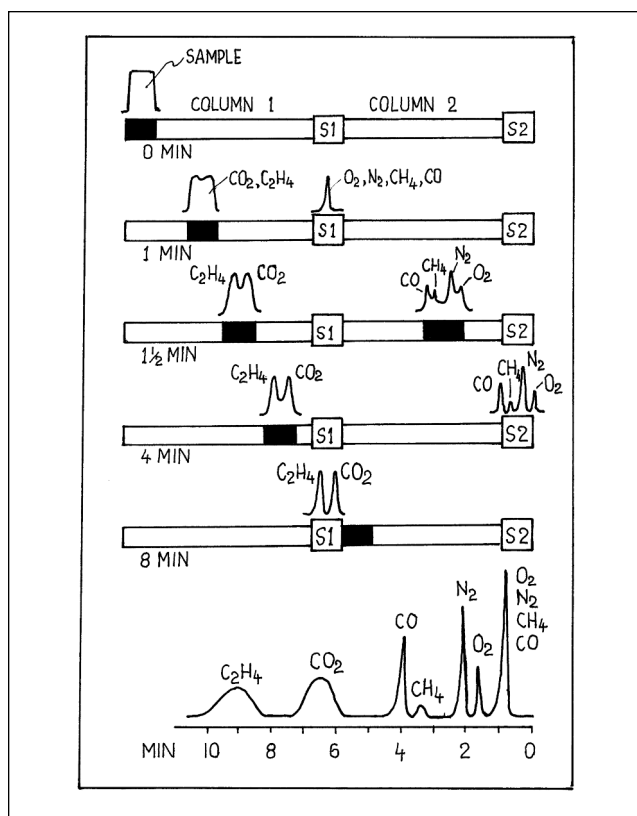


Figure 10. Functional schematic of the Model 25 Gas Partitioner from Fisher Scientific introduced in the fall of 1958. The top part of the figure visualizes the position of the individual sample components at a given time: the dark boxes indicate the bands and the degree of separation is shown by the partial chromatograms. S1 and S2 are the two sides of the thermal-conductivity detector. Column No. 2 contained molecular sieve 5A while Column No. 1 contained either silica gel or a column packing prepared with a liquid stationary phase such as 2-ethylhexyl sebacate, for example. The full chromatogram is shown at the bottom of the figure: the composite $\text{O}_2 + \text{N}_2 + \text{CH}_4 + \text{CO}$ peak and the peaks of CO_2 and C_2H_4 were recorded by S1, and the individual peaks of O_2 , N_2 , CH_4 , and CO were recorded by S2. CO_2 and C_2H_4 were irreversibly adsorbed by the second column and thus were not recorded by S2.

Model GC-1 GC introduced at the 1956 Pittsburgh Conference (38). An interesting feature of this instrument was that it utilized a flat, coiled sample instead of the standing U-shaped columns of other instruments. This fact contributed to the controversy on the proper column shape, which lasted for about 10 years⁷.

The GC-1 (Figure 11) was very simple and easy to operate; however, it had a major drawback: it could only be used at a single fixed temperature, 40°C . In order to correct this shortcoming, a new instrument, the Model GC-2, now having variable temperature capacity up to 200°C was introduced within a year (39).

Soon after the introduction of the laboratory GCs Beckman also developed instruments for process control. In addition, they also introduced a major preparative GC, the Megachrom, utilizing large-diameter columns (40). The basic drawback of this instrument was that it utilized helium as the carrier gas, which it recirculated through a purification unit. However, this instrument was withdrawn within a short time from the market.

By the second part of the 1960s, Beckman lost interest in laboratory GC; however, they remained active in the process instrumentation field for a long time.

CEC

CEC, in Pasadena, CA, has been in the MS business for some time, mainly for the petroleum industry. Thus, it was a logical extension of their activities to develop a laboratory GC. This instrument, the Model 26-201, was introduced in the summer of 1956 (Figure 12) (41,42).

The CEC instrument differed from the other contemporary GCs in that it had a modular construction with a separate control unit and analyzer, the latter containing the column oven, which could be operated at any temperature between 50°C and

⁷ The objection expressed by some was that in a coiled column, centrifugal forces might unfavorably influence separation.



Figure 11. The Model GC-1 GC from Beckman Instruments introduced in March 1956. This instrument had a single fixed temperature control at 40°C ; the Model GC-2 introduced within a year had variable temperature control possibility up to 200°C .

250°C. It is interesting to note that this instrument also employed coiled columns. The eluent fractions could be trapped after the thermal-conductivity detector for subsequent MS analysis. Special features included a heated multiport gas sampling valve and a liquid sampler.

In the following years, CEC improved their instruments, extending the upper temperature range (43). However, by the beginning of the 1960s, they diverted themselves from the chromatography line.

Hallaikainen Instruments

Besides British Petroleum Co., the laboratories of the Shell companies in England, The Netherlands and the U.S. were the first to explore GC after the seminal work of James and Martin. In the U.S., Shell Development Co. in Emeryville, CA, carried out some fundamental investigations on instrumental requirements on the standardization of certain terms and on the use of GC for hydrocarbon analysis; the results of these activities were summarized in three basic papers published in the March 1956 issue of *Analytical Chemistry* (44–46)⁸. Naturally, Shell Development Co. also developed a GC for their own purpose and they licensed Hallaikainen Instruments Co., a small company in California⁹ to build a commercial instrument, the so-called Chromagraph (48). This was the first instrument to have a separate refrigerated unit permitting column temperature down to 0°C.

The Chromagraph was sold only locally in California and had no nationwide impact; its production was ceased within a couple of years. I have never seen this instrument.

⁸ Among others, the so-called Dimbat-Porter-Stross expression for the sensitivity of the thermal-conductivity detector was the result of their work. This expression was then standardized by ASTM (47).

⁹ I was told at that time that the owner of this company was actually a former employee of Shell Development.

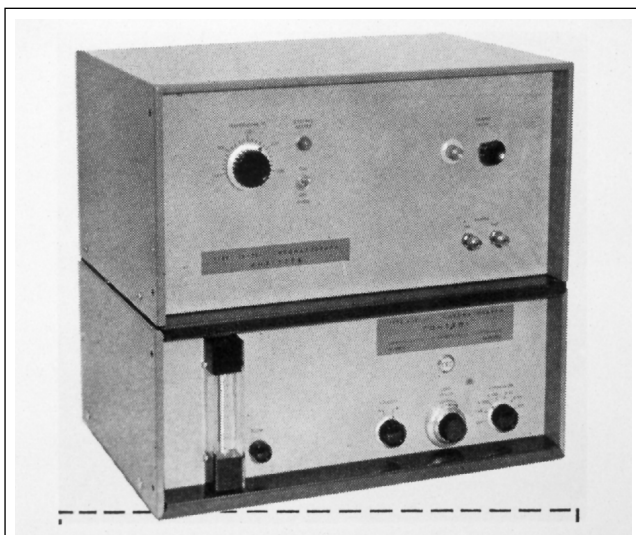


Figure 12. The Model 26-201 GC from Consolidated Electrodynamics Co. introduced in the summer of 1956. The top part was the analyzer module including the (coiled) column, the thermal-conductivity detector, a sampling valve for gaseous samples, and a flush vaporizer for liquid samples. The lower module was the control unit. It was also possible to have two analyzer modules with one control module.

The Next Generation

All the companies discussed until up to now have already been in the instrument business, and for them GC simply represented a new line. Also, their instruments followed more or less the same general principles. The three companies discussed below brought some changes in the instrument design; also, they were newcomers in the field of scientific instrumentation. Barber-Colman had been involved in the manufacture and marketing of potentiometric recorders and other electronic components; thus, at least they had some experience in electronics and basic engineering and knowledge in chromatography they learned from consultants. At the same time both Wilkens Instrument and F&M were specially formed by chromatographers to develop and produce GC instruments. Both represented real success stories, and although eventually they were acquired by large companies, their direct successors are still prominent suppliers of GC equipment. Barber-Colman also started as a success story; however, after about a decade they decided that the marketing of scientific instruments did not fit their corporate philosophy.

Wilkens Instrument & Research Inc.

This company was formed by Keene P. Dimick, a chemist at the Western Regional Research Laboratory of the U.S. Department of Agriculture (USDA) in Albany, CA; in a retrospective article he discussed in detail the circumstances leading to his involvement in GC and in company building (49). At the USDA laboratory, Dimick's project was the investigation of the volatile flavor components of strawberries. However, he could not obtain any meaningful results until, after reading the first reports on GC, he built his own instrument and applied the technique to the separation of the volatile constituents of strawberry oil (50). In the spring of 1956, Dimick presented his results in a couple of lectures, and from discussions with his peers outside his own laboratory he realized that there was a great interest in the new technique in the food/flavor field. Encouraged by these discussions he built during the summer of 1956 in his garage, together with Ken Wilkens, his brother-in-law (a high school art teacher in the Napa Valley), five GCs. It was fairly easy to sell these, and additional orders came in. As a conclusion Wilkens rented a former bicycle shop, Dimick set up a small laboratory in his house, and on December 14, 1956, together with his wife (under the pseudonym Doris Lausten) and his brother-in-law, Keene Dimick incorporated the new company¹⁰.

Their first instrument, the Aerograph Model A-90 (Figure 13) employed a thermal-conductivity detector. It was a simple machine, easy to operate, and thus particularly favored by novices in the field who simply used GC as a tool. In the next decade the company introduced a number of new instruments from which particularly two should be mentioned: one can say that these two made really the company.

The first, introduced in 1961, was the Aerograph Model 600

¹⁰ At that time Dimick was still employed by the USDA; thus, he could not use his own name for the new company. Since they used "Aerograph" as a common name for their instruments, most people did not even know the official name of the company, which was commonly called simply as "Aerograph."

Hy-Fi, a small and simple instrument with an FID (its name was concocted from the initials of hydrogen flame ionization). It became an instant success, and as mentioned by Dimick (49), soon they sold 80–100 units per month! The second big success was the Aerograph Model 700 Autoprep semi-preparative instrument (Figure 14) introduced in early 1962. The fundamental difference between this instrument and other preparative GCs was that actually the Autoprep was a laboratory instrument, in fact, almost identical to their basic model, and used regular columns; with single, manual injection, it performed just as the other laboratory instruments. However, it also permitted the repeated automatic injection of the sample and the joint collection of the same fractions from the repetitive injections. In this way the laboratory instrument could be used as a preparative GC. Within a year this instrument represented about 25% of the company's sales.

All these instruments—even the more complex models introduced in the 1960s—were fairly small. They may have been less sophisticated than some of the instruments of the other companies, but they were easy to operate by an average laboratory chemist. In a retrospect discussion (49), Dimick emphasized that this was because of a different company philosophy: his instruments were designed by chemists, chromatographers, and the engineers only contributed in making the new design easy to produce. In addition, they put great emphasis on the continuous communication with the users. We shall deal with this activity later.

Wilkins Instrument went through a phenomenal growth: from a sales volume of \$60,000 in 1957 it grew to \$3 million by 1962 and \$8.5 million by 1965. In the spring of 1966, the company was sold for \$12 million to Varian Associates and continued its operation as part of that company.

Barber-Colman Co.

To present-day chromatographers this company is completely forgotten, although in the 1960s it played an important role, mainly in the biochemical field. Its involvement in GC can be traced back to S.R. Lipsky, professor at Yale University Medical School in New Haven, CT, and to Evan C. Horning, at that

time associated with the National Heart Institute within the National Institutes of Health in Bethesda, MD (51,52).

We have to go back to the Burrell Corporation. From 1955 on, they introduced a number of models, and in 1957, they wanted to develop an instrument using glass columns for the emerging biochemical field. This was the time of the investigations linking the diet to the occurrence of heart diseases and GC seemed to be an ideal method for the analysis of fatty acids (in the form of their esters). One of the experts in this field was S.R. Lipsky at Yale, and thus Burrell engaged his cooperation as a consultant for this development. However, the new instrument, the Kromo-Tog Model K-5¹¹ had a number of problems, and Lipsky had to carry out a number of modifications of the instrument to be able to use it for the planned applications.

The Burrell instrument used Wheelco potentiometric recorders manufactured by the Barber-Colman Co., Rockford IL, and thus, Lipsky inadvertently established contact with this company. Barber-Colman was interested to become directly involved in the GC instrumentation field, and with Lipsky's help they developed a new instrument specially designed for the biochemical field. It was realized that thermal-conductivity detectors do not have the needed sensitivity; therefore, they selected the argon-ionization detector invented at that time by J.E. Lovelock (54,55), however, using a tritium titanium foil instead of ⁹⁰Sr as the radioactive source. At the last minute Barber-Colman's management almost decided against the project, but this was changed on the intervention of Evan Horning.

This instrument, Barber-Colman's Model 10 was finally introduced at the 1959 Pittsburgh Conference. It was a large, floor-standing instrument (Figure 15) using U-shaped glass columns in an air thermostat with a maximum temperature of 300°C. The liquid samples were introduced directly to the top of the column without going through a flash vaporizer.

In the 1960s, Barber-Colman was very active, mostly in the



Figure 13. The Aerograph Model A-90 GC from Wilkins Instrument & Research Inc. introduced in early 1957. A bubble flow meter was attached on the side for the measurement of the carrier gas flow rate. The injection port was located just left of the name plate.

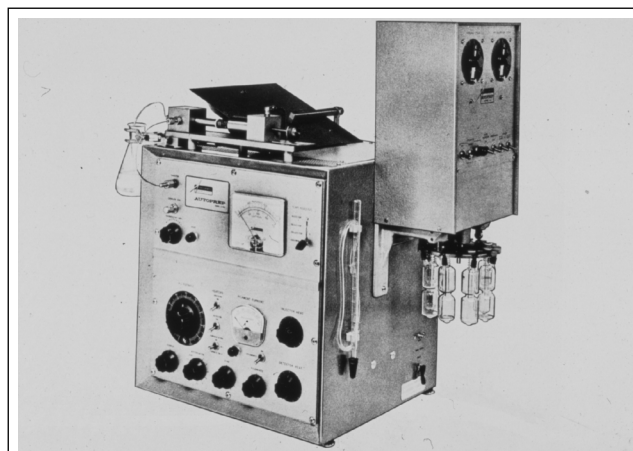


Figure 14. The Aerograph Autoprep Model A-700 GC from Wilkins Instrument & Research Inc. introduced in the first part of 1962. The device on the top of the instrument was the automatic injector and the fraction collector was on the right-hand side.

¹¹ The Kromo-Tog Model K-5 was introduced by Burrell in the first part of 1958. It employed two U-shaped columns (which could be used either simultaneously or as two independent systems) and a specially designed hot-wire detector and was also equipped with a fraction collection system (53).

biochemical field. Then, in January 1970, the company sold its GC instrument line to Nuclear Chicago, which, a couple years later, discontinued it.

It should be mentioned that in 1958–59, Jarrell-Ash Co., Boston, MA, also marketed in the U.S. the so-called Argon Chromatograph produced in England by W.G. Pye & Co., originally introduced at the International GC Symposium held in May 1958 in Amsterdam, The Netherlands. This instrument was the first utilizing Lovelock's argon-ionization detector. However, after the introduction of the Barber-Colman Model 10, the Pye instrument could not compete with it in the U.S.

F&M Scientific Co.

The second instrument company formed specially to supply GCs was F&M; it has been another success story. This company is particularly linked to the advent of temperature programming.

Programming the column temperature was first utilized in 1952 by C.S.G. Phillips (56) and in commercial instrumentation by Burrell Corp. In their instrument the thermal-conductivity detector and the column had separate temperature controls, and thus the latter could be changed without affecting detector stability.

At the 1958 Amsterdam GC Symposium, Guild actually presented a paper in which, among other things, the possibilities of column temperature programming was described¹² (57). In spite of this early work, however, practically all instruments—either home-built or available commercially—were designed for isothermal operation.

The development of modern temperature programming is usually credited to S. Dal Nogare of E.I. Du Pont de Nemours, Inc. in Wilmington, DE, who showed for the first time the advantages of linear temperature programming and automated control of the program rate (58–60). This work represented the

¹² It should be noted that in this work, column temperature was changed (increased) manually during analysis. Automated linear programming was first used by Dal Nogare and then in the F&M instrument.

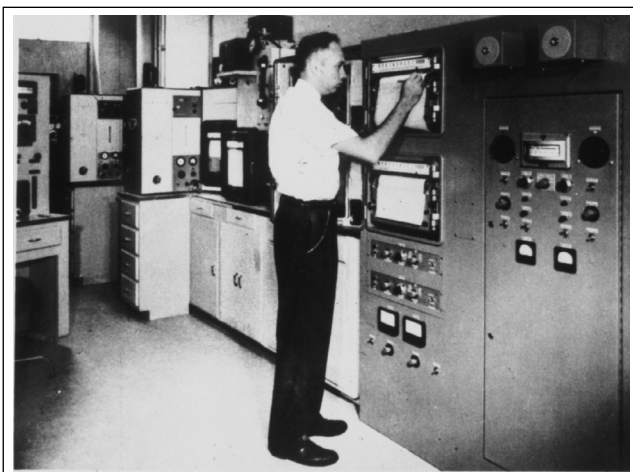


Figure 15. The Barber-Colman Model 10 GC introduced in March 1959. The instrument was equipped with a glass column and an argon-ionization detector. A number of other GCs (Model 154s) can be seen in the background. The instrument is being operated by Dr. B.M. Mitzner of International Flavors & Fragrances, Inc.

basis of the F&M Corporation.

The beginning of the company goes back to 1956 when two Du Pont glass blowers, Kraus and Martinez, were permitted by Du Pont to start a private business in addition to their main affiliation. For this work they formed a company under the name of K&M Scientific Glassware Co., representing the initials of the two owners' names; soon, however, Frank Martinez bought out his partner and changed the name to F&M, representing his own initials.

In 1956–57 Dal Nogare's group developed two special (still isothermal) instruments for internal use, one for high-temperature (61) and the other for high-sensitivity work (62). Parts of these were made of glass, and thus Martinez became involved in their construction. He continued to have close contact with Dal Nogare's group, and seeing their results on temperature programming Martinez immediately realized the great potential of the technique. Therefore, he decided to leave Du Pont and devote full time to the manufacturing and marketing of temperature-programmed GCs. He invited those who were involved in this development to join him. Two accepted this invitation and decided to join Martinez: Eugene Bennett, a research chemist in Dal Nogare's group, and Aaron J. Martin, a research supervisor. Thus, they left Du Pont, and in January 1959, jointly formed a new company, F&M Scientific Corporation. Two months later, at the 10th Pittsburgh Conference, they already exhibited the first temperature-programmed instrument, the Model 202 GC (Figure 16) (63). This instrument utilized a thermal-conductivity detector and permitted operation up to 300°C. Within a year its construction was improved (changing it to the Model 300), and then, the temperature range was also extended to 500°C (Model 500) (64). In 1960, an instrument (the Model 609) with an FID complemented F&M's product line (65).

In January 1961, ground was broken for a new plant in Avondale, PA; by then F&M already employed 50 people, and

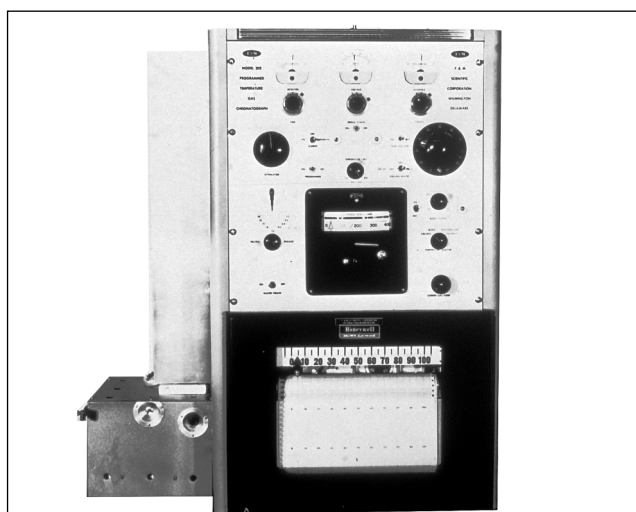


Figure 16. The Model 202 programmed-temperature GC from F&M Scientific Co. introduced in March 1959. The column was in the "chimney" (an air thermostat) on the left mounted on the box containing the thermal-conductivity detector. The injection port was just below the column.

within a few years F&M grew into a multimillion dollar company. Finally, in August 1965, it was sold to Hewlett-Packard Co., which continued its operation as a separate division. When, in July 1999, Hewlett Packard split into two separate companies, this division—now the dominant supplier of GC equipment in the U.S.—became part of the new Agilent Co.

Supply Houses

In chromatography the instrument itself is only part of the business; according to a recent survey (66) 33% is represented by companies providing the much needed supplies (gases, columns, syringes, valves, etc.) without which the proper use of the instruments would be impossible. Therefore, a discussion of the early evolution of GC would be incomplete without also dealing with the companies that provide these tools. We shall discuss here two particular fields: syringes used for sample introduction and columns and column materials.

Syringes

We have already mentioned the problem of early chromatographers with the introduction of small sample volumes: no reliable tools existed permitting the reproducible introduction of such samples into the GCs. We have discussed the situation with the so-called micro-dippers that were supposed to solve this problem: these were, however, highly unreliable. Other companies (e.g., Beckman (67) and CEC (42)) also developed special devices to improve the situation: however, these were bulky and difficult to use. Thus, the introduction of the modern microsyringes by Hamilton Co. represented a real breakthrough.

The beginnings of this company by Clark L. Hamilton were recently discussed in more detail (68). He started in 1956–58 with the 50- and 10- μ L syringes (69). Then, within a couple of years he developed the 1- and 5- μ L syringes to fulfill the demand created by the ionization detectors and capillary columns. The 1- μ L syringe really represented the ultimate in precision production; it utilized the bore of a needle (inner diameter of 0.15 mm!) as the syringe barrel with a tungsten wire of the same diameter serving as the plunger, and the subdivision of graduation was 0.01 μ L.

In the subsequent years Hamilton Co. expanded its operation by introducing a number of special syringes and other devices and later also entered the field of columns for high-performance liquid chromatography. However, after 40 years, these syringes still represent the company's major products.

Columns and column materials

At the beginning, everybody made his own (packed) columns, coating a finely divided support material with the stationary (liquid) phase. Diatomaceous earth type materials were used as the support. James and Martin in their original work (10) used Celite 545, a flux-calcinated diatomite, and the same material was also used by the groups of Tennessee-Eastman (12) and Gulf Oil (32). Celite was a product of the Johns-Manville Co. used in large quantities by the industry as a filter

aid. Naturally, the original material was not uniform and pure enough for chromatographic purposes and had to be sized and purified. Another diatomaceous earth material was firebrick, widely used in the steel industry for furnace isolation. Crushed, sized, and purified¹³ firebrick was used in the basic work of Shell Development Co. (33–36). A similar European product was the so-called Sterchamol, introduced for chromatography by the Dutch Shell group (70).

When the demand for support material increased, Johns-Manville set up a small production of crushed and sieved firebrick under the trade name of Chromosorb P (the P stood for “pink”, the color of the material resulting from impurities). Around 1958, they also prepared a more inert material named Chromosorb W (for “white”). In the early 1960s, new types were added such as a silanized version (blocking the active hydroxyl groups on the particles' surface by trimethylsilyl groups).

Parallel to the introduction of commercial instruments, out of necessity the instrument companies offered standardized packed columns to their customers. In this pioneering activity Perkin-Elmer was leading; within one year over a dozen standardized columns, with different stationary phases and specified stationary phase loading, were offered.

Turning to the stationary phases, at the beginning, standard chemicals were used for this purpose. May & Baker Ltd. of Dagenham, England, was probably the first to offer substances prepared specially for the purpose of use as liquid phases, and by 1958, they had seven such substances with the brand name Embaphase (“emba” for M&B). Then, slowly some other small companies started in this field. At first, their activities were mainly restricted to the purification of existing chemicals for chromatographic purposes; this was then followed by developing a number of new phases and supports and finally offering ready-made column packings and packed columns. In the period we are dealing with here, Applied Science Laboratories was the most prominent in this field. It was originally founded in 1951 by A. Rose, professor at Pennsylvania State University, to carry out his consulting work. At that time his main activity was the preparation and purification of fatty acids and their esters, mainly for some government laboratories (N.I.H.). When in 1958 polyester liquid phases were introduced for the analysis of fatty acid methyl esters (71,72), Applied Science started to prepare high-purity polyesters for GC purposes. It may be interesting to note that their first brochure—a two-page sheet—on pure polyester phases was published in 1960.

From this modest beginning the company expanded rapidly as a major supplier of high-purity phases and supports; their introduction of GasChrom Q, a silanized diatomaceous earth type support, in 1964 had a major impact on the application of GC in biochemistry. The company also served as the starting point for other chromatography supply houses; for example, the founders of Analabs and Supelco were originally affiliated with Applied Science Laboratories. The start of these and other

¹³ In 1957–58 when I was in charge of a service laboratory in Germany with three GCs, we had to make our columns. In order to purify Sterchamol (to get rid of the mostly iron impurities), we extracted the raw material with HCl. I remember that we had a Soxhlet extractor continuously operating in the corner of the laboratory, preparing the purified support.

companies in this field, however, is beyond the scope of this report.

Dissemination of Technical Information

In the case of a new technique it is always crucial to have as much information as possible available to those who want to apply the new technique; they have to learn the principles of the method, the proper handling of the instruments, the selection of the operation variables, and how specified methods can be developed to solve the individual problems. Practically from the beginning, the instrument companies had a major role in these fields. In fact, Ralph Müller had a special praise for these activities, saying that:

In our opinion, the instrument manufacturers of this country are doing a splendid and increasingly important job in disseminating scientific and technical information (17).

With the introduction of the first GCs a number of companies also provided the analysts with simple texts, explaining the technique and its applications in concise form. For example, Hausdorff's 31-page brochure (73), the two-part introductory paper of Podbielniak and Preston (74), and Theron Johns' manual (75) served for thousands as the basis on which they built up their knowledge¹⁴.

This activity continued in the years ahead. GC courses were regularly held by the companies, and periodically they also published application notes and data sheets providing new information. Podbielniak Co. even founded in 1956 an institute in Chicago in which two-week intensive courses on GC (and analytical distillation) were offered.

As already mentioned previously, the dissemination of technical information became a very important factor in the success of Wilkens Instrument & Research Co. A few months after establishing the company Dimick started the publication of *Aerograph Research Notes*, a quarterly publication that was eventually mailed to 15,000 chemists. Its style was best characterized by Karasek who noted that "*Research Notes* appeared to be talking directly to the chemist, conveying results much as one would in an informal bull session" (77).

When speaking about the dissemination of technical information, a few words must be said about the Applications Engineering Groups existing at that time at most instrument companies. In fact, *Analytical Chemistry* considered their function so important that in 1961 it devoted one of the Reports for Analytical Chemists to the key role of these groups (78). It is worthwhile to quote from this report:

The laboratory instrument ... is the product of almost continuous application engineering which begins during devel-

opment of the instrument and which continues behind the scenes as long as the instrument is produced.

The Applications Engineer represents the customer's viewpoint within the corporation and (the corporation's) technology to the customer.

The Applications Engineering Section also represents that group which is best identified by the customer as being representative of the technical face of the Corporation.

Today, now that GC is a mature technique, this role is less important, and in the large companies its functions are usually divided among various groups. However, one cannot discuss the early period when GC underwent its meteoric rise without re-emphasizing the role of the Applications Engineering Groups in the early development of the technique.

The Evolution of the Business of GC

As mentioned in the Introduction, the scientific instrument industry essentially evolved after World War II when companies active in supplying high-precision systems for the Allied military and war efforts were looking for new fields in which they could turn their energy. This new branch of the industry grew very rapidly and, according to Fahr (79), by 1975 its yearly sales volume reached about \$1 billion. On the other hand, according to a recent report (66), by 1999 the worldwide scientific instrument market was worth \$17.709 billion, representing an increase by a factor of 17 in 25 years.

Our interest is GC and it is worthwhile to compare data concerning the growth in our field. A 1961 survey by *Chemical & Engineering News* (30) estimated the annual GC business to be close to \$4 million, and for the mid-1970s, conservative estimates put the annual GC business over \$100 million (2). In contrast, a detailed current survey (66) indicated the GC business (without GC-MS) at \$1.039 billion in 1999. This information agrees well with the value given for the same year by *Analytical Chemistry* (80).

A problem with these data is that they do not include GC-MS systems; these are usually considered as a separate field by the surveys. Today, however, almost every fourth GC unit is integrated with an MS serving as its detector, and thus we cannot neglect this field. It should be noted that at the beginning of the 1960s, GC-MS practically did not exist, and even ten years later it barely reached a few million dollars.

Table I lists the market value of GC and GC-MS for the year 1999 based on the data given in the quoted survey (66). "Aftermarket" refers to the purchase of syringes, columns, gases, and other materials needed for the operation of the instruments, and "service" refers to the expenses needed to maintain the equipment in good shape. Table II indicates the types of laboratories in which these systems are used. It is particularly interesting to note that 30% of the GC-MS systems are used in environmental (e.g., EPA) and other government laboratories.

It is a legitimate question to ask how much of this enormous increase in the business in the last 40 years is real; in other

¹⁴ When I started in GC in 1957, I also learned the technique and its intricacies from these brochures, which dealt with the principles of the technique in a simple, easy-to-understand language. These brochures were remarkably free of any bias toward a specific instrument and could be used by anybody, regardless of what instrument was used. Let us not forget that the more detailed textbook of Keulemans (76) was only published in 1957, and it was too theoretical for the beginner; and, last but not least, we should mention that these brochures were readily available, free of charge!

words, how did inflation, the increase of prices, influence the growth. It is very difficult to directly compare prices because present-day instruments provide much more in performance, flexibility, and automation¹⁵. Around 1960, the price of a simple, isothermal GC with a thermal-conductivity detector was about \$1200, and a kit permitting the addition of an FID and the use of capillary columns added about \$2800 to the price. By the mid-1970s, the price of a temperature-programmed GC with dual FID and differential amplifier equipped for capillary column work was around \$5000–\$6000. As a contrast, a present-day automated laboratory GC for capillary column work, with an FID, costs about \$12,000–\$14,000. Naturally, to these prices one must add the cost of a recorder or data system, but this was also true 40 years ago. Around 1960, a potentiometric recorder cost about \$1000, and the price of an electric integrator (providing only peak area) was also around \$1000. The first electronic integrators (introduced in the early 1970s), which besides recording peak areas and retention times could also calculate calibration factors and normalized peak area, cost around \$4000–\$5000, almost the price of the GC. Today, for this amount one can buy a modern computerized data system!

The only modest change in the prices can also be seen in the prices of the ancillary devices. For example, while the price of a 10- μ L Hamilton syringe was \$18.00 at the time of its intro-

duction, today the same syringe costs \$19.80 (82).

Naturally, the meteoric rise in the use of GC cannot continue forever. The field matured, and because of the parallel development of other separation techniques (e.g., high-performance liquid chromatography or, more recently, electrochromatography), some samples, which 40 years ago were analyzed by GC, are now in the domain of these other techniques¹⁶. Therefore, the yearly growth in GC is now very small, not like it was 25 years ago. The already quoted survey of the analytical instrument industry (66) indicated only a 16% growth for the five year period of 1999–2004, and more recent estimates predict GC revenues to grow less than 2% per year (83). At the same time, GC–MS sales are predicted to grow at a much higher rate, close to 6%.

Naturally, however, these predictions are based on an essentially unchanged technology. On the other hand, there are clear signs that within a few years the design of our instruments may radically change, and this will undoubtedly increase the demand for new systems. Essentially, this change will result in “miniaturization”, reducing the overall size of the instruments. Here we do not refer to the “instrument on a chip” concept; at present, this still seems to be too revolutionary. On the other hand, the technology is readily available to significantly reduce the size of our instruments, using much shorter capillary columns with smaller diameters, thus speeding up in this way the time of one analysis. Another concept seeming to gain ground is to design the instruments in a modular way with easily interchangeable components. However,

much R&D work is still needed to adapt our instruments and their components (e.g., injectors and detectors) to these new concepts.

Finally, it is interesting to compare the evolution of the various companies in the last 40 years. A 1961 survey (30) listed 23 companies offering GC equipment, each trying to compete with the others. Slowly, the smaller companies disappeared, and by the 1970s, the field was dominated by a few companies only. The recent survey (66) lists five instrument companies dom-

inating the field: Agilent (the successor of Hewlett-Packard and through it of F&M), PerkinElmer, Varian (the successor of Wilkens Instrument), Shimadzu, and Thermo Electron, which is a conglomerate formed from the merge of a number of companies. This process, consolidating smaller companies, is also evident in the field of column supplies. As mentioned earlier, in the first years the instrument companies acted—out of necessity—as their own supply house, providing columns to their customers. Then, in the 1960s and 1970s, independent companies were established (e.g., Supelco, J&W, and Chrompack), which essentially took over this business. These supply houses represent an important segment of the business; the quoted survey (66) estimated for 1999 their sales volume as around 30% of the total GC and GC–MS business.

¹⁵ This is the question whether one can compare a typewriter with a personal computer, because both can be used to write a letter!

Table I. World Market of GC and GC–MS in 1999*

	GC		GC–MS		Total	
	\$ (million)	%	\$ (million)	%	\$ (million)	%
Initial systems	625	60.2	276	71.9	901	63.3
Aftermarket	340	32.7	68	17.7	408	28.7
Service	74	7.1	40	10.4	114	8.0
Total	1039	100.0	384	100.0	1423	100.0

* After reference 66.

Table II. Distribution of the GC and GC–MS Market in 1999 According to the Type of Laboratories*

	GC (%)	GC–MS (%)
Government laboratories	18	17
Environmental testing	18	13
Independent testing	17	6
Organic chemistry	23	10
Academic institutions	8	12
Petrochemical laboratories	7	9
Pharmaceutical laboratories	7	7
Agriculture/food	6	9
Others	14	17
Total	100	100

* After reference 66.

¹⁶ For example, around 1960–62, the possibility to analyze steroids by GC (as derivatives) represented a real breakthrough. Today, however, nobody would use GC for their analysis; HPLC is the method of choice.

The newest trend seems to be that the large instrument companies are starting to acquire the independent supply houses: Varian acquired Chrompack (a major European column supply house) in 1997 and Agilent acquired J&W in 2000. In this way we seem to be back at the original organization of our field.

The meteoric rise of GC in the past 40 years was unparalleled in the history of science; within a few years it became a dominant technique. By the second part of the 1960s it started to branch out, initiating the development of other chromatographic techniques (e.g., HPLC and electrochromatography). This phenomenal growth would not have been possible without the major contribution of the instrument companies. Therefore, today, on the occasion of the 50-year anniversary of the first seminal paper by James and Martin on gas-liquid partition chromatography, we should pay tribute to their contributions.

Acknowledgments

When in 1977 I wrote my original study on the contributions of the American instrument companies (2), besides my own recollections and original company literature (which at that time I still possessed), I could rely on information provided by a number of pioneers involved in GC instrumentation since its early years. At the end of the present review I would like to list them, as a lasting memory to their contribution. None of them are active today, and unfortunately many are not anymore among us. In this alphabetical list I indicate the companies where they have been active during the heydays of GC: J.E. Baudean (Perkin-Elmer), F. Baumann (Varian Aerograph), L.V. Guild (Burrell Corp.), C.L. Hamilton (Hamilton Co.), H.H. Hausdorff (Perkin-Elmer), R.S. Henly (Applied Science Laboratories), T. Johns (Beckman), S.R. Lipsky (Yale University Medical School), L. Mikkelsen (F&M), D.M. Ottenstein (Johns-Manville and Supelco), D.L. Petitjean (Barber-Colman), A.A. Poli (Fisher Scientific), S.T. Preston, Jr. (Podbielniak), W.R. Supina (Applied Science Laboratories and Supelco), E.S. Titlar (Applied Science Laboratories), and R. Vilalobos (Beckman). Special thanks are also due to Mrs. Eleonore Dempsey and Mrs. Debra Kaufman, librarians at Perkin-Elmer, for their help in hunting down some original literature sources.

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