Concentration dependence of gas discharge around drops of inorganic electrolytes

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This article is devoted to the study of image formation in gas discharge, initiated by a strong impulsive electromagnetic field around drops of four different nonorganic electrolytes. To describe the image mathematically we propose several parameters: the form coefficient (fractality), the entropy, and the average streamer width. We study the dependence of these parameters on concentration. The form coefficient turns out to have the best combination of stability and sensitivity in the whole range of concentrations. Statistically significant difference between the solutions and distilled water disappears at concentrations of about 2^{-20} N. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1360700]

I. INTRODUCTION

Image formation in gas discharge around objects of a different nature initiated by strong impulsive electromagnetic fields (also called the Kirlian effect) has been known for more then two centuries.[1,2] So far the main direction of investigation of the effect has been purely practical; It turned out that gas discharge images around biological objects can provide substantial information about the internal state of the object. In particular, the gas discharge images of human fingers and toes are actively used by physicians for diagnostic purposes (see Ref. 2 for a review). This generated a number of research works devoted to the physical nature of image formation.[1,3–7] It turns out that one of the most difficult problems is finding an adequate quantitative description of the process. The main obstacle to an effective mathematical description is the high nonlinearity of gas discharge. Moreover, this nonlinearity comes on top of extreme complexity of the biological objects themselves.

The authors of Ref. 6 compared gas discharge images around drops of water solutions of several nonorganic salts using the traditional method of gas discharge photography. The authors introduced several numerical parameters characterizing gas discharge around solution drops, which in particular correspond to size and shape of separate streamers. Using these parameters they were able to demonstrate essential quantitative differences between gas discharge pictures around drops of solutions of different salts at different concentrations.

However, there are several reasons which led us to attempt to further understand gas discharge around drops of different inorganic electrolytes. First, traditional gas discharge photography is largely replaced by computer image processing.[2] The images produced in this way turn out to be essentially different from the images obtained by use of standard photographic tools. Computerized gas discharge cameras have many obvious advantages, although it turns out to be difficult to directly apply the mathematical description of Ref. 6 to computer images of gas discharge. Ideally, adequate mathematical tools should be stable with respect to the method of measurement, and, in particular, to the type of device used to obtain a gas discharge image of the given object. However, they should also be sensitive enough to reveal small fluctuations of the object.

As far as water solutions of different salts are concerned, in our opinion, this is an excellent laboratory for testing the methodology of gas discharge visualization before trying to further apply rigorous mathematical tools to gas discharge images of more complicated objects.

Therefore, the purpose of this article is twofold. First, we introduce several mathematical parameters characterizing the gas discharge image (the form coefficient, entropy, average streamer width). All these parameters are borrowed from the theory of signal and image processing, and appropriately adjusted to describe the gas discharge images. Second, we test the applicability of these parameters to explicit description of gas discharge images around drops of different electrolytes: NaCl, $NaNO_3$, KCl, KNO_3. In particular, we evaluate the average error resulting from the stochastic nature of the gas discharge process itself. The main focus will be on the dependence of these parameters on concentration.

II. BASIC PRINCIPLES OF GASEOUS DISCHARGE VISUALIZATION TECHNIQUE: EXPERIMENTAL SCHEME

The scheme of the experiment is shown in Fig. 1. By a vacuum photogalvanoplastic process a thin metal grid with 10 mkm wires is evaporated onto the bottom surface of the glass plate. A train of duration 0.1 s of triangular 10 mks electrical impulses of amplitude 3 kV, steep rate 10^6 V/s and repetition frequency 10^3 Hz is applied to this grid. This gen-

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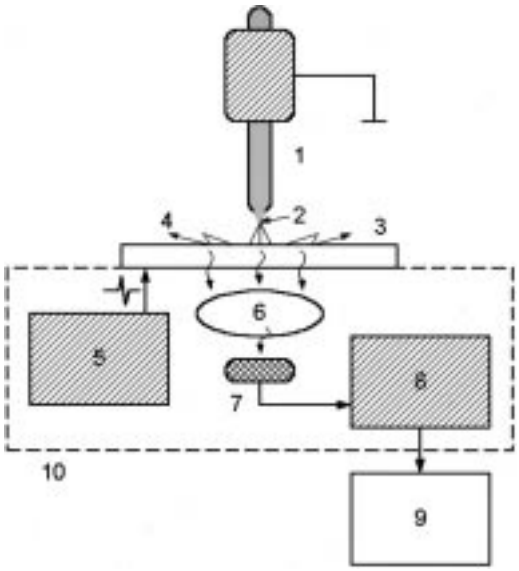

FIG. 1. The experimental scheme. (1) container with liquid; (2) liquid drop; (3) optical glass with coating underneath; (4) gaseous discharge; (5) impulse generator; (6) optical system; (7) CCD camera; (8) video digitizer; (9) IBM PC; (10) gaseous discharge device as a whole.

erates electromagnetic field around the solution drop 2 located at a distance of 3 mm from the top surface of the plate. Under the influence of the field the drop produces a burst of electron-ion emission and optical radiation light quanta in the visual and ultraviolet range. These particles and photons initiate electron-ion avalanches, giving rise to a sliding gas discharge along dielectric surface.[8] The spatial distribution of discharge channels is recorded through the glass plate by the optical system 6 with charge coupled device TV camera 7 and digitized in the computer 9 using video-blaster 8. In short, this technique is called the BEO GDV technique and images after processing are called GDV grams.[2]

The main aim of our experiment was investigation of concentration dependence of a gas discharge image around the solution drop. The experiment was carried out for four salts: NaCl, KCl, $NaNO_3$, and KNO_3 in an attempt to understand the influence of different ions on the parameters of gas discharge images. We used the salts refined to degree analytical reagent grade (AR); It turns out that the use of less refined salts essentially distorts the final results. The distilled water used in the experiments was refined up to conductivity 10^{-6} $cm^2\,\Omega^{-1}$(g.-equv.)$^{-1}$. We studied the dependence of form coefficient, entropy, and average streamer width of the gas discharge image (see Sec. III for precise definitions of these parameters) on concentration.

The experiments were carried out as follows: we start from standard 1 N solution and make subsequent double dilutions until the significant difference between GDV images of solution and GDV images of distilled water disappears.

The temperature was kept in the interval 22,0°–22,5 °C; humidity was kept in the interval 42%–44%. For each concentration of each salt 40 pictures of the gas discharge were taken to make possible a sufficiently reliable statistical analysis of experimental data at each point. These 40 pictures were produced in the following way: we took eight

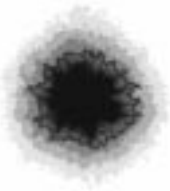

FIG. 2. Typical image of gas discharge around a drop of 1 N solution of NaCl. We depict the isoline corresponding to the average luminosity of the image. Form coefficient of this image is equal to 2.09, entropy is equal to 5.46, average streamer width is equal to 12.3.

different drops of the solution, and for each drop we studied five subsequent images of gas discharge. The reason for change of the drop after five measurements was the observed distortion of parameters of GDV image of the drop under the influence of larger number of subsequent measurements.

III. MATHEMATICAL DESCRIPTION OF GAS DISCHARGE IMAGES AROUND SOLUTION DROPS

In Fig. 2 we show a typical gas discharge image around the drop of 1 N solution of NaCl. This image is a set of pixels with a certain luminosity assigned to every pixel. Generically, in the center of the image the luminosity is maximal; Moreover, as a rule, it monotonically decreases along any radial direction.

The mathematical description of gas discharge images of drops of inorganic solutions is less complicated compared to biological objects, since we can assume that the drop is rotationally symmetric. Therefore, all the deviations from rotational symmetry observed in the gas discharge image are due to random processes of gas discharge itself. This allows one to radically decrease the number of numerical parameters sufficient for characterization of the image.

To process the image numerically, we first find the position of its centroid (with luminosity playing the role of image density). Then we construct curves of constant luminosity (isolines), which are closed curves surrounding the centroid. In principle, it is sufficient to restrict ourselves to any of the isolines to extract relevant mathematical information. However, it turns out that to achieve the best combination of stability and sensitivity of the parameters, it is convenient to take the isoline corresponding to the average luminosity of the image (Fig. 2).

This curve can be described numerically as a set of numbers f_n, where $n=1,\ldots,N$ is a ''discrete angle'' (our images have resolution sufficient to divide the whole 2π angle into 1028 pieces, i.e., we take $N=1028$). Number f_n measures (in pixels) the distance from the centroid to the isoline.

The following integral parameters characterizing the set f_n turn out to provide an optimal mathematical description of the image:

(1) Form coefficient (fractality). There are several ways to describe the fractality of any plane figure numerically. Here we shall use the notion of form coefficient which measures the deviation of our line of constant luminosity from a circle. It is given by

$$F=\frac{L}{2\pi\bar{f}}, \tag{1}$$

where L is the length of the line, and

$$\bar{f}=\frac{1}{N}\sum_{n=1}^{N} f_n, \tag{2}$$

is the mean value (i.e., ''average radius'' of the isoline). Obviously, if the isoline is a circle (i.e., all f_n are equal) then F is (very close to) 1. The discrete version of formula 1 in terms of the numbers f_n is as follows:

$$F=\frac{\sum_{n=1}^{N}\sqrt{\left(\frac{2\pi}{N}\right)^2 f_n f_{n+1}+(f_{n+1}-f_n)^2}}{\frac{2\pi}{N}\sum_{n=1}^{N} f_n} \tag{3}$$

(we assume $f_{n+N}\equiv f_n$). Generically, the greater the value of F compared to 1, the more fractal is the isoline, and, therefore, the more chaotic is the gas discharge process.

(2) Average streamer width (maximal angle of essential autocorrelation): We define the autocorrelation function[9] which for any periodic function $f(x)$ with period 2π and zero mean value has the following form

$$K(y)=\frac{\int_0^{2\pi} f(x)f(x+y)dx}{\int_0^{2\pi} f^2(x)dx}. \tag{4}$$

Obviously, $K(0)=1$; for all other values of y we have $K(y)\leqslant 1$. If at some value of y function $K(y)$ reaches its (local) maximum, it means that there exists correlation between functions $f(x)$ and $f(x+y)$ at that value of y. If function $f(x)$ is random, the autocorrelation function is equal to 1 in a very small neighborhood of $y=0$ and $y=2\pi$ and fluctuates around zero at all other values of y. The discrete version of formula (4) looks as follows:

$$K_m=\frac{\sum_{n=1}^{N}(f_n-\bar{f})(f_{n+m}-\bar{f})}{\sum_{n=1}^{N}(f_n-\bar{f})^2}, \tag{5}$$

where we subtract the mean value $\bar{f}$ to guarantee that

$$\sum_{n=1}^{N}(f_n-\bar{f})=0. \tag{6}$$

Taking into account the rotational symmetry of water drop, we conclude that the rotational asymmetry of a gas discharge process should be essentially random, at least at the surface of the drop. Therefore, the correlations of a gas discharge image at different values of rotation angle should be low. However, the autocorrelation function always has value 1 at $m=0$. Thus it has to fall towards the zero value at some relatively small m. This value can be naturally interpreted as the average width of streamers.

In the sequel we shall denote the average streamer width as W with value (measured in degrees):

$$W=\frac{360m}{N}, \tag{7}$$

where m is the minimal index for which

$$K_m\leqslant 0.3,\quad K_{m-1}\geqslant 0.3 \tag{8}$$

(an empirically chosen value of 0.3 provides optimal stability and sensitivity of parameter W).

(3) Entropy: The notion of entropy plays an important role in probability theory. It is central in statistical physics, the theory of information, and coding theory. In particular, it was successfully applied to analysis of human encephalograms.[10] The entropy turns out to be relevant as well in the mathematical description of gas discharge images. In principle, to each image one can naturally associate several different notions of entropy.[11] Here we introduce the notion of entropy which seems to be most suitable in applications to gas discharge images. If we consider f_n as a random variable (which is a reasonable assumption if the autocorrelation function K_m is confined to a small neighborhood of 0 for all values of m except for the values close to 0), we can find the corresponding probability distribution $p(f)$. In practice, we approximate f_n by the closest integer number of pixels. Suppose that we have M integers confined between the minimal ($f_{\min}$) and maximal ($f_{\max}$) values of f. For each kth value from this set we denote by P_k the number of f_n's equal to that value. Then the probability of random variable f to take its kth value may be identified with

$$p_k=\frac{P_k}{N}, \tag{9}$$

and the corresponding entropy is given by the formula

$$\mathcal{E}=-\sum_{j=1}^{M} p_j \log p_j. \tag{10}$$

We call $\mathcal{E}$ the entropy of gas discharge image.

IV. RESULTS

(1) For each concentration of each solution the distribution of all parameters (form coefficient, entropy, and average streamer width) turns out to be close to Gaussian. The relative reliability interval of these parameters corresponding to 40 measurements of each point varies from 1% to 8% depending on parameter, salt and concentration. The relative reliability interval is minimal for form coefficient of distilled water (about 1%) and maximal for form coefficients of electrolytes at large concentrations (about 7%–8%).

(2) For distilled water the form coefficient is equal to 1.38 ± 0.01; subsequent measurement of the same drop shows a slow increase of the form coefficient; for example, after the fifth measurement we get value 1.42 with the relative reliability interval about 1%. After seven to eight measurements the tendency of F to increase persists, but the relative standard deviation gets increased about three times. For electrolyte solutions the change of the object under the

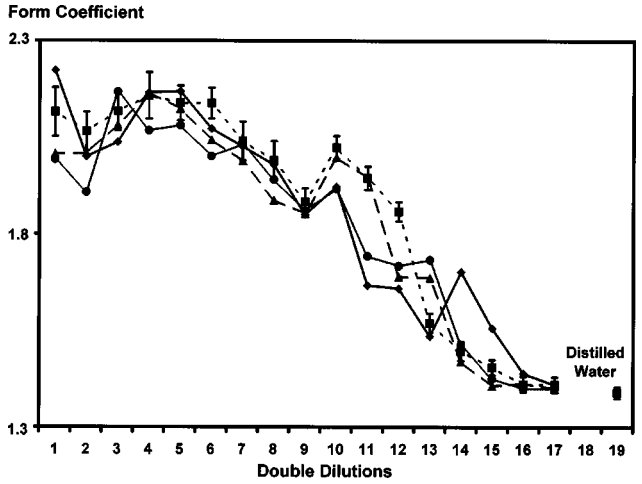

FIG. 3. Dependence of form coefficient on concentration for NaCl (boxes), $NaNO_3$ (triangles), KCl (diamonds), and KNO_3 (circles). The reliability intervals are shown only for NaCl to keep the image clear.

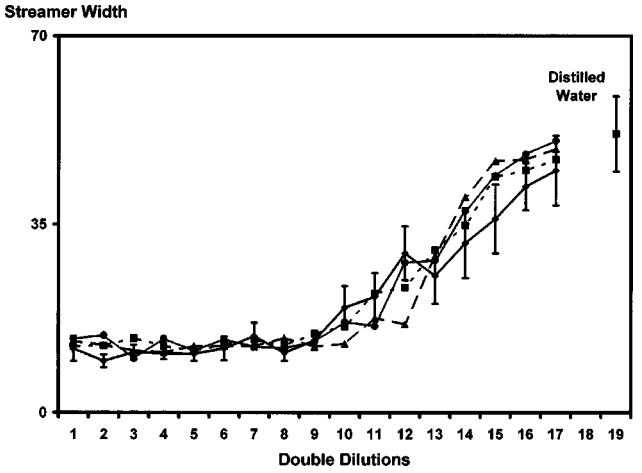

FIG. 5. Concentration dependence of average streamer width (measured in degrees). Notations are the same as in Figs. 3 and 4. The reliability intervals are shown for KNO_3.

influence of the measurement process turns out to be even higher. We conclude that measurements of the same drop for more then seven times highly distorts all the results; This is the reason for restricting the number of measurements of the same drop to 5.

(3) Dependence of form coefficient on concentration for all four salts together with reliability intervals for NaCl is shown in Fig. 3. The general form of the curve looks rather similar for different electrolytes. The most significant difference is observed at a concentration of 2^{-11} N between the pair KCL and KNO_3 and the pair NaCl and $NaNO_3$. An unexpected feature is the essentially nonmonotonic character of the curve at concentrations 2^{-1} N and 2^{-8}–2^{-9}N. It is worth noticing that the point 2^{-1} N is the point of complete dissociation of all these electrolytes. The form coefficient shows good sensitivity and stability within the whole range of concentrations.

(4) Dependence of entropy on concentration (Fig. 4) has monotonically decreasing character; curves corresponding to different electrolytes are almost undistinguishable. For an illustration we also show the reliability intervals for $NaNO_3$. The reliability interval corresponding to 40 measurements varies from 3% at higher concentrations to 8% at lower concentrations. The entropy shows very good stability properties, but it is less sensitive to the electrolyte type than the form coefficient. Nevertheless, the entropy has better sensitivity at concentrations lower than 2^{-10} N.

(5) The average streamer width (Fig. 5) depends monotonically on concentration for all four electrolytes and is insensitive to the type of electrolyte. It is essentially independent of concentration at concentrations higher then 2^{-10} N. The reliability intervals are shown for KNO_3. At lower concentrations the average streamer width is a sensitive parameter, although there are large relative reliability intervals corresponding to 40 measurements of each point (about 9%).

(6) The statistically significant difference between parameters of electrolyte solutions and distilled water disappears at a concentration of 2^{-20} N.

(7) Figure 6 depicts the dependence of form coefficient on conductivity of solutions for KNO_3. It turns out that for KNO_3 (as well as for other salts) this dependence may be

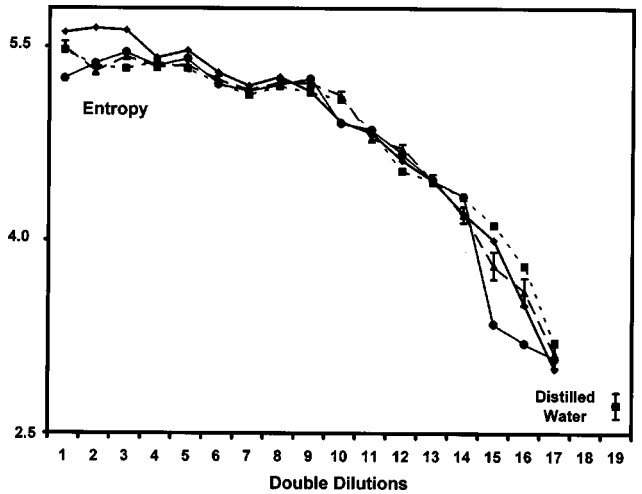

FIG. 4. Dependence of entropy of gas discharge image on concentration. Notations are the same as in Fig. 3. The reliability intervals are shown for $NaNO_3$.

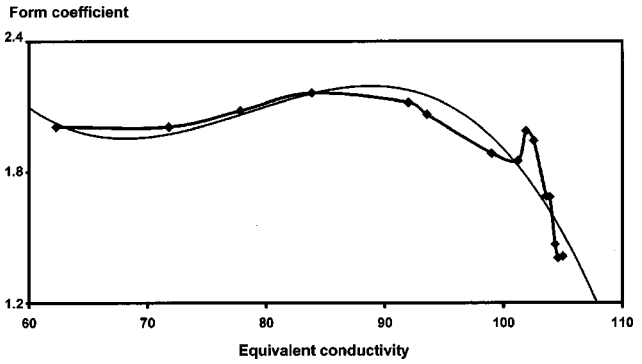

FIG. 6. Dependence of form coefficient on equivalent conductivity [measured in $cm^2\,\Omega^{-1}(g.-equiv.)^{-1}$] for KNO_3 is shown in bold line. Approximation of this dependence by the third order polynomial $-5\times10^{-5}x^3+0.013x^2-x+27.2$ is shown in regular line.

nicely approximated by a third order polynomial. It seems tempting to speculate about a possible correspondence between coefficients of these polynomials and basic properties of solutions.

V. CONCLUSIONS AND OPEN PROBLEMS

We conclude that the concentration dependence of gas discharge images around electrolyte drops admits a rigorous mathematical description using any of three introduced parameters (form coefficient, entropy, and average streamer width). The form coefficient is the most universal parameter which shows a good combination of stability and sensitivity properties in the whole range of concentrations. However, at low concentrations the average streamer width and entropy are more sensitive than the form coefficient. We deliberately did not consider such parameters of the gas discharge image as total area or total brightness of the image which were actively used in this context previously.[2] The main reason for omitting them is that these parameters are not dimensionless and, therefore, are extremely sensitive to the change of resolution, type of device, experimental conditions, etc.

As far as the methodology of the gas discharge visualization is concerned, it turns out that the properties of the electrolyte drops change under the measurement process. Therefore, to make sure that different measurements of the same drop are independent, one has to restrict the number of measurements of the same drop to a maximum of 5. We expect that the measurement has an even stronger influence on the properties of organic objects. Another feature of the method is an essential distortion of all results if one uses less refined than analytical reagent grade (say, chemically pure grade) salts or less refined distilled water.

We plan to continue the present research in several directions.

(1) As far as gas discharge processed around nonbiological objects is concerned, one must clarify the meaning of nonmonotonic parts of dependence of form coefficient on concentration (Fig. 3). The main problem here is to achieve better understanding of the points where one gets statistically significant distinction between different electrolytes. It would certainly be desirable to clarify which properties of electrolytes are responsible for this distinction. For that purpose one has to repeat the same experiment for an essentially larger variety of different solutions.

(2) A problem of intermediate difficulty is the analysis of gas discharge around biological liquids (blood, urine, etc.) and their dilutions, also often used by physicians for diagnostic purposes. On one hand the rotational symmetry of the object (solution drop) is preserved, which should make possible the use of analytical methods proposed in this work. On the other hand, the extremely high complexity of biological liquids should probably lead to a very essential change of the properties of the drop under the influence of measurement.

(3) The development of a mathematical description of gas discharge images around fingers and toes seems to be an essentially more appealing problem due to basic asymmetry of the object. Therefore, to extract the relevant information from these images one has to exploit a much larger set of parameters.

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