

The Research of the Time Dynamics of the Gas Discharge Around Drops of Liquids

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I. INTRODUCTION

The problem of detecting individual differences of chemically similar liquids, as well as highly dilute aqueous electrolyte solutions, remains to be unsolved for various areas of the natural sciences, such as medicine, biology, nutrition, and cosmetics¹. For example, some certain subtle differences of smell and taste 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.

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, NaNO₃, and KNO₃, 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 the present article, it is shown that regular dynamics of the change of the gas discharge characteristics is observed when the drops of both conductive and non-conductive liquids are exposed to an electromagnetic field (EMF) within two seconds to 10 seconds.

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 electrode³. The

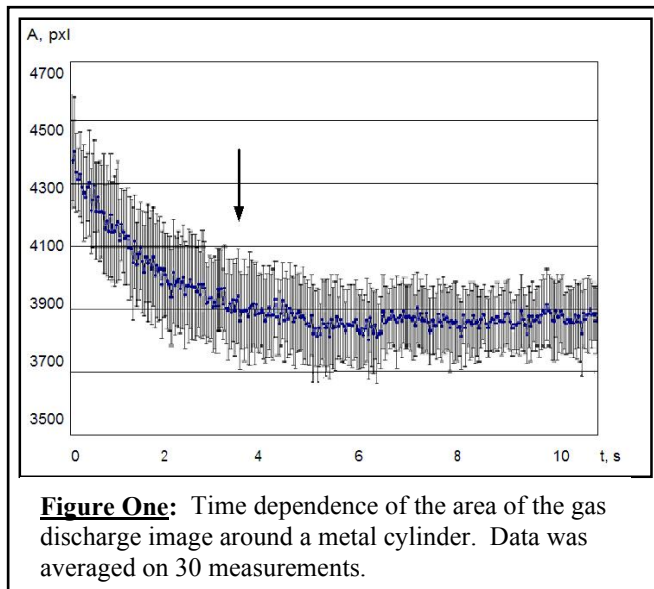
volume of the liquid was about 4×10^{-3} mL. The temperature was kept in the interval of 22.0°C to 22.5°C. The relative humidity was maintained at 42% to 44%. The train of triangular bipolar 10 microsecond electrical impulses of amplitude 3 kV, at a steep rate of 10^6 V/s and a repetition frequency of 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 surface⁴. A spatial distribution of discharge channels was registered via glass plate by the optical system with the Charge Coupled Device TV Camera, and then it was digitized in the computer.

Drops were exposed to an EMF for two seconds to 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 converted into a series of “BMP” files, and the area (the number of light-struck pixels) and averaged intensity (ranked from “0” for absolute black to “255” for absolute white) parameters for every image were calculated by the software. The time series was averaged on 10 measurements that provided the statistical reliability at the confidence level of 0.95 with an experimental sensitivity of 75%.

In the current work, the deterministic trends of the averaged dynamical time series were being studied. Noise filtration and smoothening were done with the sliding average method. The Fisher criteria⁵ was utilized for the comparison of the average values of different time series at a specified time.

The reproducibility of the method was verified by measuring the time dynamics of the gas discharge around a metal cylinder that was 10 mm in diameter, and was placed in the center of the optical lens. Thirty subsequent measurements provided statistical reliability at the confidence level of 0.95 with an experimental sensitivity of 95%. As shown in Figure One, deviation at every point of the time series is less than 5%. The same level of deviation was obtained for most of the studied liquids.

Monotone decreasing of the area at the initial seconds of measurement is due to the electro-positive discharge accumulation that takes place at the surface of the glass lens-electrode where the discharge is occurring. As was demonstrated by computer modelling^{6, 7}, an electrical field created by positive ions decreased the applied EMF so that the length of the discharge streamers decreased in every subsequent discharge. It should be noted that the frequency of both the impulses and subsequently the discharges were 1000 Hz. After some



time, when the dynamic equilibrium between the speed of the positive ions accumulated, their neutralization and leakage along the surface was established, and the length of the streamers became quasi-constant. This moment is denoted by the arrow according to the graph of Figure One.

III. RESULTS

The analysis of the results of the research will be broken down into two parts. The first part will discuss the investigation of the peculiarities of the dynamic characteristics of the GDV (Gas Discharge Visualization) process of conducting liquids (specifically, strong electrolytes). The method involved in this investigation helped to improve the results of the statistical analysis of the parameters of single GDV-grams³.

The second part deals with the investigation of the peculiarities of the dynamics of GDV curves of weakly conducting liquids. The method involved here allowed for detecting the 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.

The Analysis of the Data on the Investigation of Strong Electrolytes—the Conducting Liquids

For strong electrolytes, the dynamical curves display the same behavior as that of the same metal cylinder that was used to verify the reproducibility of the Dynamic GDV- graphy method. It was found, that the area of the GDV image is proportional to the concentration of the strong electrolyte (see Figure Two). The approximation of the trends of time series is given by the following power function:

$$y = Ax^\alpha + B. \quad (1)$$

According to Figure Two, the approximation demonstrates that with the dilution of the solvent, parameter “ α ” decreased from 0.35 for a 1 N (that is, one **Normal**) solution to 0.08 for a concentration of 2^{-14} N. At the diluted concentration of 2^{-14} N, no difference between the

solution and the distilled water sample was found. For the distilled water sample, the trend of the dynamical curve was slightly increasing, which was opposed to what was observed for the metal cylinder and the strong electrolytes. This may be attributed to the neutralization of the positive ion charge by the negative ions from the water vapors.

The derivative of the power function (1) was negative for the solvents and positive for the distilled

water. This difference might be traced even at a concentration of 2^{-14} N, where no difference in the area was found (see Figure Two). For the other GDV parameter of averaged intensity, the direction of the trends was the opposite. It was positive for the solvents and negative for the distilled water.

It was shown earlier³ that the parameters of the GDV-grams for different electrolytes did not differ for all concentrations. In particular, at a concentration of 1 N, the parameters of the gas discharge images from the drops of NaCl and KCl solutions had no statistically significant differences.

The analysis of the deterministic trends of the corresponding time series demonstrated a statistically significant difference from 0.3 seconds to six seconds of measuring the time dynamics (see Figure Three). Parameter “ α ” was found to be 0.04 for the 1 N NaCl solution and 0.01 for the 1 N KCl solution. In addition, no significant difference for the averaged intensity parameter of these two solvents was found.

Another situation was found for KCl and KNO₃ solutions, which did not have any difference between the averaged parameters of the sampling of the GDV images³. For these two solutions, no difference for the area time series was found, but for the averaged intensity time series, the difference was statistically significant (see Figure Four). Parameter “ α ” was found to be the same for both solutions, but the absolute values of the parameter “B” were different.

Therefore, the Dynamical GDV analysis was found to be very useful for the evaluation of the difference between combinations of highly diluted aqueous solutions of strong electrolytes. This even includes solutions that are diluted to a concentration, such as 2^{-14} N, that are very close to that of distilled water.

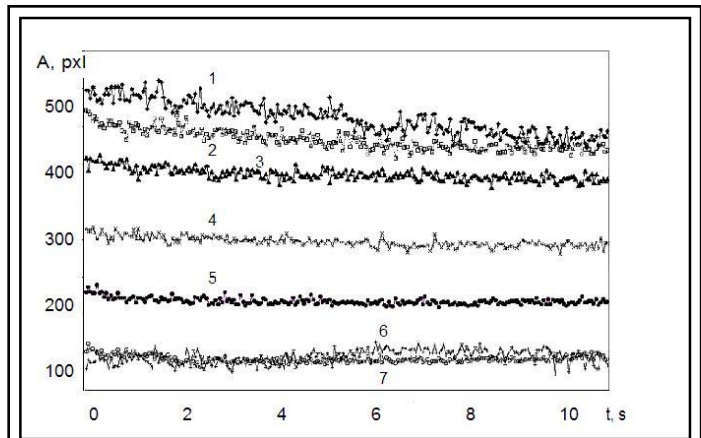


Figure Two: Time dependence of the area of the gas discharge image around drops of an NaCl solution of different concentrations (1-6) and distilled water (7). Data averaged on 10 measurements. Concentrations—**1:** 1 N; **2:** 1/6 N; **3:** 1/64 N; **4:** 1/1024 N; **5:** 1/4096 N; **6:** 1/32768 N.

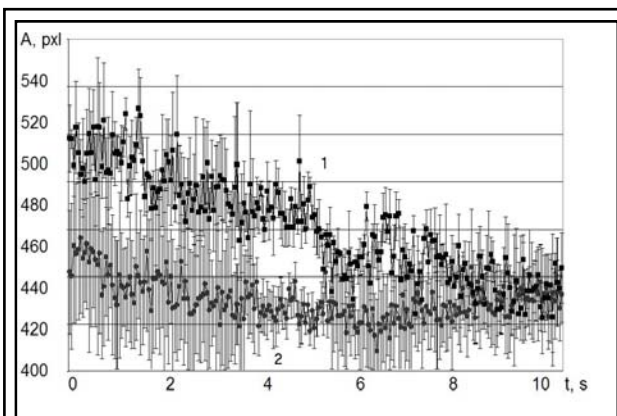


Figure Three: Time dependence of the area of the gas discharge image around drops of NaCl (1) and KCl (2) solutions both of 1 N concentration. Data averaged on 10 measurements.

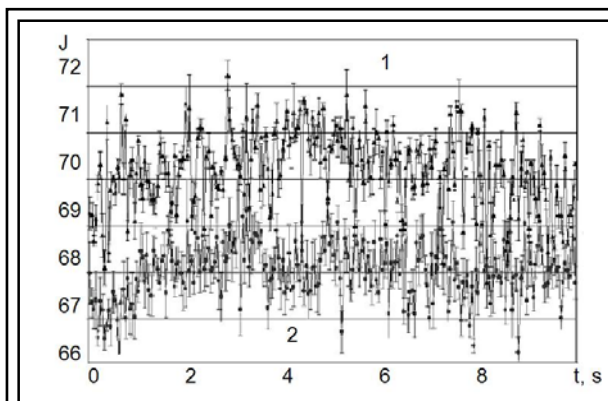


Figure Four: Time dependence of the averaged intensity of the gas discharge image around drops of KNO₃ (1) and KCl (2) solutions both of 1 N concentration. Data averaged on 10 measurements.

The Analysis of the Data on the Investigation of Essential Oils and Their Synthetic Counterparts (Liquids)—the Weakly Conducting Liquids

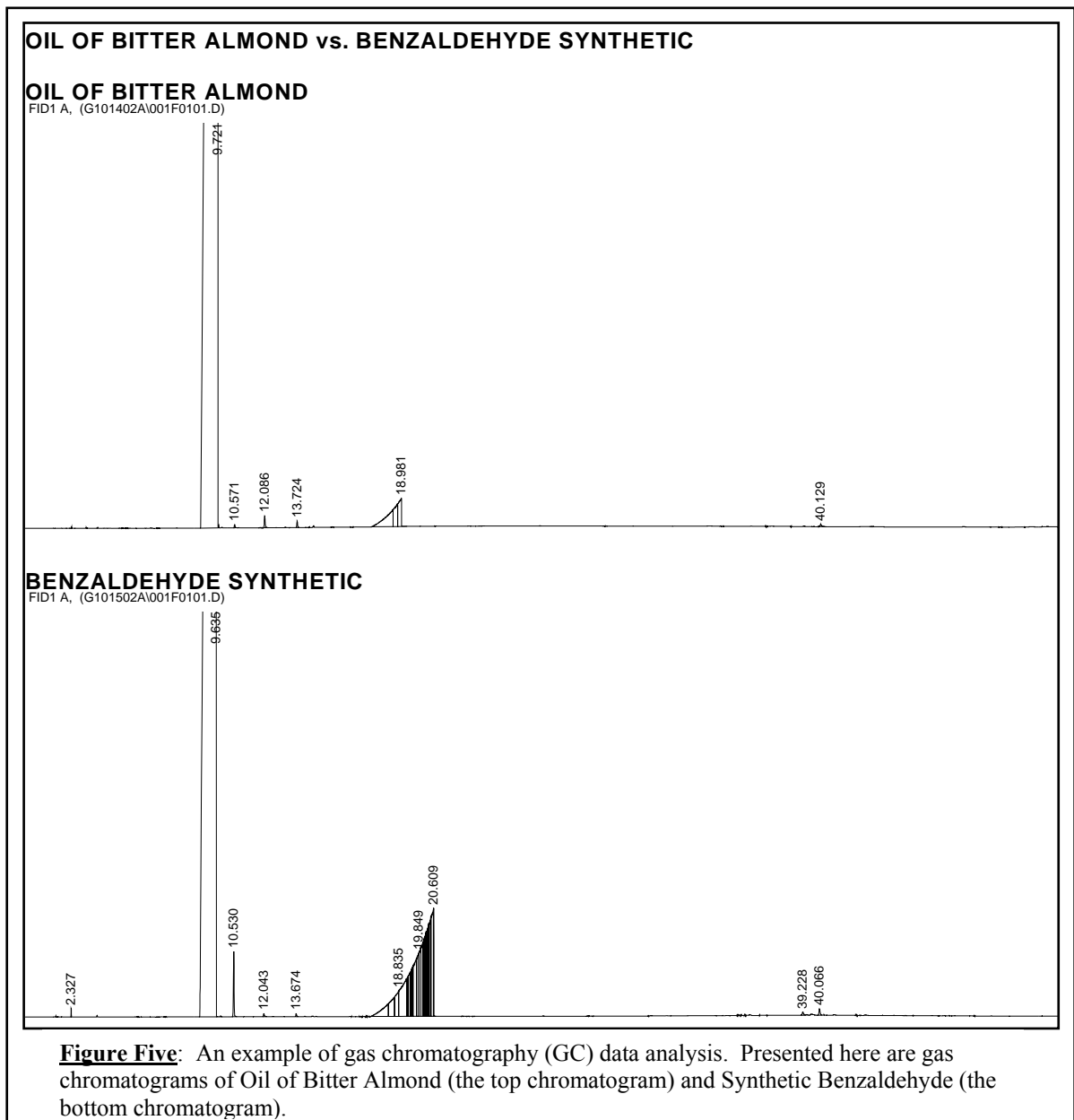
As was previously addressed, the Dynamical GDV process can be utilized for detecting or registering subtle differences amongst extremely diluted electrolyte solutions. Therefore, it was assumed that this process can be applied to the investigation of weakly conducting liquids. Such liquids included the essential oils and their synthetic counterparts (liquids), of which the same chemical compound or ingredient was naturally and synthetically derived, respectfully. This case-scenario will be discussed shortly.

Out of 84 of the studied samples of essential oils and liquids (again, keeping in mind that 42 pairs were investigated), three samples each had a dynamical curve that displayed a trend of descending character. However, this is in contrast to what was observed with conductive liquids, such that the line was quasi-linear with a small declination angle ($\alpha \sim 10^{-3}$). For the rest of the tested oils and liquids, slow inclination curves of the GDV time series with several types of behavior were observed as well.

The difference between the area and the averaged intensity for the time curves was found for the 42 pairs of investigated oils and liquids, after about two seconds of exposure to the EMF. Again, for each pair of oils and liquids, they had similar chemical contents in accordance with the gas chromatography (GC) analysis, but each differed in origin. That is, for a particular pair of oils and liquids, both could have been sourced differently, or one liquid was synthetically derived while the other was naturally derived, and so forth. In all cases, however, no distinct difference was observed between the chemical spectra of the studied pairs of oils and liquids, in accordance with the gas chromatography data.

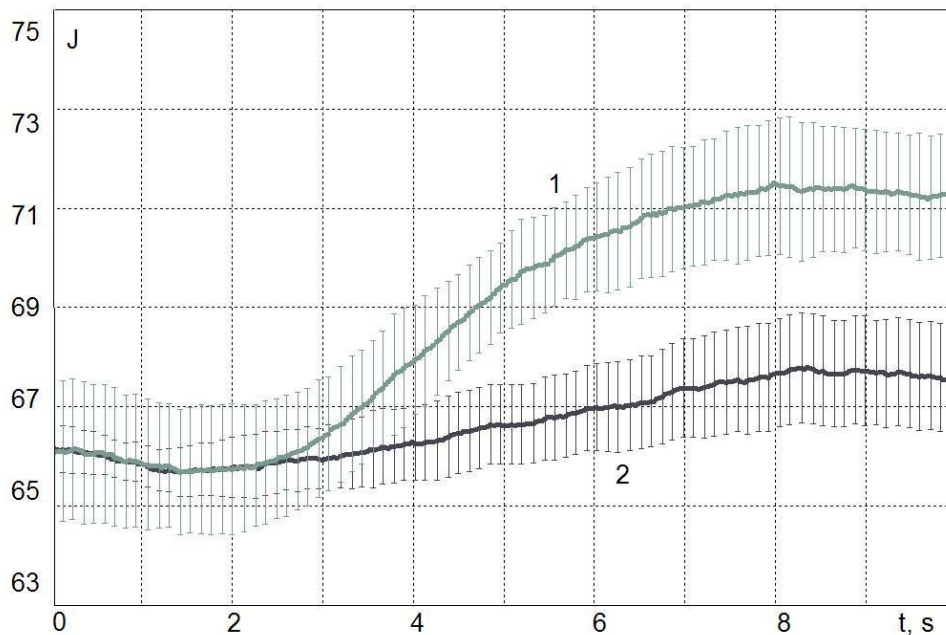
As was already mentioned, the focus of the present research was on weakly conducting liquids that were represented by essential oils and their synthetic counterparts (liquids), of which the same chemical compound or ingredient was naturally and synthetically derived, respectfully. An example of this 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 Five). Specifically, Oil of Bitter Almond contains about 95.00% pure natural benzaldehyde⁸, while Synthetic Benzaldehyde (which is commercially available as is as a result of organic chemical syntheses techniques) is 99.00+% to 99.5+% pure⁹.



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. Therefore, the 10 realizations for both Oil of Bitter Almond and Synthetic Benzaldehyde were received.



1. Fig. 6. Time dependence of the intensity of the gas discharge image around drops of Bitter Almond (1) and Synthetic Benzaldehyde (2) oils. Data averaged on 10 measurements.

For the present research, data from the first session (as an example) will be presented. Figure Six 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.

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.

IV. DISCUSSION OF RESULTS

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.

The influence of the discharge on electrolyte solutions was accompanied by the dissociation of the molecules in the solvent. The chemical interactions of the formed active particles therefore caused changes in the solution's characteristics as a whole.

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 discharge⁴. 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.

As a result of the bombardment of the solution by the energetic ions, which are emitted from the plasma, ionization and dissociation of the water molecules takes place. In this set of processes, the ions H^+ , OH^- , and solvated electrons are produced. This allows for the generation and accumulation of hydrogen peroxide (H_2O_2) in the solution. In addition, it is important that NO_2 gas molecules could be formed.

Therefore, it can be assumed that the reproducible differences of the GDV parameters of the time series of area and averaged intensity for both various concentrations of electrolytes and oils and liquids of different origin became apparent. This was done at the expense of the specific self-organization of the investigated liquids into periodically repeating structures.

V. CONCLUSION

1. The investigation of both conductive and weakly conductive liquids 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. Dynamical studies enable one to find differences in the characteristics of liquids (several concentrations of strong electrolytes and different essential oils and certain liquids), which did not differ in the parameters of single GDV-grams.

2. For all concentrations of strong electrolytes (NaCl, KCl, NaNO₃, and KNO₃), the trend was monotonously decreasing, while for distilled water the trend was slightly increasing.

3. A concentration of 2^{-14} N for the strong electrolytes was found such that this concentration is where the difference between absolute parameters of distilled water and the particular electrolyte disappeared, while the difference in the character of the dynamical trends was still apparent.

4. 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 investigation of Oil of Bitter Almond versus Synthetic Benzaldehyde, in which the same chemical ingredient (that is, Benzaldehyde) 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.

5. From the 42 pairs of studied oils, 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.

6. Parameter “ α ” from the approximation power function ($y = Ax^\alpha + B$) had both positive and negative values. This may be correlated with the increase and decrease of the electron-free

movement path in the near-surface space. It is in this space that every subsequent discharge process mostly depends on the accumulation of positive and negative ions on the dielectric surface.

7. Parameter “ α ” was also used for analyzing the time series realizations of area and averaged intensity of the discharge process for the different liquids. Parameter “ α ”, in this case, was shown to have had a reverse polarity that was probably correlated with the intensity of the electron and photon emissions in the discharge process. This topic needs more intensive study.

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⁸ Ziegler, E. “Botanical Species: Prunus dulcis (Miller) D. A. Webb (P. amygdalus Batsch, P. communis L.)” *Die Natuerlichen Und Kuenstlichen Aromen*. (Dr. Alfred Huethig Verlag, Heidelberg, 1982, 61)—This information was obtained from a search on Oil of Bitter Almond from the ESO 00 Database Report (© 1999, BACIS, The Netherlands).

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