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Time Dynamics of the Gas Discharge Around Drops of Liquids

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I. INTRODUCTION

Oils used in personal care products can be made by either one of two conventional ways. They can be synthesized by using precise organic chemical synthesis techniques. On the other hand, the oils can be naturally derived, predominantly from a plant source. In either case, innovative technologies are being applied in order to get the purest ingredient that is necessary

for obtaining the desired oil. These techniques are using conventional chemical and physical analytical tools that by themselves cannot distinguish between the same oils of natural and synthetic origin.

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 cosmeticsⁱ. 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^{ii, iii}. 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.

Results of^{iv} demonstrated that in controlled blinded randomized assessment of four split samples of 30c potencies of three homeopathic remedies from different kingdoms GDV technology allowed differentiation of the remedies from the solvent controls.

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 surface^v. 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.

Dynamic GDV-graphy uses video-files (GDV-video), instead of separate snapshots, for capturing the development of the image of gas discharge during the time of the exposure to the EMF. GDV-video is comprised of the following:

- The GDV Camera instrument that has a digital USB-interface (or an older model of an instrument with a connected "GDV + " device).
- The GDV Capture program, which is needed for the registration and division of video-files into separate image files.
- The GDV Video Analyzer or GDV Processor program for the calculation of quantitative parameters of the images.
- Programs for the mathematical estimation of the time series of the parameters of the GDV-gram (for example, Excel, Statistica, «Caterpillar»-SSA, etc.).
- Pentium II and higher class PCs can be used for the registration and work with GDV-video.

In the present research, all of the tested oils were subjected to the influence of a strong electromagnetic field from about two to 10 seconds on the first voltage mode of the camera. Recording was carried out in the form of a video-file with the frequency of 30 shots per second. The received video-files were converted into a series of GDV images. The extracted images had a size of 160x120 and 320x240 pixels. Therefore, a time series was presented with equal intervals of 0.034 seconds, where GDV-gram Area and Average Intensity were both used as parameters. The choice of these parameters was determined by their maximum reproducibility in Dynamic GDV-graphy. For all measurements, statistical analysis of these two parameters utilized Fisher's test at 95% Confidence Intervals.

A few techniques of the investigation of oils were studied:

- Placement of a drop of an oil on the glass with an opening covered by a high-density paper filter from above.
- Placement of a drop of an oil on the glass with an opening, but without filter.
- Touching a drop of an oil with a grounded needle.
- Suspension of a drop of an oil in a syringe.

It is worth mentioning that the most reproducible technique was the second one listed, which was the placement of a drop of an oil into the hole of the glass with a grounded needle.

In this case, a sample of an oil was placed into a cylindrical hole with dimensions of 2 mm in depth and 6 mm in diameter on a glass plate. A minimum volume for the sample was selected in such a way that in the end of the time series, the oil covered the bottom of the hole evenly. A volume of about four to 20 microliters was determined as optimal, and was applied by a standard micropipet. The hole was centered with the help of the GDV Capture program. The filter was grounded by a standard set of needles from the GDV Material Testing Kit.

Advantages of placing a drop of an oil on the glass and grounding with a metal rod include good reproducibility and the opportunity to observe structural changes in the volume of the liquid. However, the disadvantages of the previously mentioned technique along with the others include the following:

- For certain oils, they might spread heavily along the containing perimeter.
- Additional fluctuations might be seen because of the possible quick evaporation of a specific oil.

A new device that connects the GDV Camera with a computer was developed. It provides for the simultaneous application of the gas discharge and for the start of shooting. Each shooting was carried out on a new sample of oil. Isopropyl alcohol was used to clean the glasses between measurements.

It is also worth mentioning that some ethyl oils solidify even with a small decrease in temperature. Furthermore, the formation of a gas discharge is sensitive to a strong drop in both temperature and relative humidity. Therefore, a thorough control of the mentioned physical characteristics is required. All research was carried out in the temperature range of 21°C to 23°C and a relative humidity range from 42% to 44%.

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 criteria^{vi} 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^{vii, viii}

, 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 Fig.1.

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.

3.1. 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.

3.2. 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 Fig. 5). 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.

For the present research, data from the first session (as an example) will be presented. Fig. 6 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.

For the oils of Moroccan Rose, Bulgarian Rose, and Russian Rose, they are chemically similar; however, as can be seen, they are sourced from different locations. During the initial moments of measurement, the time series for area for each oil coincided with each other. However, in less than one second, each of the curves began to deviate and differ statistically significantly from one another (see fig. 7). Parameter " α " in the approximation for the different Rose oils was $2 \cdot 10^{-2}$, $7 \cdot 10^{-3}$ and $-2 \cdot 10^{-2}$, respectively. Positive and negative values of " α " for Moroccan Rose oil and Russian Rose oil, respectively, with the same absolute values of area illustrated the sensitivity of this parameter to the physical and chemical properties of the liquids. In addition, no statistically significant difference between Moroccan Rose and Bulgarian Rose oils was found, but they both differed statistically significantly from Russian Rose oil.

In more precise studies, highly purified Citral Natural and highly purified Citral Synthetic oils were used. Both oils abruptly changed their behavior in interaction with the electromagnetic field after about 1.7 seconds (see fig.8). Parameter " α " for the area time series changed its value from $-6 \cdot 10^{-2}$ to $5 \cdot 10^{-3}$. This could be correlated with the process of surface charging in accordance with the previously mentioned discussion. After 1.7 seconds of exposure to the EMF, a statistically significant reproducible difference was detected between the synthetic and natural Citral essential oils. The same type of behavior was also found for the intensity time series. However, the intensity of the gas discharge glow for the Citral synthetic oil was higher than that for the Citral natural oil.

3.4. Data Analysis.

In the present work, the following samples of oils were studied:

1. Oils of Essential and Synthetic Nature

- 1.1. Bitter Almond versus Synthetic Benzaldehyde
- 1.2. Linalool Synthetic versus Linalool Extracted from Bergamot
- 1.3. Linalool Synthetic versus Linalool Extracted from Bois de Rose
- 1.4. Linalool Synthetic versus Linalool Extracted from Pinene
- 1.5. Linalool Synthetic versus Linalool Extracted from Shiu Oil

2. Oils of Organic and Regular Origin

- 2.1. Peppermint Organic versus Peppermint Regular
- 2.2. Lavender Organic versus Lavender Regular

- 2.3. Cloveland Organic versus Cloveland Regular*
3. Oils Received in Different Climatic Conditions and Extracted by Different Ways (Oils from Different Sources)*
 - 3.1. Jasmine Oils: Algerian, Indian, and Moroccan
 - 3.2. Orange Oils: Cold-Pressed Valencia FCC⁺, Midseason FCC⁺, and Brazil Cold-Pressed High Aldehyde Content
 - 3.3. Rose Oils: Bulgarian, Bulgarian (Otto), Bulgarian Alba Organic, Moroccan (Otto), Russian, and Turkish Bulgarian Type
4. Oils of Various Optical Activity
 - 4.1. Dextro Carvone versus Laevo Carvone
 - 4.2. Dextro Limonene versus Laevo Limonene
 - 4.3. Dextro Linalool versus Laevo Linalool
5. Natural Oils: Fresh and Oxidized in Various Ways*
 - 5.1. Lime Oil Italian Refrigerated versus Lime Oil Italian Oven Treated at 120°F
 - 5.2. Lime Oil Italian Refrigerated versus Lime Oil Italian Xenon Arc Treated
 - 5.3. Lime Oil Mexican Refrigerated versus Lime Oil Mexican Oven Treated at 120°F
 - 5.4. Lime Oil Mexican Refrigerated versus Lime Oil Mexican Xenon Arc Treated

*) Denotes research that was done in Russia.

⁺) FCC is an acronym for Food Chemical Codex.

1. Essential Oils versus Synthetic Oils.

1.1. 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 Appendix for these and other GC data). However, GC analysis by itself has its limitations; that is, it cannot reveal the subtle differences between the two materials. This is why analysis by Dynamic GDV-graphy has been applied to this situation.

The comparison between Bitter Almond and 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 Bitter Almond and Benzaldehyde were received.

For the first session, the graph for the averaged realizations of Area using 95% Confidence Intervals demonstrates that the average values of realizations of Bitter Almond and Benzaldehyde oils have no statistically significant difference. However, the same realizations for the Average Intensity parameter (see fig.6) show a statistically significant difference, again using 95% Confidence Intervals, after four seconds of measurements.

For the second session, the averaged realizations of Area (fig.9), using 95% trusting intervals, show a statistically significant difference that appears after six seconds from the initial moment of registration of the Dynamic GDV processes. The same realizations for the Average Intensity (fig.10) demonstrate that a statistically significant difference, using 95% trusting intervals, does exist during the entire time of registration.

However, for the third session, a statistically significant difference for all parameters was not observed.

The main trends of these experiments for these two oils reveal a higher position of realizations of synthetic oils by Area and a lower position by Average Intensity. Therefore, the desired parameters for revealing significant differences between Oil of Bitter Almond and Synthetic Benzaldehyde in terms of both Area and Average Intensity are characteristic of those demonstrated in the second session of the experiments. Also, it is proven 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.

1.2. Linalool Natural versus Linalool Synthetic

For this set of experiments, the oils of concern were non-conductive liquids, and because of that, a certain high voltage was not enough for taking gas discharge images. Therefore, gas discharges were observed only during the second and third sessions. All Linalool oils (both of synthetic and of natural origins) were identified through GC analysis (see Appendix for these and other GC data).

For the second session, 10 realizations for every type of oil were received. Statistically significant differences, using 95% Confidence Intervals, were obtained by the Average Intensity parameter for Linalool Synthetic and all Linalool Natural oils, except Linalool Extracted from Shiu oil (fig.11). For the realization of these oils by Area, significant differences were not revealed.

For the third session, 10 realizations for Linalool Extracted from Bergamot and Linalool Synthetic were received. A statistically significant difference, again using 95% Confidence Intervals, was obtained by Area during the first second (fig. 12). There was no significant difference by Average Intensity during the entire time of registration of the Dynamic GDV processes.

Therefore, the desired parameters for revealing significant differences between natural and synthetic Linalool oils in terms of Area are characteristic of those demonstrated only in the third session of the experiments. However, the second session of the experiments has the appropriate parameters for revealing significant differences between natural and synthetic Linalool oils in terms of Average Intensity. Also, it is proven that the same chemical ingredient in both materials (which is Linalool for this situation) can be distinguished from one another by using Dynamic GDV-graphy.

2. Oils of Organic and Regular Origin.

2.1. Peppermint Organic versus Peppermint Regular

Nine realizations for Peppermint Organic oil and ten for Peppermint Regular oil were received. In this section, please make note of the terms organic and regular. The term “organic”, in this sense, refers to an agricultural process of growing natural plant raw materials without the use of pesticides, insecticides, plant fertilizers, and the like. On the other hand, “regular” refers to conventional agricultural practices, which are usually not restricted from the use of these chemicals.

The graph for the averaged realizations of Area used 95% Confidence Intervals for every moment of registration of the Dynamic GDV processes for the two oils (fig. 13) demonstrates that the average values of the realizations of Peppermint Organic and Peppermint Regular oils for Area show a statistically significant difference from one to three seconds and from seven to 10 seconds. Again, a statistically significant difference was not observed for the Average Intensity parameter for these realizations.

The trends of the compared oils are different. Peppermint Regular oil has a trend that shows linearly increasing behavior. Peppermint Organic oil has a trend that is close to a polynomial form of the 3rd order. Apparently, the ionization process considerably exceeds the evaporation process of Peppermint Regular oil. However, at the same time, the evaporation process for Peppermint Organic oil sometimes competes with the ionization process. Still, the latter dominates for the larger part of the time of registration.

2.2. Lavender Organic versus Lavender Regular

Eight realizations for Lavender Organic oil and five for Lavender Regular oil were received. The graphs for the averaged realizations of Area and Average Intensity used 95%

Confidence Intervals for every moment of registration of the Dynamic GDV processes for the two oils (fig. 14) demonstrates that the average values of the realizations of Lavender Organic oil and Lavender Regular oil for Area show a statistically significant difference during the first six seconds of the registration of the Dynamic GDV processes. The trends of the compared oils do not differ; they are approximately linear in character. However, a statistically significant difference was not observed for the Average Intensity parameter for these realizations.

2.3. Cloveland Organic versus Cloveland Regular

For Cloveland Organic oil and Cloveland Regular oil, four and six realizations were received, respectively. The graph (fig. 15) for the averaged realizations of Area used 95% Confidence Intervals for each moment of registration of the Dynamic GDV processes for the compared oils. It follows from this graph that only the average values of the realizations of Cloveland Organic oil and Cloveland Regular oil for the first 0.2 seconds of observation demonstrate a statistical difference, again in terms of Area only. However, a statistically significant difference was not observed for the Average Intensity parameter for these realizations.

Still, the trends of the compared oils do differ. Cloveland Organic oil has a trend that shows linearly increasing behavior. However, Cloveland Regular oil has a trend that is close to a polynomial form of the 3rd order. Apparently, the ionization process considerably exceeds the evaporation process of Cloveland Organic oil. At the same time, the Cloveland Regular oil evaporation process sometimes competes with the ionization process. However, as was already observed in earlier experiments, the latter dominates for the larger part of the time of registration.

3. Oils Received in Different Climatic Conditions and Extracted by Different Ways (Oils from Different Sources).

3.1. Jasmine Oils: Algerian, Indian, and Moroccan

For Algerian, Indian, and Moroccan Jasmine oils, five, six, and six realizations were obtained, respectively. The graph (fig. 16) for the averaged realizations of Area used 95% Confidence Intervals for every moment of registration of the Dynamic GDV processes for the three given oils demonstrates that the average value of the realizations of Algerian Jasmine oil shows a statistically significant difference from both Indian Jasmine and Moroccan Jasmine oils, in random moments of time. Still in terms of the Area parameter, the two latter oils have no statistically significant difference amongst themselves; however, as already mentioned, they show a statistically significant difference from Algerian Jasmine oil. Because statistically significant differences were not observed for the Average Intensity parameter for these oil realizations, this graphical data is not presented in the Appendix.

The trends of these oils are similar in character and can be approximated by polynomials of the 3rd order. However, both Indian and Moroccan Jasmine oils reach a “stationary mode” at about 1.7 seconds, while Algerian Jasmine oil reaches this point at the middle of the registration time (that is, at about one second). At about 1.4 seconds, the Algerian Jasmine oil curve starts to decrease quasilinearly, which is evidence for the change from the dominating mode of ionization to evaporation.

3.2. Orange Oils: Cold-Pressed Valencia FCC[‡], Midseason FCC[‡], and Brazil Cold-Pressed High Aldehyde Content ([‡]FCC is the acronym for Food Chemical Codex.)

Seven realizations for each of the following oils were received at 95% Confidence Intervals: Cold-Pressed Valencia FCC, Midseason FCC, and Brazil Cold-Pressed High Aldehyde Content. In this section, please make note of the acronym FCC, which stands for Food Chemical Codex. FCC refers to a compilation of specific tests that have been done on certain

foods and/ or food chemicals.

From the graph (fig. 17), one can conclude that the average values of the realizations in terms of the Area parameter have statistically significant differences in almost all moments of observation. However, the exception to this observation is the pair Midseason FCC Orange Oil and Brazil Cold-Pressed High Aldehyde Content Orange Oil, where significant differences between the two start to develop on the 13th frame (that is, at about 0.43 seconds). Again, because statistically significant differences were not observed for the Average Intensity parameter for these oil realizations, this graphical data is not presented in the Appendix.

The form of the trends of each of the studied orange oils is the same in character. That is, they each exhibit the behavior of a quasilinearly increasing function. This indicates that the process of ionization plays a dominating role in the Dynamic GDV processes of these oils for the entire time of observation.

3.3. Rose Oils: Bulgarian, Bulgarian (Otto), Bulgarian Alba Organic, Moroccan (Otto), Russian, and Turkish Bulgarian Type

Six realizations were received for each of the following oils: Bulgarian, Bulgarian (Otto), and Bulgarian Alba Organic. For Moroccan (Otto), Russian, and Turkish Bulgarian Type oils, six, seven, and six realizations were received, respectively.

Figure 18 demonstrates that the average value of the Area realizations of Russian Rose oil has a statistically significant difference from all of the other compared oils. Bulgarian Rose, Moroccan (Otto) Rose, and Turkish Bulgarian Type Rose oils each start demonstrating statistically significant differences from Bulgarian (Otto) Rose oil after about 0.67 seconds of the Dynamic GDV processes' registration, and are indistinguishable from Bulgarian Alba Organic Rose oil. The first three oils (Bulgarian Rose, Moroccan [Otto] Rose, and Turkish Bulgarian Type Rose oils), as well as the latter two (Bulgarian [Otto] Rose oil and Bulgarian Alba Organic Rose oil), each have no statistically significant differences between themselves. However, as already mentioned, they show a statistically significant difference from Russian Rose oil. Because statistically significant differences were not observed for the Average Intensity parameter for these oil realizations, this graphical data is not presented in the Appendix.

All trends, with the exception of those of Russian Rose and Bulgarian Alba Organic Rose oils, have a similar character, and may be approximated by polynomials of the 3rd order. These mentioned trends each reach a "stationary mode" at the level of about 1.8 seconds. For Bulgarian Alba Organic Rose oil, this mode starts at the middle of the registration time, which is at about one second. At the level of about 1.4 seconds, the curve starts to decrease quasilinearly (as in the third paragraph of section 5.3.1), which indicates the change of the dominating mode from ionization to evaporation.

Finally, the trend of Russian Rose oil has the following behavioral form: $y = \{(\text{initial value}) - x^2\}$ to 1.7 seconds, where the curve receives its stationary form. It is obvious that the process of evaporation dominates in this case.

4. Oils with Different Optical Activity.

4.1. Dextro Carvone versus Laevo Carvone

Both Dextro Carvone and Laevo Carvone oils were identified through GC analysis (see Appendix for these and other GC data). For both Dextro Carvone and Laevo Carvone oils, ten realizations were obtained. Average values of the realizations for both Area and Average Intensity (taken at 95% Confidence Intervals) do not have statistically significant differences during all moments of observation.

The trends of both oils have the form of a decreasing power function. It is obvious that the process of evaporation dominates in this case.

4.2. Dextro Limonene versus Laevo Limonene

Both Dextro Limonene and Laevo Limonene oils were identified through GC analysis (see Appendix for these and other GC data). Eight and six realizations were received for Dextro Limonene and Laevo Limonene, respectively. The graph (fig.19) shows that the average values of the realizations of Dextro Limonene oil and Laevo Limonene oil have a statistically significant difference, again in terms of Area only. However, average values of these oil realizations for the Average Intensity parameter do not have a statistically significant difference during all moments of observation.

The trends of both oils have the form of a decreasing power function as well. Again, it is obvious that the process of evaporation dominates in this case.

4.3. Dextro Linalool versus Laevo Linalool

Both Dextro Linalool and Laevo Linalool oils were identified through GC analysis (see Appendix for these and other GC data). Five and four realizations were received for Dextro Linalool and Laevo Linalool, respectively. From the graphs (fig.20 and 21), one can conclude that the average values of the oil realizations for both Area and Average Intensity (taken at 95% Confidence Intervals) have statistically significant differences after about 2.5 seconds from the beginning of the observation.

The Area trends of the compared oils do differ however. Dextro Linalool oil has a trend that practically exhibits a linearly increasing character. On the other hand, Laevo Linalool oil has a trend that is close to that of an increasing power function. Apparently, the ionization process for Laevo Linalool oil considerably exceeds the process of its evaporation. At the same time, the Dextro Linalool oil evaporation process sometimes competes with its ionization process; however, the latter dominates for the larger part of the time of the registration.

5. Fresh and Oxidized by Various Ways-- The Natural Oils.

Natural oils derived from citrus products (such as Lime oil, which will be discussed in this section) contain trace amounts of a specific class of chemicals known as furanocumarins. Citrus oils that contain these chemicals, when used topically, are known to cause skin irritations (such as photodermatitis). Because of this, a series of accelerated stability/ degradation tests have been performed on these oils for the purpose of determining the furanocumarin content. In performing such tests, citrus oils are typically stored under one or more of the following extreme conditions: accelerated heat/ temperature (oven treatment at 120°F simulating approximately nine months of shelf-life), under a xenon arc lamp (photochemical effects), and in an air-oxygen environment (an oxidation process).

While still fresh, these oils are usually refrigerated for normal stability. Therefore, when it comes to these tests, one is investigating an increase or decrease in the furanocumarin content of the oil from the initial reference (that is, the refrigerated oil—the fresh case scenario) to the final case of subjecting the oil to one of the three extreme conditions. The final case, in addition to measuring the changes in furanocumarin content, is also measuring the potential degradation of these oils, resulting from extreme storage conditions. Because furanocumarins, such as bergaptenes (a subclass of furanocumarins), can fluoresce, this increase or decrease can be easily detected and monitored by some means of chemical analysis (such as UV/Vis spectrophotometry).

5.1. Lime Oil Italian Refrigerated versus Lime Oil Italian Oven Treated at 120°F

Both Lime Oil Italian Refrigerated and Lime Oil Italian Oven Treated at 120°F were both identified through GC analysis (see Appendix for these and other GC data). For each of these oils, six realizations were obtained. The given pair of oils (Lime Oil Italian Refrigerated and Lime Oil Italian Oven Treated at 120°F) does not have a statistically significant difference for

the averaged realizations in terms of neither Area nor Intensity.

The character of the trends of both oils, as in some of the previous cases, has the same form of a polynomial of the 3rd order, with a “stationary mode” at the level of about 1.8 seconds. Therefore, the already mentioned assumptions on the form of the trends of the compared oils, as in the previous examples, are valid for this pair of oils.

5.2. Lime Oil Italian Refrigerated versus Lime Oil Italian Xenon Arc Treated

Both Lime Oil Italian Refrigerated and Lime Oil Italian Xenon Arc Treated were identified through GC analysis. Results of the analysis of Lime Oil Italian Refrigerated are already presented in section 5.5.1. For Lime Oil Italian Xenon Arc Treated, six realizations were obtained. The given pair of oils, as well as in the previous case, does not have a statistically significant difference for the averaged realizations in terms of Area or Intensity. The trends of these compared oils have the same form (as was already mentioned in the previous case), and are indistinguishable from those of the previous case.

5.3. Lime Oil Mexican Refrigerated versus Lime Oil Mexican Oven Treated at 120°F

Both Lime Oil Mexican Refrigerated and Lime Oil Mexican Oven Treated at 120°F were also identified through GC analysis. For Lime Oil Mexican Refrigerated, seven realizations were received. Six realizations were received for Lime Oil Mexican Oven Treated at 120°F. The graph (fig.22) demonstrates that the given pair of oils has a statistically significant difference (in terms of Area) within the entire time of the registration, owing to the absence of the intersections of the 95% Confidence Intervals. No statistically significant difference was observed for the Average Intensity parameter for these oil realizations.

The trends of the compared oils are different. The Lime Oil Mexican Refrigerated trend exhibits a quasilinear increasing form up to about 1.9 seconds. From about 1.9 to 2 seconds, the curve is stationary. However, the trend of Lime Oil Mexican Oven Treated at 120°F is close to a polynomial form of the 3rd order. Apparently, the ionization process significantly exceeds the process of evaporation for Lime Oil Mexican Refrigerated. From the previous discussions, one can conclude that for Lime Oil Mexican Oven Treated at 120°F, the process of evaporation sometimes “competes” with the process of ionization; however, the latter significantly dominates longer.

5.4. Lime Oil Mexican Refrigerated versus Lime Oil Mexican Xenon Arc Treated

Both Lime Oil Mexican Refrigerated and Lime Oil Mexican Xenon Arc Treated were both identified through GC analysis. Four realizations were received for Lime Oil Mexican Xenon Arc Treated. No statistically significant difference for the Area and Intensity was found.

In general, the trends of the given pair of oils do not show a strong difference. It is worth stressing that for Lime Oil Mexican Xenon Arc Treated, the trend deviates from quasilinear behavior to strong nonlinear behavior. However, for the already mentioned pair of oils, the differences are insignificant.

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 , $NaNO_3$, and KNO_3), 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.

The development of the Dynamic GDV-graphy technique presented the opportunity to reveal statistically significant differences when comparing a wide range of oils. These differences are manifested both in the absolute volumes of the parameters of the studied oils and in the change of the form of the deterministic components in a certain moment of time. Further automation of this technique will enable one to rapidly obtain results of the analysis of the compared pairs of oils.

The technique that was utilized in these experiments showed high reproducibility. Again, the reproducible experimental technique that showed the most promising results was that of placing a drop of oil on the glass and subsequent grounding with a metal rod. At the same time, it was necessary to improve the device for the purpose of minimizing the influence of an operator during the centering and filling of the opening with the liquid (oil), as well as the influence of the uneven distribution of the tested liquid along the glass surface.

ACKNOWLEDGMENTS

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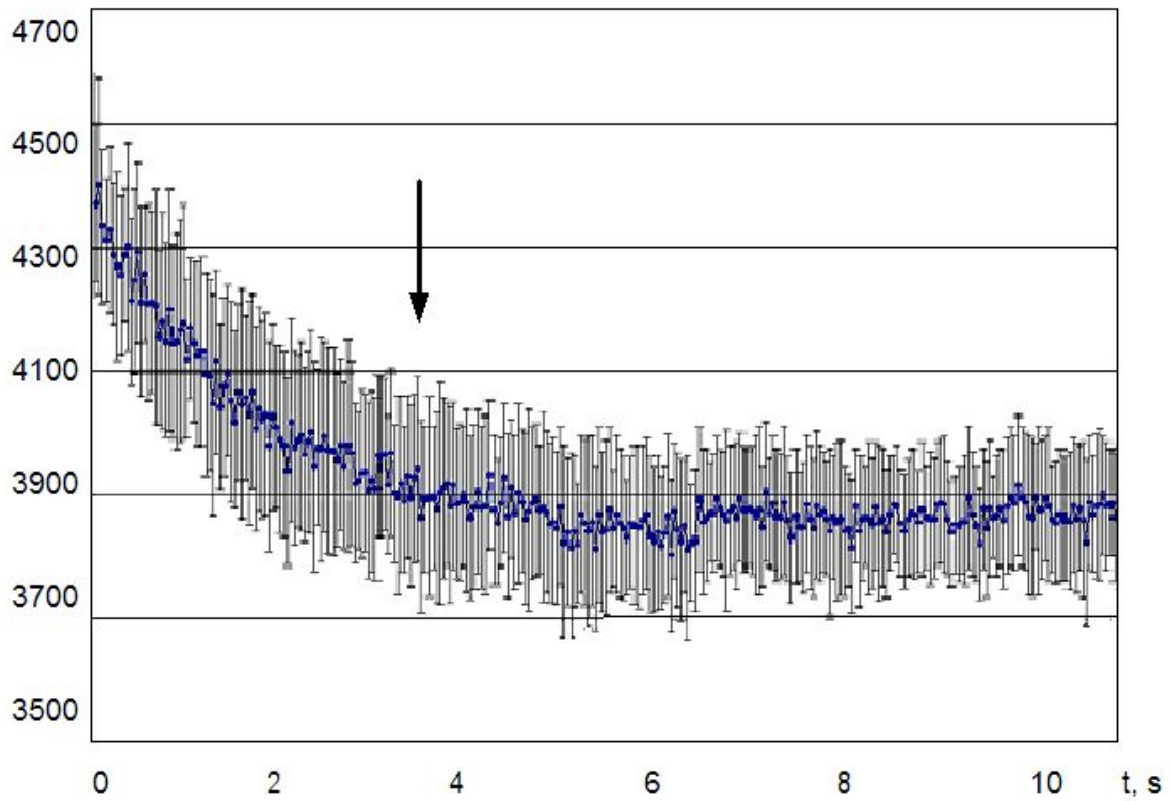
⁹ Information on Benzaldehyde (Product Numbers 41, 809-9 and B133-4) Taken from the 2003-2004 Aldrich® Handbook of Fine Chemicals and Laboratory Equipment.

Table 1. Results of study of different pairs of oils

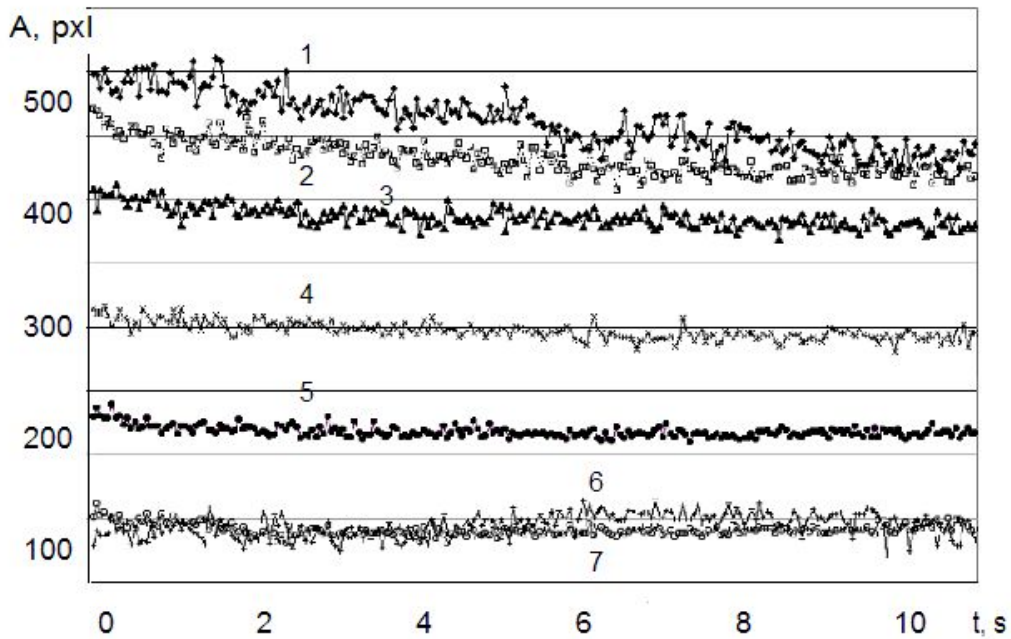
The Individual Case Scenarios with Their Corresponding Oil Comparisons	Area	Intensity
Oils of Essential and Synthetic Nature		
Bitter Almond versus Synthetic Benzaldehyde (Session 1)		X
Bitter Almond versus Synthetic Benzaldehyde (Session 2)	X	X
Bitter Almond versus Synthetic Benzaldehyde (Session 3)		
Linalool Synthetic versus Linalool Extracted from Bergamot (Session 2)		X
Linalool Synthetic versus Linalool Extracted from Bergamot (Session 3)	X	
Linalool Synthetic versus Linalool Extracted from Bois de Rose (Session 2)		X
Linalool Synthetic versus Linalool Extracted from Pinene (Session 2)		X
Linalool Synthetic versus Linalool Extracted from Shiu Oil (Session 2)	X	
Oils of Organic and Regular Origin		
Peppermint Organic versus Peppermint Regular	X	
Lavender Organic versus Lavender Regular	X	
Cloveland Organic versus Cloveland Regular	X	
Oils Received in Different Climatic Conditions and Extracted by Different Ways (Oils from Different Sources)		
Indian Jasmine versus Moroccan Jasmine		
Algerian Jasmine versus Indian Jasmine	X	
Algerian Jasmine versus Moroccan Jasmine	X	
Cold-Pressed Valencia FCC* Orange versus Midseason FCC* Orange	X	
Cold-Pressed Valencia FCC* Orange versus Brazil Cold-Pressed High Aldehyde Content Orange	X	
Brazil Cold-Pressed High Aldehyde Content Orange versus Midseason FCC* Orange	X	
Bulgarian Rose versus Russian Rose	X	
Bulgarian (Otto) Rose versus Russian Rose	X	
Bulgarian Alba Organic Rose versus Russian Rose	X	
Moroccan (Otto) Rose versus Russian Rose	X	
Turkish Bulgarian Type Rose versus Russian Rose	X	
Bulgarian Rose versus Bulgarian (Otto) Rose	X	
Bulgarian (Otto) Rose versus Moroccan (Otto) Rose	X	
Bulgarian (Otto) Rose versus Turkish Bulgarian Type Rose	X	
Bulgarian Rose versus Bulgarian Alba Organic Rose		
Bulgarian Alba Organic Rose versus Moroccan (Otto) Rose		
Bulgarian Alba Organic Rose versus Turkish Bulgarian Type Rose		
Bulgarian Rose versus Moroccan (Otto) Rose		
Bulgarian Rose versus Turkish Bulgarian Type Rose		
Moroccan (Otto) Rose versus Turkish Bulgarian Type Rose		
Bulgarian (Otto) Rose versus Bulgarian Alba Organic Rose		
Oils of Various Optical Activity		
Dextro Carvone versus Laevo Carvone	X	
Dextro Limonene versus Laevo Limonene	X	
Dextro Linalool versus Laevo Linalool	X	X

Natural Oils: Fresh and Oxidized in Various Ways		
Lime Oil Italian Refrigerated versus Lime Oil Italian Oven Treated at 120°F	X	
Lime Oil Italian Refrigerated versus Lime Oil Italian Xenon Arc Treated	X	
Lime Oil Mexican Refrigerated versus Lime Oil Mexican Oven Treated at 120°F	X	
Lime Oil Mexican Refrigerated versus Lime Oil Mexican Xenon Arc Treated	X	

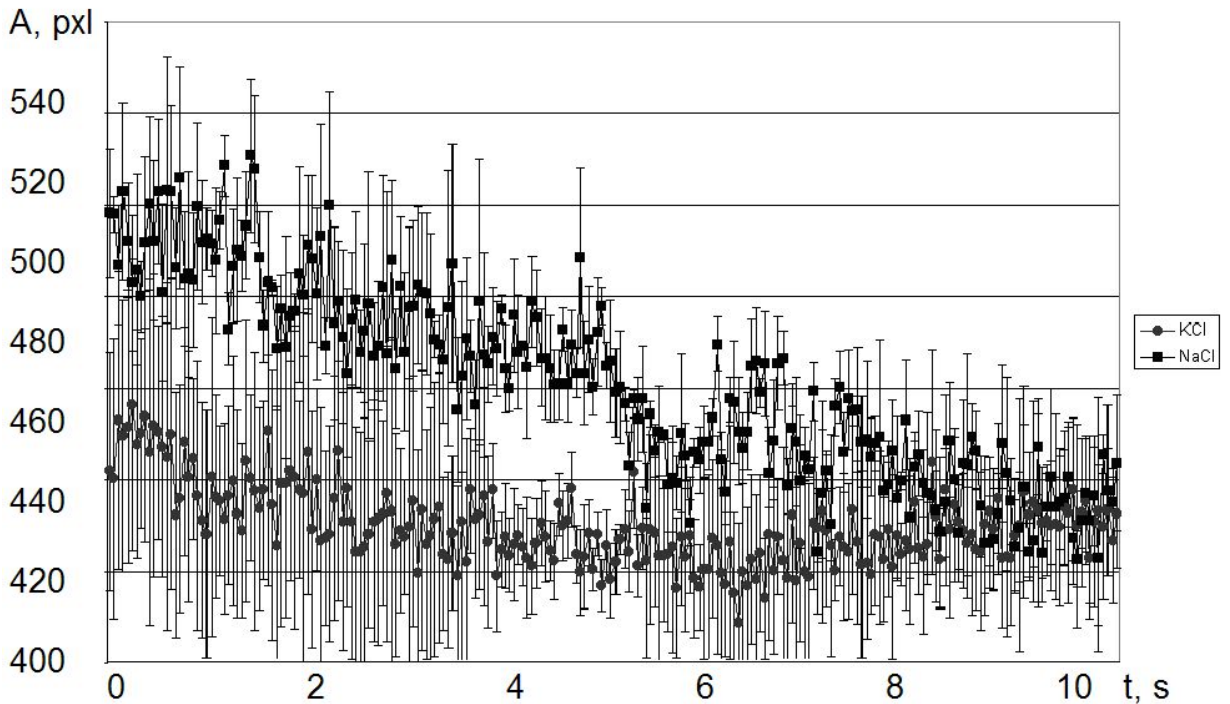
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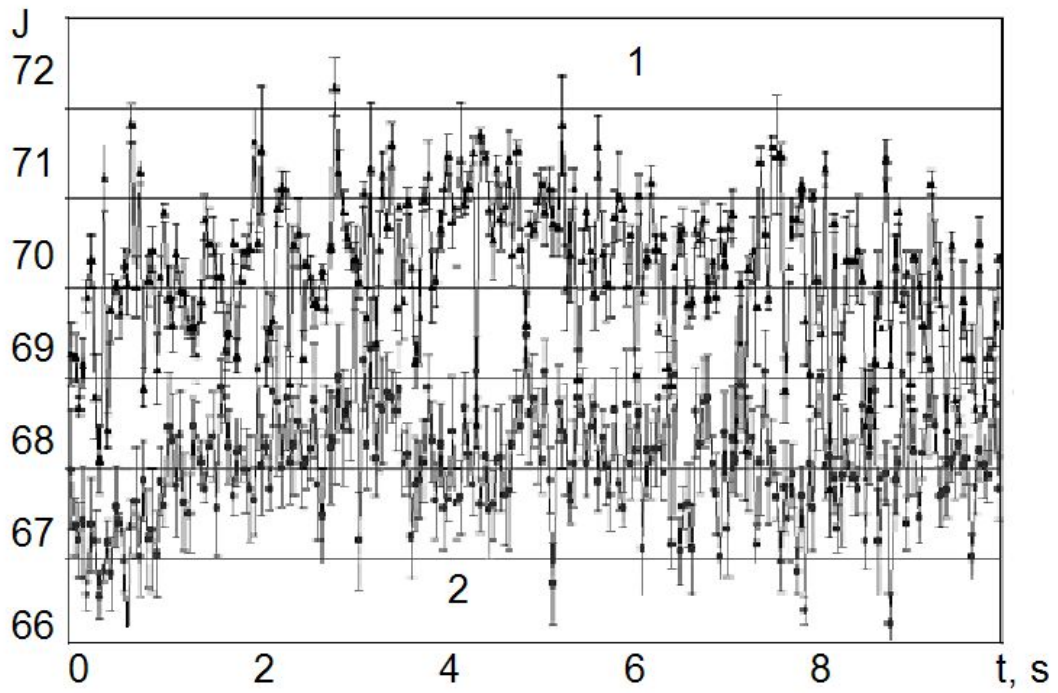
1. Time dependence of the area of the gas discharge image around a metal cylinder. Data was averaged on 30 measurements.



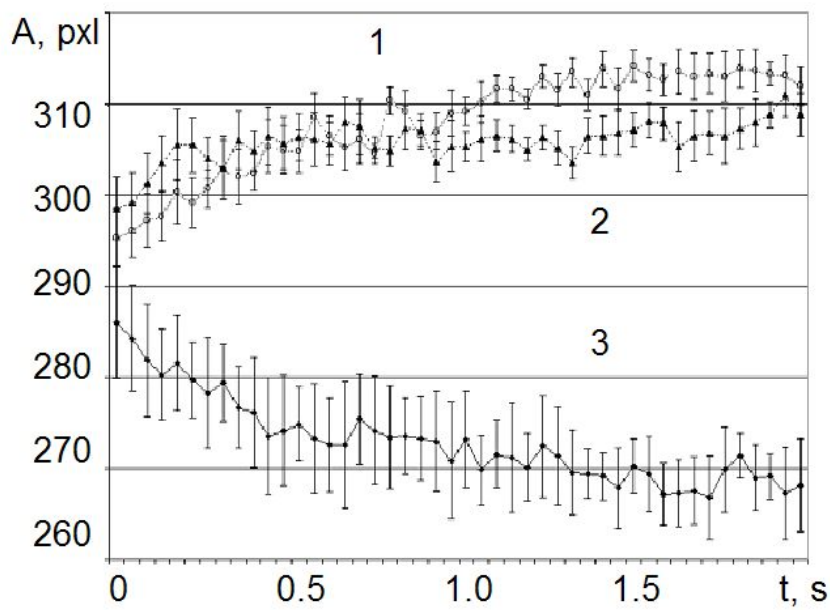
2. Time dependence of the area of the gas discharge image around drops of an NaCl solution of different concentrations (1-6) and distilled water (7). Data averaged on 10 measurements. Concentrations: 1: 1N; 2: 1/6 N; 3: 1/64 N; 4: 1/1024 N; 5: 1/4096 N; 6: 1/32768 N.



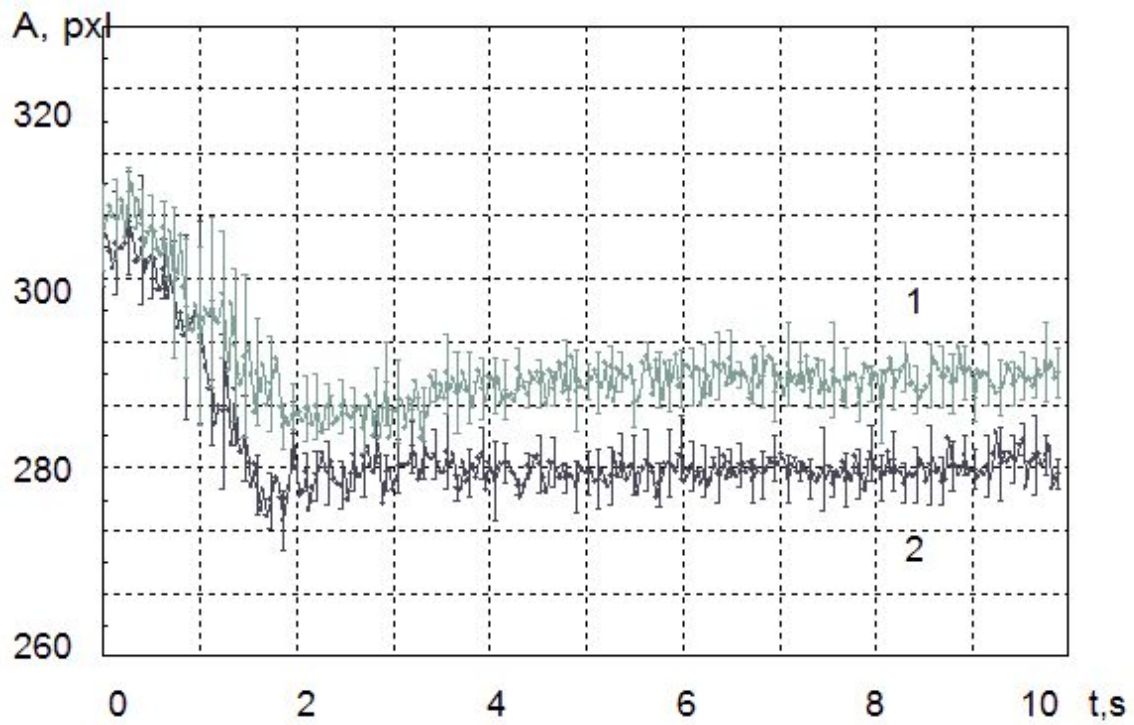
3. Time dependence of the area of the gas discharge image around drops of NaCl (1) and KCl (2) solutions both of 1N concentration. Data averaged on 10 measurements.



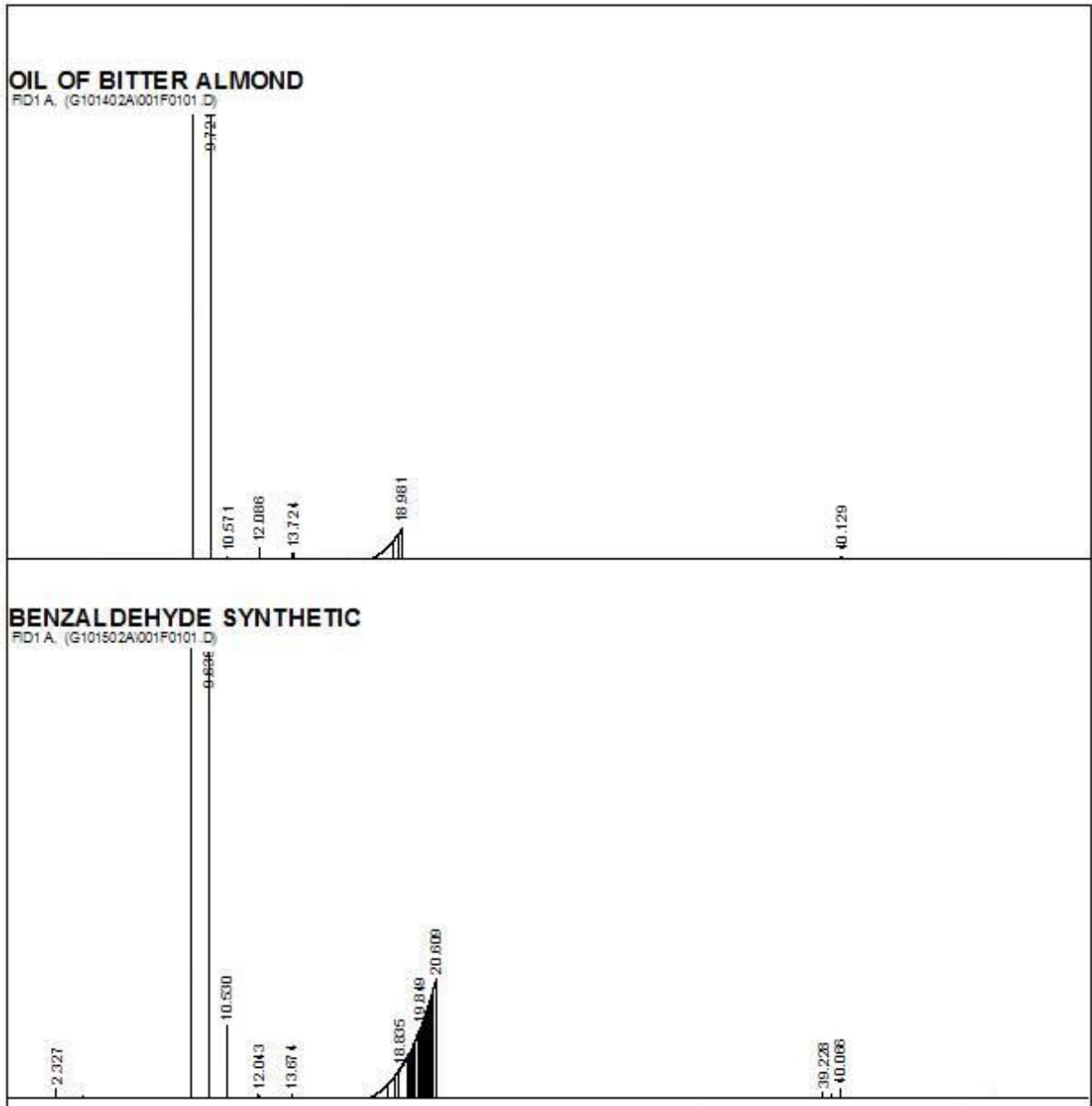
4. Time dependence of the intensity of the gas discharge image around drops of KNO_3 (1) and KCl (2) solutions both of 1N concentration. Data averaged on 10 measurements.
- 5.



5. Time dependence of the area of gas discharge image around drops of Rose oil of different origin. Data averaged on 10 measurements. 1 – Morocco Rose; 2 – Bulgarian Rose; 3 – Russian Rose.



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.



7. 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).

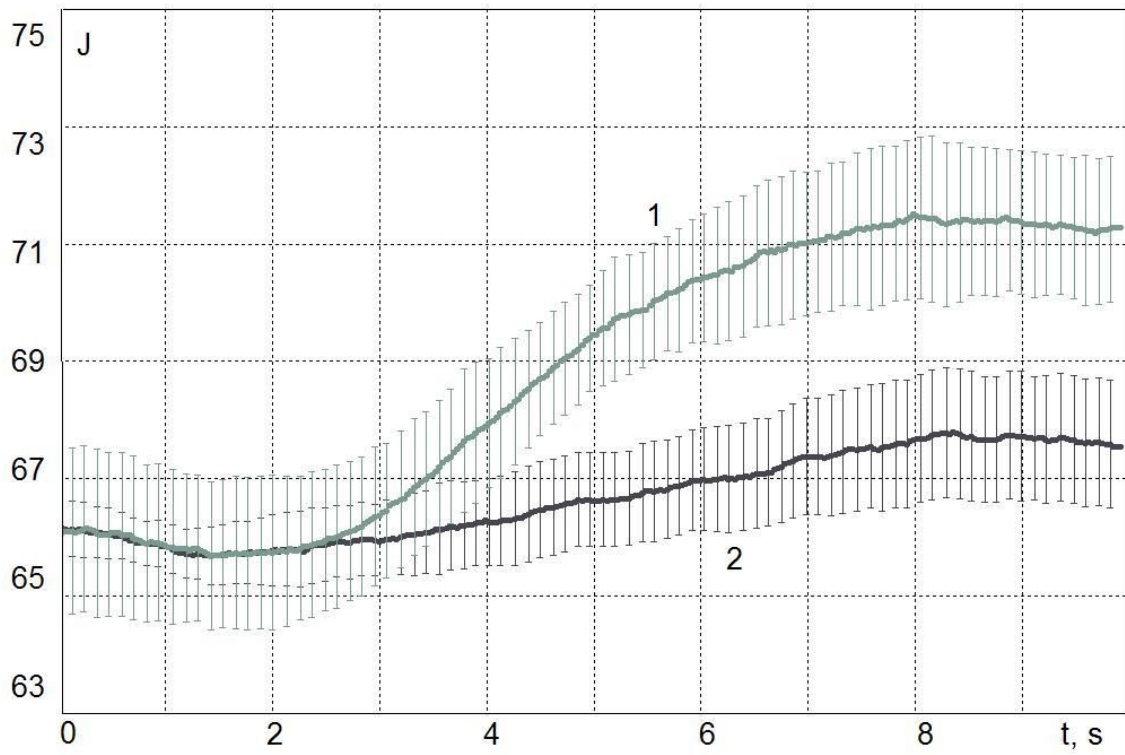


Fig.8. Time dependence of the area of gas discharge image around drops of oils Citral natural (1) and Citral synthetic (2). Data averaged on 10 measurements.