The nature of oscillations of ion currents in the ion track electronics

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Abstract

The paper contains the description of the main features of ion current pulsations in track devices. The conditions under which the pulsations arise are discussed. We describe different approaches that are used for interpretation of the effect of ion current pulsations. In particular the generalized model of current spikes in track devices is considered. To create this model a special modification of the classical molecular dynamics was developed. The results of application of this model coincide with the main experimental data concerning the ion current pulsations in track devices.

Keywords: track devices ion current pulsations nanoporous membranes

1 Introduction

Since the sixties of the past century it is known that energetic (with tens of MeV or more) heavy (with atomic masses being usually larger than that of Ar) ion irradiation ("swift heavy ions", SHI) introduces very narrow (~ some nm) but long (typically 10-100 μm) parallel trails of damage in irradiated polymer foils, the so-called latent ion tracks. The damage shows up primarily by the formation of radio-chemical reaction products. Whereas the smaller ones readily escape from the irradiated zone thus leaving behind them nanoscopic voids, the larger ones tend to aggregate towards carbonaceous clusters. Thus emerging structural disorder along the tracks modifies their electronic behavior.

The newly created intrinsic free volume enables electrolytes to penetrate into the polymer, thus forming parallel liquid nanowires. In case that the tracks penetrate through all the foil the conducting connections emerge between the foil front and back sides.

Upon proper design, the irradiated polymer foils may exhibit electronic properties that mimic bioelectronic functionalities, as they resemble somewhat biological membranes which also contain a number of parallel electrolytes - filled nanopores.

The ion track technology is, in particular, directed towards biosensing applications. In this case the ion tracks are functionalized directly by attaching organic or bioactive compounds (such as enzymes) to their walls. The recent advances in this field allow monitoring and tracking biomolecules in areas such as environment, food quality and health. The presently developed ion track-based nanosensors provide high sensitivity, low power and low cost [1].

The creation of new biosensors and their further improvement requires a careful study of the mechanisms of electrolytes passage through the tracks.

2 Collective interaction in ion track electronics

In [2] the capability of a multitude of parallel electroactive nanostructures in a given substrate to show collective interactions has been considered. Two types of electroactive nanostructures are described: (a) electrolyte-filled current spike-emitting latent ion tracks in thin polymer foils, (b) metal cluster-filled etched ion tracks in TEMPOS structures (i.e. in thin SiO_2 layers on Si substrates). Both these electroactive nanostructures can be operated either by applying a constant voltage to them, or by application of a sinusoidal voltage at low frequency, and in both cases current spike emission can be triggered. An electroactive nanostructure can influence the performance of neighboring nanostructures by modifying the entrance or exit potentials (or both) of the latter one, via lateral charge exchange through the common front or backside conductors or contacts. For these two cases, this leads to two different effects: The collective interaction of many current spike-emitting latent tracks in electrolytic ambient leads to pulse-locked synchronization similarly to its representation in Neural Network theory.

In TEMPOS structures with etched tracks in SiO_2 on Si and with metal nanoclusters coverage of the oxide layer and the track walls (with at least two contacts on the oxide surface and one on the Si substrate), the collective track interaction can induce negative differential resistance [3]. This is the consequence of a chain reaction triggered by spontaneous opening of previously closed (or closing of previously open) neighbored tracks. Periodic repetition of such opening/closing processes leads to self-pulsating devices.

3 Oscillating currents through nanoporous membranes in electrolytes

The effect of current pulsations when ions pass through the tracks is of great interest for creation of new biosensing devices. [4-7]. There are different attempts to explain the phenomenon of current oscillations in track-containing foils embedded in electrolytes.

To describe the properties of nanopores (in particular the ion transport in tracks) different models and mathematical methods are used. For example, the current through nanotracks is described by stationary Poisson Nernst Planck equations [8, 9]. Molecular dynamics simulation is used in [10, 12] to describe the ion current rectification.

One of the models connects these oscillations with carbonaceous clusters that form along the latent tracks [7]. These clusters might behave as obstacles for the smooth ionic current passage along them, upon application of a DC or low frequency AC voltage across the track-containing polymer foil. As a result, charges may pile up in front of them until the intrinsic electric field across them exceeds the breakthrough field strength. At that moment current spikes eventually associated with negative differential resistances emerge. As the spike height is decreased by eventual surface adsorption layers, pulsating tracks can also be exploited for biosensing [13]. Foils with current spike emitting tracks are thought to mimic neurons. In a multitude of such tracks, the individual randomly emitted spikes synchronize themselves towards phaselocked oscillations [14, 15], similarly as they occur for neurons in the human brain, where their interaction results in the formation of brain waves. The frequency of these collective track pulsations is around 0.1…30 Hz [2]. Hence in order of magnitude which is similar to brain waves. The presently available neural network theory describes the behavior of pulsating tracks at least qualitatively well.

In [16] it was suggested that the current oscillation mechanism is linked to the competition of two processes in the tracks: a) The adsorption of charged ions at the internal track walls and b) The ion spike when the number of accumulated ions reaches some critical value. The negative charge of the pore walls [17] causes the adsorption of positive ions. Also chemical reactions may influence this adsorption. During accumulation of the adsorbed ions at the inner surfaces of the tracks the repulsive forces begin to prevent the penetration of new positive ions into the tracks, hence the ion current decreases. At some threshold voltage applied to the track and for some critical number of adsorbed ions an ion spike emerges which leads to desorption of the ions accumulated inside the track, hence to an increase of the ion current. The current maximum corresponds to the open track with a minimum of adsorbed ions. Thereafter newly adsorbed ions can be accumulated so that the process repeats. Hence an oscillation current emerges with a frequency $\nu = 1/\tau_1$, (τ_1 being the period of oscillations) that is determined by the rate of accumulation of adsorbed ions and by the probability that an ion spike occurs, the latter depending on the applied voltage. In order that ions can penetrate at all through the track, the track radius must exceed a certain threshold value r_{min}, which is determined by the thickness of the ion adsorption layer. On other hand, for too large track radii r_{max} the adsorption layer does not control the ion penetration through the track. Thus, current oscillations will occur only for track radii r with $r_{min} < r < r_{max}$.

The above model does not take into account the possible interaction of nanopores.

Hence this refers to the case of one track per membrane only. In the case of many tracks per membrane the statistical interaction between them modifies the conditions that determine the oscillation frequencies. Here, the ion current minimum corresponds to the situation when most tracks are closed simultaneously. Such a situation is realized as a result of statistical interaction processes and demands some time (τ_2). This interaction may be ionic charge equilibration processes between neighboring tracks via currents running from one charge carrier cloud near a track entrance and/or exit to the other, or via the (more improbable) ionic diffusion across the tracks, and by different adsorption conditions in different tracks. A realization of the situation when almost all tracks are open demands also a given time (τ_3).

It is clear that we have $\tau_1 << \tau_2 + \tau_3$: in the case of many tracks the frequency of current oscillations is much less than in the case of one individual track. In the case of many tracks per membrane the current oscillations do not drop to zero as in the case of individual tracks because in the first case some tracks are open always.

4 Generalized model of current spikes in track devices

For ion current pulsations in track-containing foils the following features are established [18, 19]:

- Spike emission depends on the amplitude and frequency of the applied voltage;
- The high ion track density (higher than some threshold density) is necessary to obtain the effect of current spikes;
- Current spikes preferentially occur at pronounced, rather equidistant applied voltages;
- Maximal spike heights do not seem to be affected markedly by the frequency of the applied voltage;

- Current spike spectra are not always reproducible though their principle features remain the same;
- Current spike emission appears to vanish rapidly with the frequency of applied voltage. Its decrease indicates the existence of a threshold frequency for spike emission.

The model described in [20] allows studying the general case of the ion current pulsations in the track-containing polymer foils embedded in electrolyte. A schematic representation of the appropriate structure can be seen in Fig. 1. To construct the model the classical method of Molecular Dynamics (MD) was modified so that a new MD approach allows describing subthreshold radiation effects [21].

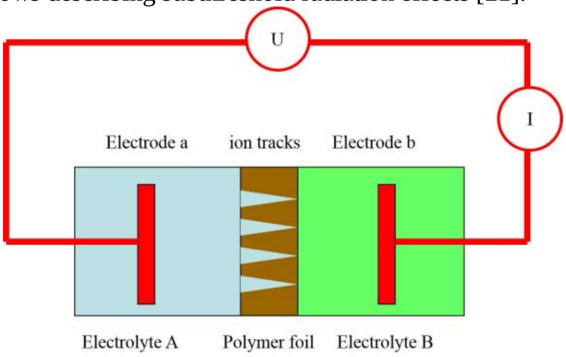

FIGURE 1 Principle arrangement of experimental setup to study current spike emission in ion track-containing foils embedded in electrolytes (current/voltage measurements)

4.1 MODIFICATION OF CLASSICAL MOLECULAR DYNAMICS

To create the model of current spikes in track devices a modification of classical MD was performed in [22-24]. In the MD method the classical equations of motion with an appropriate potential of the interaction between particles are solved. The Verlet algorithm [25] is usually used to solve these equations.

To simulate the effect of atomic collisions in the modified MD method the "shock function" (FSH) is introduced. Then in the Verlet algorithm the total force acting on atom i is presented by expression:

$$\vec{F}_i^n = -\sum \frac{\partial \Phi(\vec{r}_{ij})}{\partial x_i} + \vec{F}_{SH} , \quad (1)$$

where $\Phi(\vec{r}_{ij})$ is the interatomic potential and $r_{ij} = |\vec{r}_i - \vec{r}_j|$ is the distance between atoms *i* and *j*.

The pulses that are transferred to lattice atoms during irradiation are characterized by special random function (RF). This function shows which atom in the irradiated sample is knocked, what energy value is chosen from the selected energy interval, and what the direction of hit is. RF inserted to the computer program performs three tasks:

- Selects an atom in the target lattice which gets a hit;
- Selects an energy value from the interval ($\varepsilon_1, \varepsilon_2$);
- Selects an orientation of the pulse transferred to the target atom.

A linear congruent generator for RF is an algorithm that yields a sequence of pseudo-randomized numbers calculated with a discontinuous piecewise linear equation. This generator presents one of the best-known pseudorandom number generator algorithms that are easily implemented and fast, especially on computer hardware which can provide modulo arithmetic by storage-bit truncation. These generators based on linear congruent method are especially useful for non-cryptographic applications, such as modeling. They are effective and most used in empirical tests and show good statistical characteristics.

The generator is defined by the recurrence relation:

$$X_{n+1} = \left(aX_n + c\right) < m , \quad (2)$$

where X_0 is an initial value. In our model the parameters are: m = 232, a = 214013, c = 2531011.

In order to determine the kinetic energy transferred to the lattice atom the scaling of the forces FSH should be implemented. As a reference point we used the energy that is necessary for irreversible displacement of lattice atom to interstitial position in elastic collision (Ed). Then the following relation may be written:

$$\frac{(F_{SH}t)^2}{2M} = E_d , \quad (3)$$

where *t* is duration of the action of the force in the process of one hit.

In computer experiment the value of the force FSH was gradually increased to the point where the atoms begin to move irreversibly to interstitial positions. This magnitude of the force FSH corresponds to Ed ≈ 25 - 30 eV.

To apply the proposed approach a model crystal with a cubic lattice that consists of 8000 atoms is constructed. Lenard-Jones potential [26] was used. These parameters were slightly varied to stabilize the lattice of the model crystal.

The proposed approach allows studying so called subthreshold radiation effects in solids [21]. Such mechanisms are used in the model.

4.2 DESCRIPTION OF THE MODEL AND RESULTS OF SIMULATION

In the model the track structure is represented as a two-dimensional system of currents. Really, in any plane that intersects the tracks one can fix some current value in each track (see Fig. 2). Therefore in the indicated plane we have *a set of currents* that perform oscillations in the appropriate conditions. The model describes a system of pulsating currents regardless of the material, in which the tracks are created, and does not include the description of real material and tracks themselves. Current pulsations are simulated by oscillations of model particles. Their masses are varied in a large range of values (five orders) in order to get a wide variety of oscillation spectra and compare their qualitative view with views of experimental pulsations of currents. To simulate the real experimental conditions the value of SF and its frequency were varied.

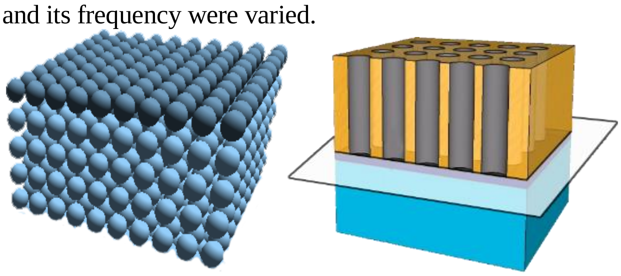

FIGURE 2 The model crystal (on the left) and the model for pulsating ion currents (on the right)

In the simulation experiment it was assumed that the average amplitude of the model particles oscillations correspond to the average spike heights, the value of SF – to the value of applied voltage, and the frequency of SF action – to the frequency of applied voltage. The values of SF were chosen so that model particles (MP) did not leave irreversibly their nodes under the action of SF. The directions of SF action were determined by the random function and change within the upper hemisphere which corresponds to the directions of motion of the ions in the track. As a result of action of SF, regular small oscillations of MP as well as large amplitudes arise as shown in Figure 3. This figure demonstrates the similarity of oscillation spectra in real experiment and as a result of computer modelling.

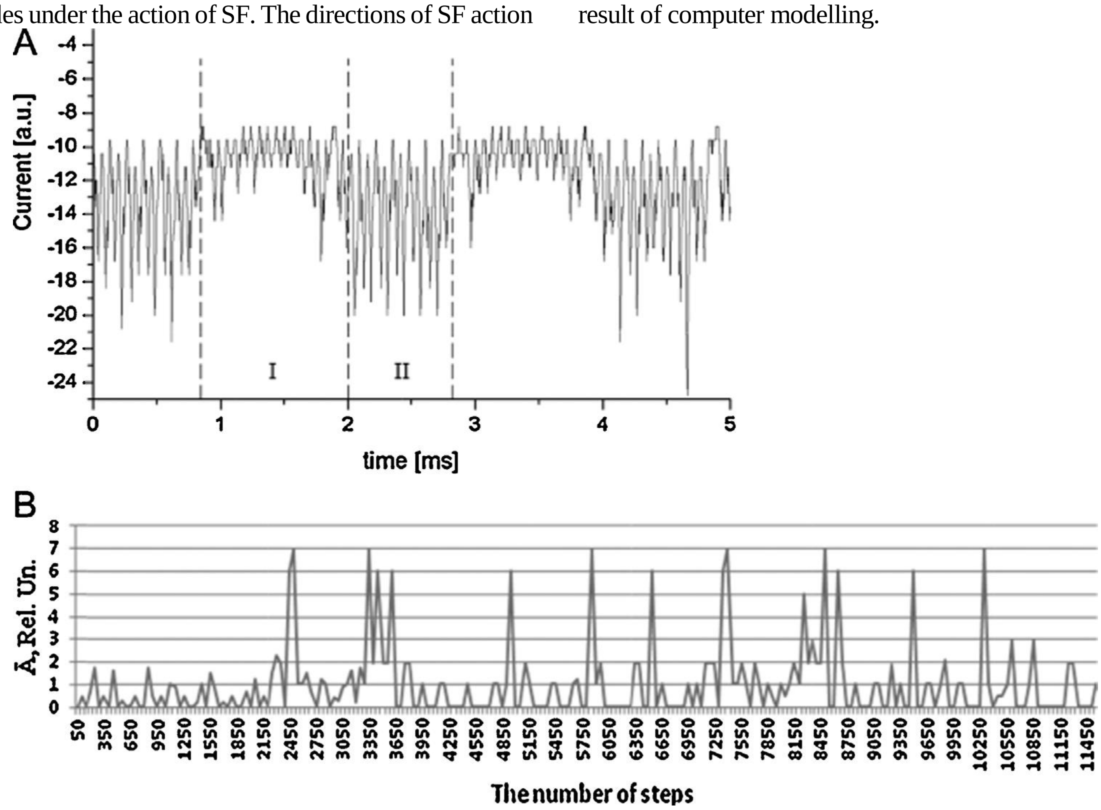

FIGURE 3 Current spikes in conditions of real experiment (on top). Region I corresponds to regular oscillations, region II corresponds to current spikes. Illustration of model spikes in the model experiment (down). At the horizontal axis is the average amplitude of MP oscillations.

The dependence of the average amplitude A of MP oscillations on the value of SF is displayed in Figure 4 (on top). This dependence is in good agreement with the dependence of the average spike height on the value of applied voltage shown in Figure 4 (down). In Figure 5 (on top) the dependence of the average amplitude of MP oscillations on the frequency of SF action is shown. The corresponding dependence of the average spike height on the frequency of applied voltage is presented in Figure 5 (down).

The dependence in Figure 5 can be explained by a "memory effect" which manifests itself in the mechanism of spikes formation: each next spike "remembers" the information about previous spikes. As a result of this effect according to the model there is some optimum frequency of the external exciting factor that provides a maximum average spike height. It means that at higher frequencies of the SF action the model lattice is too disordered after the previous spikes, and the conditions for the synchronization of individual spikes are less favorable. On the other hand, at too low frequencies the model lattice is completely restored after the previous spikes ("forgetting about the previous disordering") and this worsens the conditions for the formation of new integrated spikes.

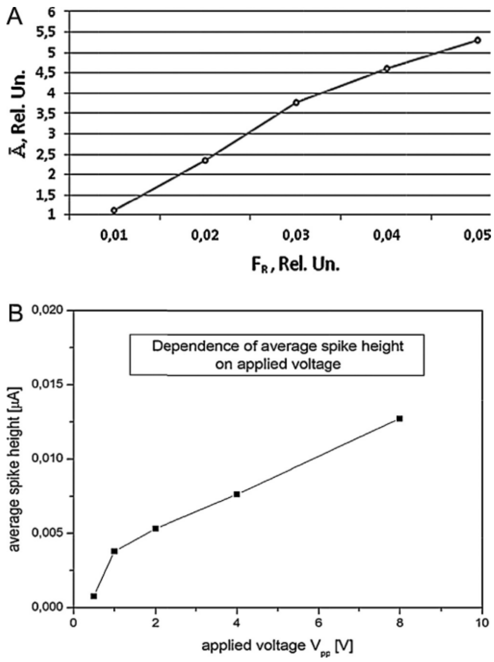

FIGURE 4 Dependence of the average amplitude of MP oscillations on the value of SF (on top); Dependence of the average spike height on the value of the applied voltage (down)

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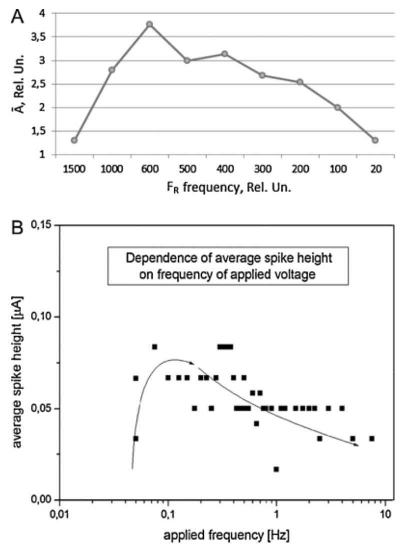

FIGURE 5 Dependence of the average amplitude of MP oscillations on the frequency of SF action (on top); Dependence of the average spike height on the frequency of applied voltage (down), here the curve is drown by the method of least squares.

5 Conclusion

The effect of current spikes in track devices is described. Different explanations of this effect are discussed. A phenomenological model for ion current spikes in track-containing foils is considered in more detail.

Experimental results show that ion current spikes in track-containing foils arise for different forms of tracks and different types of materials and that the current spike effect is determined significantly by the mean distance between tracks. A computer experiment with the developed model for current spikes showed that this model reflects the main features of the ion current spikes in track structures. It is shown that taking into account only one factor (the interaction of currents in the system of tracks) leads to the result of an emergence of current spikes. The occurrence of current spikes takes place in a wide range of the potential parameters.

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