Investigation of Essential Oils and Synthetic Fragrances Using the Dynamic Gas Discharge Visualization Technique

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I. INTRODUCTION

The problem of detecting individual differences of chemically similar liquids remains to be unsolved for various areas of the natural sciences, such as medicine, biology, nutrition, and cosmetics[1]. This problem is of crucial importance for the cosmetic sciences, including perfumery and aromatherapy. For example, some certain subtle differences of smell and taste between chemically similar liquids are very difficult to detect by using conventional methods of analysis, such as gas chromatography (GC) and the like (except under unique advanced techniques that are expensive, time consuming, and are not readily available). However, a trained human nose and/ or mouth of a perfumer and/ or taster can detect differences of that kind.

There are two conventional ways to make a fragrance. It can be replicated via synthesis from inexpensive petrochemical sources using precise organic chemical synthesis techniques. On the other hand, a fragrance can be naturally derived, predominantly from a plant source. In both cases, innovative methods are being applied in order to get the purest ingredient that is necessary for obtaining the desired fragrance. In many cases, such methods include the use of conventional chemical and physical analytical tools that by themselves cannot distinguish between given ingredients of natural and synthetic origin. It has also been assumed that during their development and processing, both natural and synthetic ingredients used to create fragrances have to go through different biological, physical, and chemical pathways. These pathways can leave some informational and energetical traces that are implied to the ingredients from which they are being made.

The technique of the research of liquids by way of investigating the characteristics of the gas discharge around the drops of those liquids has been shown in previous works[2,3]. These works demonstrated that strong electrolyte solutions, such as NaCl, KCl, $NaNO_3$, and KNO_3, have differences in the characteristics of the gas discharge images (GDV [Gas Discharge Visualization]- grams) around the drops of liquids. These differences were found in both cases in comparing differences between neighboring concentrations of one electrolyte solution and between the same concentrations of various electrolyte solutions.

In this article, the new method of Dynamic GDV- graphy is explained and demonstrated. Specifically, this new technique has shown high sensitivity to measure and analyze the subtle energetical changes of different subjects. GDV (or Gas Discharge Visualization) is a technique based on the Kirlian Effect, which is a physical phenomenon observed as a glow around the subject or subjects placed in certain electromagnetic field (EMF) conditions.

The sensitivity of this technique allows for one to register the differences between these natural and synthetic pathways. In addition, significant differences can be found through such studies that have shown high reproducibility.

II. METHODS OF RESEARCH

The investigated subject (a liquid drop) was placed on a glass plate or suspended at a distance of 3 mm from the top surface of the plate while covering the electrode[3]. The volume of the liquid was about 4 X 10^{-3} mL. The temperature was kept in the interval of 22.0°C to 22.5°C. The relative humidity was maintained at 42% to 44%. The train of triangular bipolar 10 microsecond electrical impulses of amplitude 3 kV, at a steep rate of 10^6 V/s and a repetition frequency of 10^3 Hz, was applied to the electrode, thus generating an electromagnetic field (EMF) around the drop. Under the influence of this field, the drop produced a burst of electron-ion emission and optical radiation light quanta in the visual and ultraviolet light regions of the electromagnetic spectrum. These particles and photons initiated electron-ion avalanches, which gave rise to the sliding gas discharge along the dielectric surface[4]. A spatial distribution of discharge channels was registered via glass plate by the optical system with the Charge Coupled Device TV Camera, and then digitized in the computer.

Drops were exposed to an EMF for 10 seconds, and short gas discharge "films" were recorded in the computer as "AVI" files. The frame rate (frequency of record) was set at 30 images per second, as dictated by the speed of the camera/ computer interface. All "AVI" files were then converted into a series of "BMP" files. The area (the number of light-struck pixels) and the averaged intensity (ranked from "0" for absolute black to "255" for absolute white) parameters for every image were then calculated by the software. The time series was averaged on 10 measurements that provided the statistical reliability at the confidence level of 95%. Hence, the Fisher criteria[5] was utilized for the comparison of the average values of different time series at a specified time.

III. RESULTS AND DISCUSSION

This investigation addresses the peculiarities of the dynamics of GDV curves of weakly conducting liquids. This method allows for the detection of subtle energetic differences between the same chemical compounds derived from certain essential oils and other weakly conducting liquids. Forty-two pairs of such oils and liquids have been studied. In the present research, the focus was on weakly conducting liquids that were represented by essential oils and their synthetic counterparts (liquids), of which the same chemical compound or ingredient was naturally and synthetically derived, respectfully. An example of this case-scenario is that of the investigation of Oil of Bitter Almond versus Synthetic Benzaldehyde.

Oil of Bitter Almond was chosen as a material of comparison to Synthetic Benzaldehyde because it shows a chemistry that is quite similar to that of Synthetic Benzaldehyde. Analysis by gas chromatography (GC) has proven this statement; both chromatograms for the respective materials are statistically identical (see Figure One). Specifically, Oil of Bitter Almond contains about 95.00% pure natural benzaldehyde[6], while Synthetic Benzaldehyde (which is commercially available as is as a result of organic chemical syntheses techniques) is 99.00+% to 99.5+% pure[7].

However, given this information, GC analysis by itself has its limitations. That is, it cannot reveal the subtle differences between the two materials (however, once again, except under unique advanced techniques that are expensive, time consuming, and are not readily available). This is why analysis by Dynamic GDV-graphy has been applied to this situation.

The comparison between Oil of Bitter Almond and Synthetic Benzaldehyde was studied in three sessions. Every session had a certain voltage with values of 1700 V for the first session, 2000 V for the second session, and 2500 V for the third session. For this case-scenario, ten realizations for both Oil of Bitter Almond and Synthetic Benzaldehyde were measured.

For the present research, data from the first session (as an example) will be presented. Figure Two shows a graph for the averaged realizations of the averaged intensity parameter for

Oil of Bitter Almond and Synthetic Benzaldehyde, taken during this first session of the experiment. Fisher's Test was used at 95% Confidence Intervals for every moment of registration of the Dynamic GDV processes for the two liquids. The graph clearly demonstrates that the average values of the realizations for Oil of Bitter Almond and Synthetic Benzaldehyde show a statistically significant difference for the averaged intensity parameter after four seconds of measurements.

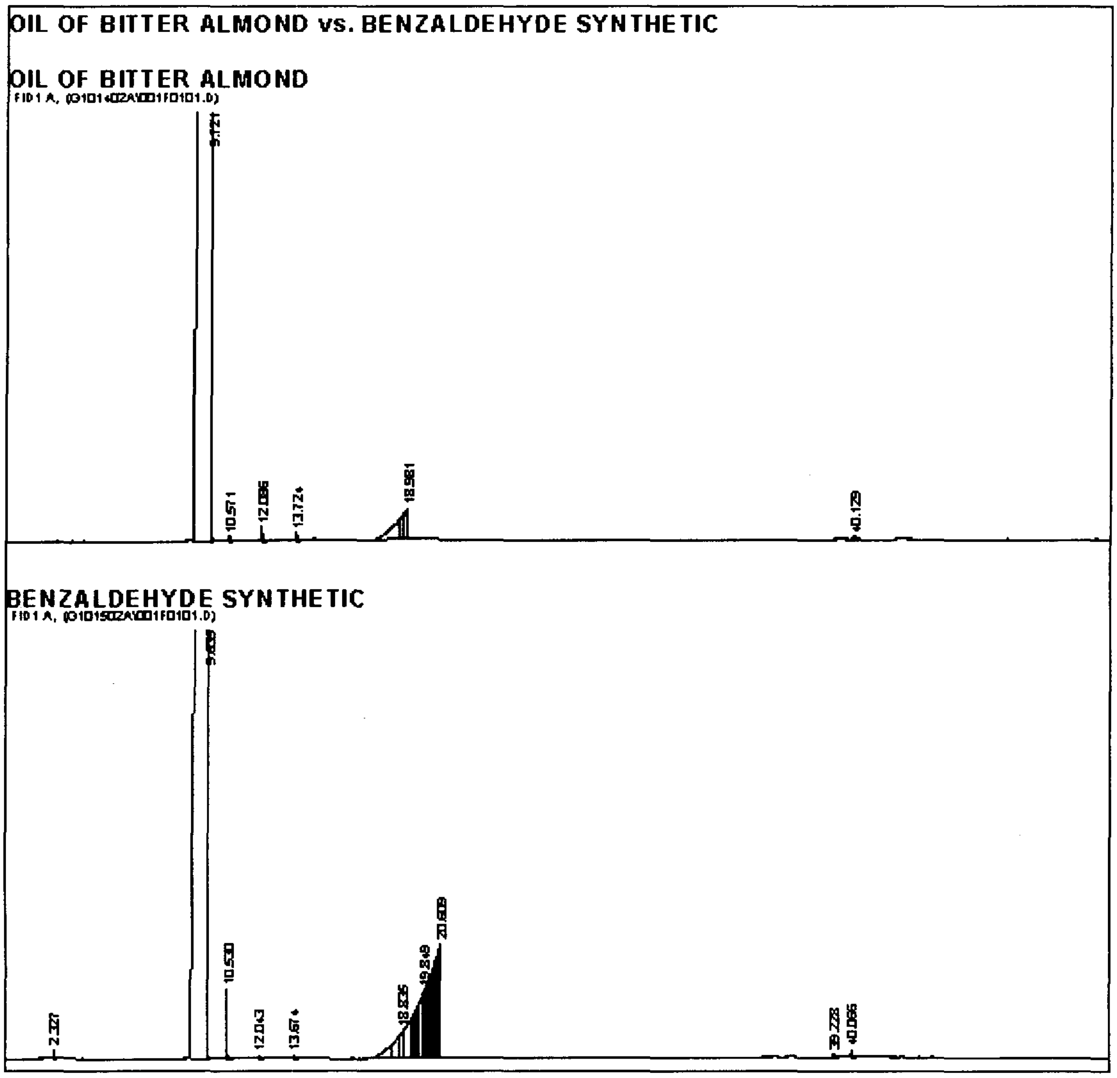

Figure One: An example of gas chromatography (GC) data analysis. Presented here are gas chromatograms of Oil of Bitter Almond (the top chromatogram) and Synthetic Benzaldehyde (the bottom chromatogram).

The main trends of these experiments for these two liquids revealed a higher position of realizations for Oil of Bitter Almond and a lower position of realizations for Synthetic Benzaldehyde for the averaged intensity parameter. In later experiments, it was shown that in terms of the area parameter, the trends were reversed for the same two liquids. That is, a higher position of realizations for Synthetic Benzaldehyde and a lower position of realizations for Oil of Bitter Almond were revealed in the case of the area parameter. Therefore, the results show that the same chemical ingredient in both materials (which is Benzaldehyde for this situation) can be distinguished from one another by using Dynamic GDV-graphy.

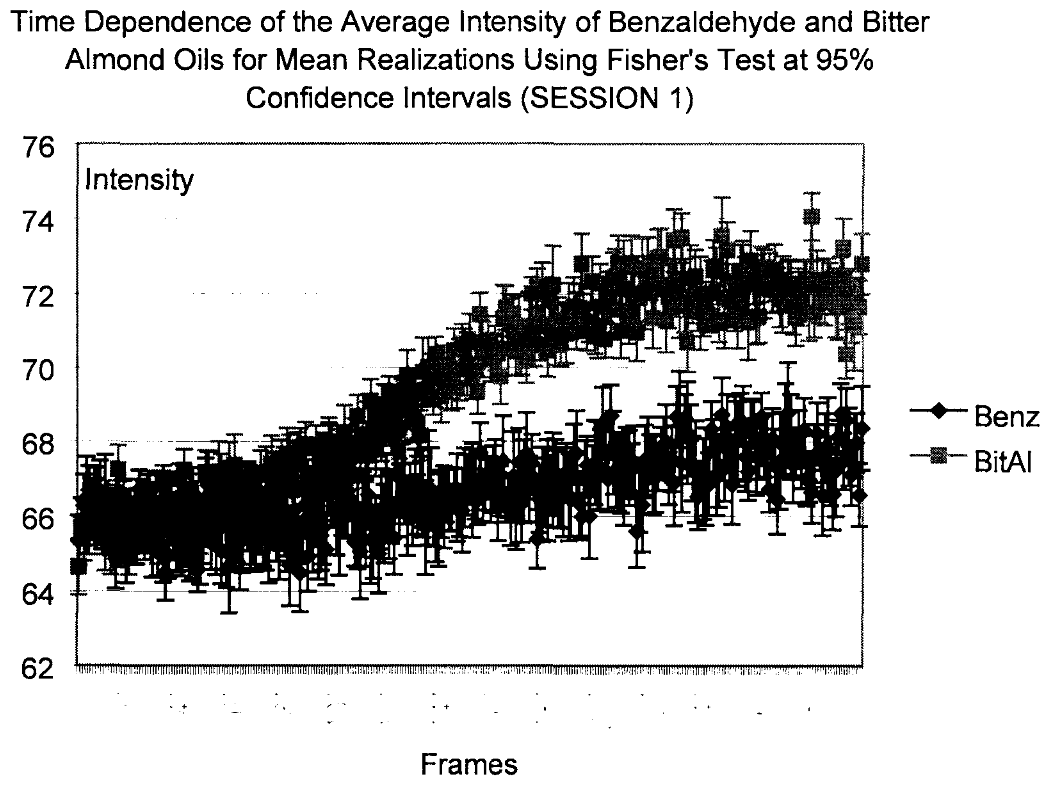

Figure Two: A graph showing the averaged realizations of the averaged intensity parameter for Oil of Bitter Almond and Synthetic Benzaldehyde, taken during session one (at 1700 Volts) of the experiment. Fisher's Test was used at 95% Confidence Intervals for this analysis.

Another example of the previously established case-scenario is that of the investigation of Natural Benzyl Acetate versus Synthetic Benzyl Acetate. Regardless of their origins, both ingredients are chemically identical, each having a purity greater than or equal to 98%. Once again, analysis by gas chromatography (GC) has proven this statement; both chromatograms for the respective materials are statistically identical (see Figure Three).

However, GC analysis by itself cannot reveal the subtle differences between the two materials (however, once again, except under unique advanced techniques that are expensive, time consuming, and are not readily available). Therefore, the results obtained by Dynamic GDV-graphy for the two materials will be presented.

The comparison between Natural Benzyl Acetate and Synthetic Benzyl Acetate was studied in one session, which was conducted at 2000 Volts. For this case-scenario, ten realizations for both ingredients were measured. Figure Four shows a graph for the averaged realizations of the glow area parameter for Natural Benzyl Acetate and Synthetic Benzyl Acetate, taken during the second session of the experiment. Fisher's Test was once again used at 95% Confidence Intervals for every moment of registration of the Dynamic GDV processes for the two liquids. The graph clearly demonstrates that the average values of the realizations for the two liquids show a statistically significant difference for the glow area parameter all throughout the measurements.

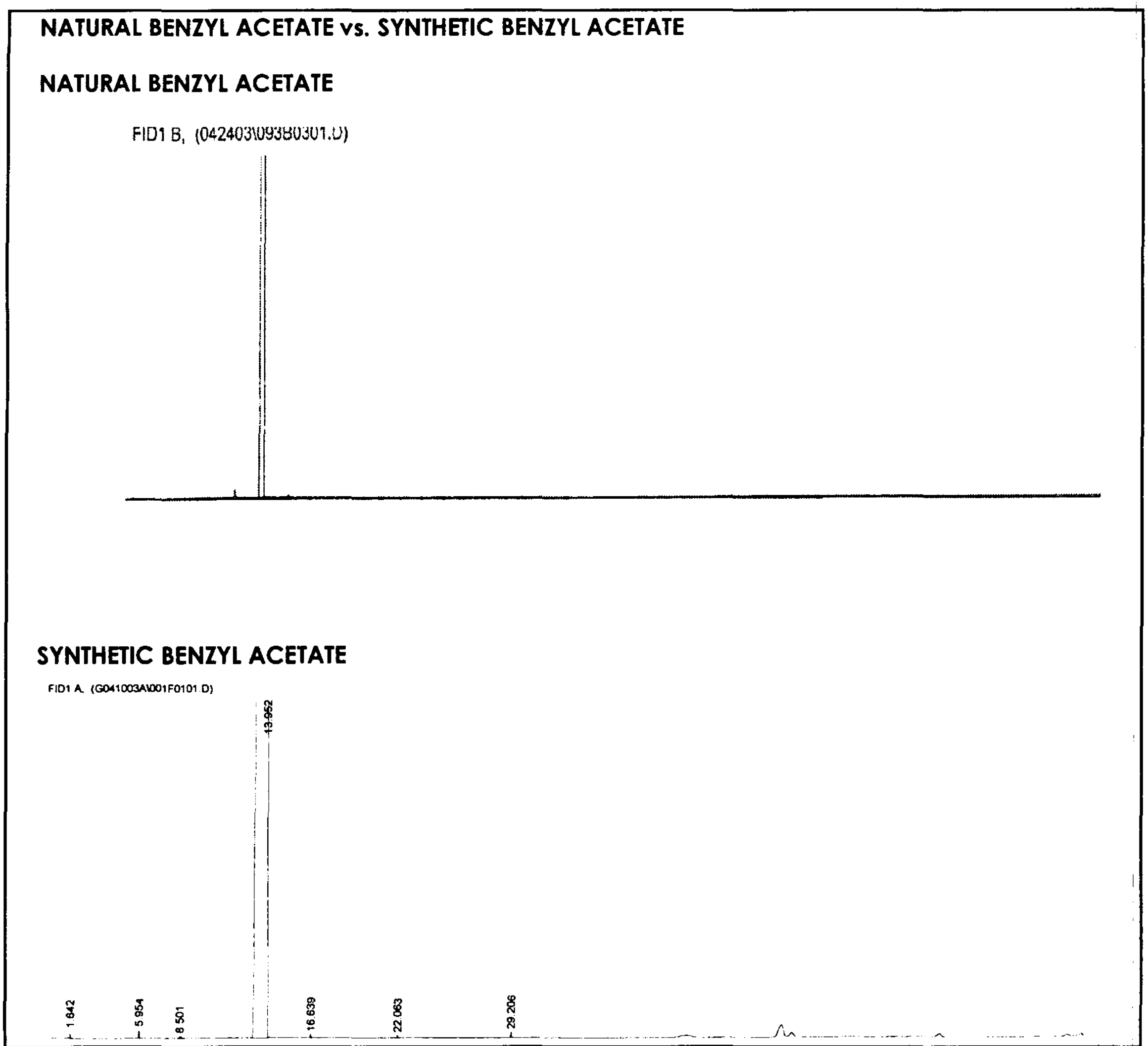

Figure Three: An example of gas chromatography (GC) data analysis. Presented here are gas chromatograms of Natural Benzyl Acetate (the top chromatogram) and Synthetic Benzyl Acetate (the bottom chromatogram).

The main trends of these experiments for these two liquids revealed a higher position of realizations for Natural Benzyl Acetate and a lower position of realizations for Synthetic Benzyl Acetate for the glow area parameter. In later experiments, it was shown that in terms of the averaged intensity parameter, the trends were the same for the same two liquids. That is, a higher position of realizations for Natural Benzyl Acetate and a lower position of realizations for Synthetic Benzyl Acetate were revealed in the case of the averaged intensity parameter. Therefore, again, the results show that the same chemical ingredient in both materials (which is

Benzyl Acetate for this situation) can be distinguished from one another by using Dynamic GDV-graphy.

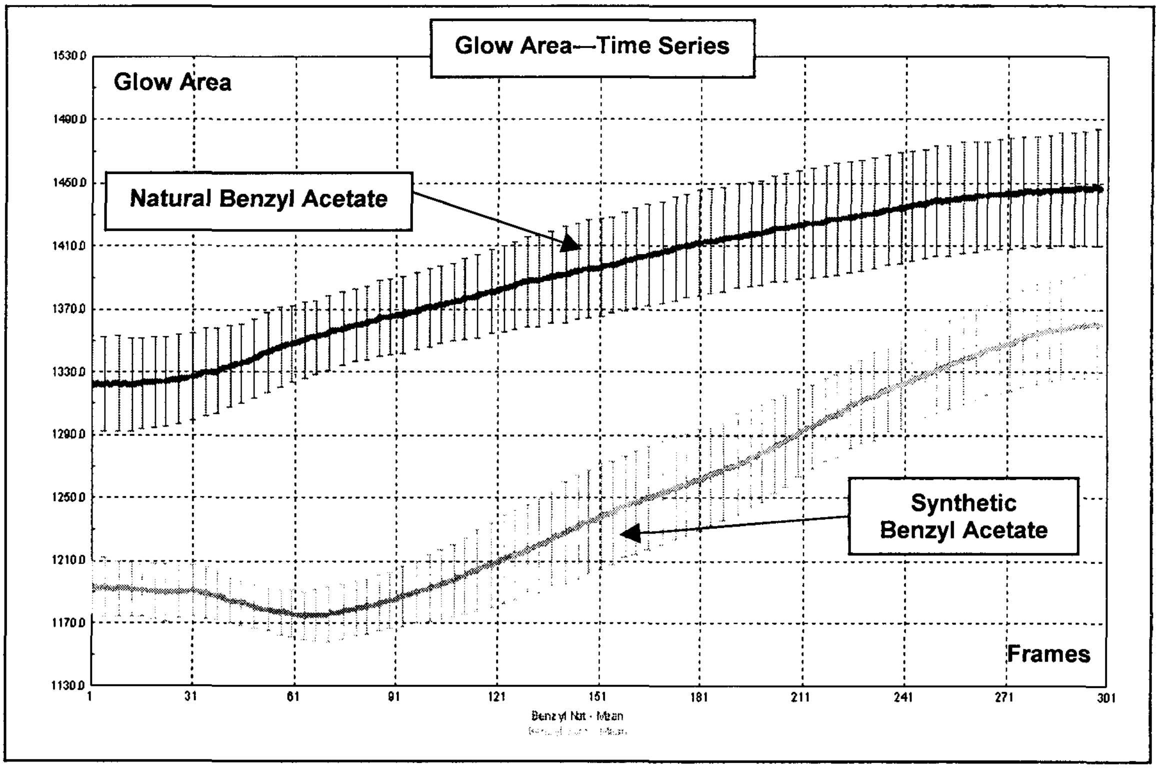

Figure Four: A graph showing the averaged realizations of the glow area parameter for both Natural and Synthetic Benzyl Acetate, taken during session two (at 2000 Volts) of the experiment. Fisher's Test was used at 95% Confidence Intervals for this analysis.

In order to understand the previously mentioned results, it is necessary to investigate the whole complex of physio-chemical processes that take place in the system—liquid, gas discharge plasma, and EMF.

After the start of the ionization process, a channel of plasma starts to appear in the vapors of the investigated liquid. The generation of chemically active particles then takes place in this channel in the gaseous phase under the influence of electrons that are amplified by the field characteristic of the impulse series. Penetration of the discharge current through the liquid initiates potential jumps at the interface of the plasma and solution. Consequently, neutral and charged particles were transferred from the solution to the plasma through the interface. This is the emission of the charged particles.

Changes that took place in the investigated liquid led to changes in the contents and configuration of the plasma. As a result, the corresponding gas discharge characteristics, such as the GDV-gram area, were also changed. EMF strength, as well as evaporation, pressure, and temperature, influenced the parameters of gas discharge[4]. Physio-chemical effects in the liquid, plasma, and interface, as well as the previously mentioned forces and evaporation, determined the dynamics of the gas discharge. Therefore, the structure of the time series of the gas discharge area was also changed. In addition, it can also be assumed that the reproducible differences of the GDV parameters of the time series of area and averaged intensity for oils and liquids of different origin became apparent.

IV. CONCLUSIONS

The investigation of weakly conductive liquids, such as essential oils and synthetic fragrances, demonstrated that the Dynamic GDV technique does reveal statistically significant differences. Specifically, it does so when comparing a wide spectrum of the similar liquids, in cases where traditional techniques (such as gas chromatography) were not effective (except under unique advanced techniques that are expensive, time consuming, and are not readily available). Differences were shown in the changes of the type of deterministic components of the time series of GDV-gram area and averaged intensity. The data also demonstrated the high reproducibility of the Dynamic GDV technique.

The dynamical study enabled one to find the subtle differences in the characteristics of low-conductive liquids (the essential oils and their synthetic [liquid] counterparts) having similar chemical contents, but each having different origins. For example, this is what was observed in the investigations of Oil of Bitter Almond versus Synthetic Benzaldehyde and Natural Benzyl Acetate versus Synthetic Benzyl Acetate, in which the same chemical ingredient (that is, Benzaldehyde and Benzyl Acetate, respectively) was naturally and synthetically derived, respectively. In all cases, there was no difference between the chemical spectra of the studied pairs of oils in accordance with the gas chromatography data.

With this new technique of Dynamic GDV- graphy, 42 pairs of oils and liquids have been investigated, while this concept is still being researched and utilized. From these 42 pairs of oils and liquids, there were 33 cases in which statistically significant differences in the dynamical characteristics were found. In 10 cases, there was a difference in both the area and averaged intensity time series data. In 12 cases, the only difference existed in area, and in 11 cases, the difference existed in averaged intensity.

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[7] Information on Benzaldehyde (Product Numbers 41, 809-9 and B133-4) Taken from the 2003-2004 Aldrich® Handbook of Fine Chemicals and Laboratory Equipment.