UDC 539.2
DOI https://doi.org/10.32782/2450-8640.2025.1.9

Dietmar Fink,
Nuclear Physics Institute, Czech Academy of Sciences, Řež, Czech Republic
Universidad Autónoma Metropolitana-Iztapalapa, D.F., México
Scopus Author ID: 55439567000, e-mail: fink@xanum.uam.mx

Alina Vinkovskaya,
The State Institution «South Ukrainian National Pedagogical University named after K.D. Ushynsky», Ukraine
e-mail: alina.vinkovskaya@gmail.com

Taras Kavetskyy,
Drohobych Ivan Franko State Pedagogical University, Drohobych, Ukraine
Institute of Physics, Slovak Academy of Sciences, Slovak Republic
The State Institution «South Ukrainian National Pedagogical University named after K.D. Ushynsky», Ukraine
orcid.org/0000-0002-4782-1602, Scopus Author ID: 57220358576,
e-mail: kavetskyy@yahoo.com

Nataliia Hoivanovych,
Drohobych Ivan Franko State Pedagogical University, Ukraine
orcid.org/0000-0002-3442-0674, Scopus Author ID: 57203341250,
e-mail: natahoyvan@gmail.com

Yuliia Kukhazh,
Drohobych Ivan Franko State Pedagogical University, Ukraine
Scopus Author ID: 56507384300, e-mail: juljakhj@i.ua

Mariana Kravtsiv,
Drohobych Ivan Franko State Pedagogical University, Ukraine
Scopus Author ID: 56919697400, e-mail: marinettakr@gmail.com

Oles Matskiv,
Drohobych Ivan Franko State Pedagogical University, Ukraine
e-mail: omackiv@gmail.com

Ondrej Šauša,
Institute of Physics, Slovak Academy of Sciences, Slovak Republic
Department of Nuclear Chemistry, Comenius University, Slovak Republic
orcid.org/0000-0001-9958-5966, Scopus Author ID: 6602354298,
e-mail: ondrej.sausa@savba.sk

Arnold Kiv,
The State Institution «South Ukrainian National Pedagogical University named after K.D. Ushynsky», Ukraine
Ben-Gurion University of the Negev, Israel
orcid.org/0000-0002-0991-2343, Scopus Author ID: 6602488378,
e-mail: kiv.arnold20@gmail.com

MECHANISM OF OSCILLATIONS OF ION CURRENTS IN THE NANOTRACKS

Abstract. The paper contains the description of the features of ion current pulsations in track devices. The conditions under which the pulsations arise are discussed. We discuss different approaches that are used for explanation the effects of ion current pulsations. Ion current pulsations when passing through nanotracks play a dual role in practical applications. In biosensor systems, these effects significantly affect the parameters of devices. In other cases, these effects serve as the basis for the mechanisms of their new device operation.

Our studies, as well as the results of other authors, show that the mechanisms of ion current oscillations in nanotracks are associated with three reasons. The first reason is the collective interactions of currents in the nanotracks, the second is a certain defect structure of the internal walls of nanotracks, and the third is carbonization in the case of nanotracks in carbon materials. The article shows that the knowledge of the nature of ion current fluctuations in nanotracks allows modifying these effects to obtain the necessary parameters for electronic devices. These effects are especially important in the creation and improvement of modern biosensors.
Key words: ion currents, nanotracks, nanosensors, biosensors.

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 films (for example, polymer foils), the so-called latent ion tracks. The damage shows up primarily by the formation of radiochemical 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, for examples 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.

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 [1].

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

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. 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 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 the structures with etched tracks in the films of SiO2 on Si 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.

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].

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 phase-locked 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 it is in an 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 [15] 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 [16] 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 mediated by 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.

THE MODEL OF CURRENT SPIKES IN TRACK DEVICES

For ion current pulsations in track-containing foils the following features are established [17, 18]:

- 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 the applied voltage. Its decrease indicates the existence of a threshold frequency for spike emission.

The model described in [19] 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

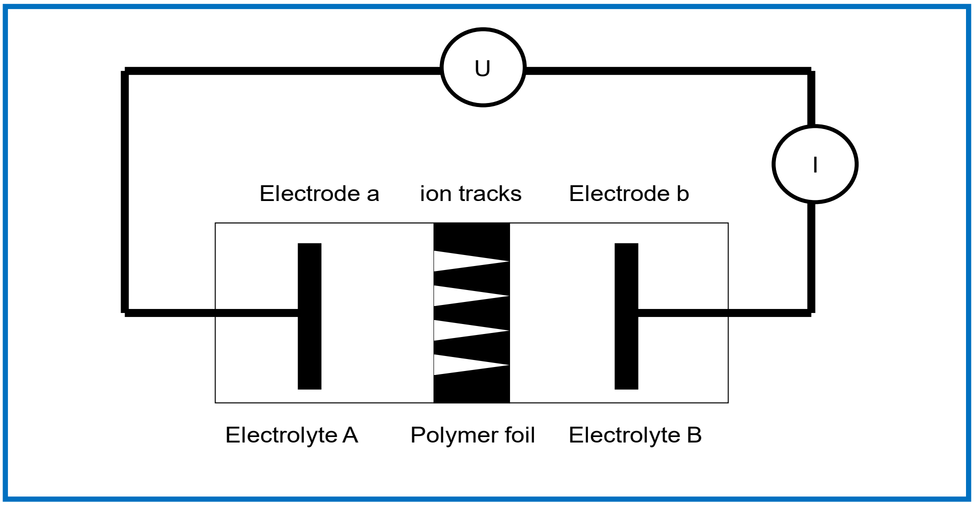

Fig. 1. Experimental setup to study current spike emission in ion track-containing foils embedded in electrolytes (current/voltage measurements)

Molecular Dynamics (MD) was modified so that a new MD approach allows describing also subthreshold radiation effects.

CONCLUSION

The effect of current spikes in track devices is discussed. Different explanations of this effect are considered. 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.

ACKNOWLEDGMENTS

This work was supported in part by the Ministry of Education and Science of Ukraine (projects Nos. 0122U000850, 0122U000874, 0125U001054, 0125U002005, and 0125U002033), National Research Foundation of Ukraine (project No. 2020.02/0100), Slovak Grant Agency VEGA (projects Nos. 2/0166/22 and 2/0131/25), and Slovak Research and Development Agency (project No. APVV-21-0335). T.K. and Y.K. acknowledge the SAIA for scholarships in the Institute of Physics of Slovak Academy of Sciences in the framework of the National Scholarship Programme of the Slovak Republic. This work has also received funding through the MSCA4Ukraine project (grant No. 1128327), which is funded by the European Union, and the EURIZON project (grant EU-3022), which is funded by the European Union (EURIZON H2020 project) under grant agreement No. 871072.

BIBLIOGRAPHY

1. Alfonta L., Bukelman O., Chandra A., Fahrner W. R, Fink D., Fuks D., Golovanov V., Hnatowicz V., Hoppe K., Kiv A., Klinkovich I., Landau M., Morante J.R., Tkachenko N.V., Vacik J., Valden M. Strategies towards advanced ion track-based biosensors. *Radiat. Eff. Defects Solids.* 2009. Vol. 164. P. 431–437.
2. Fink D., Gerardo Munoz H., Alfonta L., Mandabi Y., Dias J.F., de Souza C.T., Bacakova L.E., Vacik J., Hnatowicz V., Kiv A.E., Fuks D., Papaleo R.M. Status and Perspectives of Ion Track Electronics for Advanced Biosensing. Eds. Shunin, Y.N., & A.E. Kiv, A.E., Nanodevices and Nanomaterials for Ecological Security, NATO Science for Peace and Security, Series B: Physics and Biophysics, Springer, 2012, P. 269–279.
3. Fink D., Petrov A., Hoppe K., Fahrner W.R. Characterization of "TEMPOS" structures. *Proceedings of MRS Meeting* (Boston, 1-5 December 2003).
4. Powell M.R., Sullivan M., Vlassiouk I., Constantin D., Sudre O., Martens C.C., Eisenberg R.S., Siwy Z.S. Nanoprecipitation-assisted ion current oscillations. *Nat. Nanotech.* 2008. Vol. 3. P. 51–59.
5. Fink D., Cruz S., Munoz G., Kiv A. Current spikes in polymeric latent and funnel-type ion tracks. *Radiat. Eff. Defects Solids.* 2010. Vol. 166. P. 373–388.
6. Fink D., Munoz G.H., Vacik J., Alfonta L. Pulsed biosensing. *IEEE Sens. B.* 2011. Vol. 11. P. 1084–1089.
7. Fink D., Kiv A., Fuks D., Tabacnics M., de Rizutto M.A., Silva A.O.D., Chandra A. Irradiation induced pulsations of reverse biased metal oxide/silicon structures. *Appl. Phys. Lett.* 2007. Vol. 91. P. 083512–83520.
8. Cervera J., Schiedt B., Ramirez P. Poisson-Nernst-Planck model for ionic transport through synthetic conical nanopores. *Europhys. Lett.* 2005. Vol. 71. P. 35–41.
9. Cervera J., Schiedt B., Neumann R., Mafe S., Ramirez P. Ionic conduction, rectification and selectivity in single conical nanopores. *J. Chem. Phys.* 2006. Vol. 124. P. 104706–104710.
10. Cruz-Chu E.R., Aksimentiev A., Schulten K. Ionic current rectification through silica nanopores. *J. Phys. Chem. C.* 2009. Vol. 113. P. 1850–1862.
11. Cruz-Chu E.R., Ritz T., Siwy Z.S., Schulten K. Molecular control of ionic conduction in polymer nanopores. *Faraday Discuss.* 2009. Vol. 143. P. 47–62.
12. Siwy Z., Apel P., Dobrev D., Neumann R., Spoor R., Trautmann C., Voss K. Ion transport through asymmetric nanopores prepared by ion track etching. *Nucl. Instrum. Meth. B.* 2003. Vol. 208. P. 143–148.
13. Fink D., Vacik J., Garcia Arellano H., Muñoz G.H., Alfonta L., Fahrner W.R., Hoppe K., Kiv A. Biosensors with nuclear tracks and embedded membranes. *Key Eng. Mater.* 2014. Vol. 605. P. 83–86.
14. Fink D., Muñoz G.H., Vacik J., Alfonta L. Pulsed biosensing. *IEEE Sen. J.* 2011. Vol. 11(4). P. 1084–1087.
15. Fink D., Kiv A., Fuks D., Saad A., Vacik J., Hnatowicz V., Chandra A. Conducting swift heavy ion track networks. *Radiat. Eff. Defects Solids.* 2010. Vol. 165. P. 227–244.
16. Fink D., Hnatowicz V., Apel P.Y. Transport Processes in Tracks. Transport Processes in Ion Irradiated Polymers. Ed. Fink, D. *Springer Series in Materials Science*, 2004. Vol. 65. P. 128–130.
17. Fink D., Kiv A., Fuks D., Munoz G.H., Cruz S.A., Fahrner W.R., Hoppe K. Collective interaction in ion track electronics. *Computer Modelling and New Technologies.* 2011. Vol. 15. P. 7–18.
18. Fink D., Cruz S., Munoz G.H., Kiv A. Current spikes in polymeric latent and funnel-type ion tracks. *Radiat. Eff. Defects Solids.* 2011. Vol.166. P. 373–388.
19. Mykytenko N., Fink D., Kiv A. Computer modeling of ion current pulsations in track-containing foils. *J. Comput. Sci.* 2015. Vol. 6. P. 34–39.

REFERENCES

1. Alfonta, L., Bukelman, O., Chandra, A., Fahrner, W. R, Fink, D., Fuks, D., Golovanov, V., Hnatowicz, V., Hoppe, K., Kiv, A., Klinkovich, I., Landau, M., Morante, J.R., Tkachenko, N.V., Vacik, J., & Valden, M. (2009). Strategies towards advanced ion track-based biosensors. *Radiat. Eff. Defects Solids.* Vol. 164. P. 431–437.
2. Fink, D., Gerardo Munoz, H., Alfonta, L., Mandabi, Y., Dias, J.F., de Souza, C.T., Bacakova, L.E., Vacik, J., Hnatowicz, V., Kiv, A.E., Fuks, D., & Papaleo, R.M. (2012). Status and Perspectives of Ion Track Electronics for Advanced Biosensing. Eds. Shunin, Y.N., & A.E. Kiv, A.E., Nanodevices and Nanomaterials for Ecological Security, NATO Science for Peace and Security, Series B: Physics and Biophysics, Springer, P. 269–279.
3. Fink, D., Petrov, A., Hoppe, K., & Fahrner, W.R. (2003). Characterization of "TEMPOS" structures. *Proceedings of MRS Meeting* (Boston, 1-5 December 2003).
4. Powell, M.R., Sullivan, M., Vlassiouk, I., Constantin, D., Sudre, O., Martens, C.C., Eisenberg, R.S., & Siwy, Z.S. (2008). Nanoprecipitation-assisted ion current oscillations. *Nat. Nanotech.* Vol. 3. P. 51–59.
5. Fink, D., Cruz, S., Munoz, G., & Kiv, A. (2010). Current spikes in polymeric latent and funnel-type ion tracks. *Radiat. Eff. Defects Solids.* Vol. 166. P. 373–388.
6. Fink, D., Munoz, G.H., Vacik, J., & Alfonta, L. (2011). Pulsed biosensing. *IEEE Sens. B.* Vol. 11. P. 1084–1089.
7. Fink, D., Kiv, A., Fuks, D., Tabacnics, M., de Rizutto, M.A., Silva, A.O.D., & Chandra, A. (2007). Irradiation induced pulsations of reverse biased metal oxide/silicon structures. *Appl. Phys. Lett.* Vol. 91. P. 083512–83520.
8. Cervera, J., Schiedt, B., & Ramirez, P. (2005). Poisson-Nernst-Planck model for ionic transport through synthetic conical nanopores. *Europhys. Lett.* Vol. 71. P. 35–41.
9. Cervera, J., Schiedt, B., Neumann, R., Mafe, S., & Ramirez, P. (2006). Ionic conduction, rectification and selectivity in single conical nanopores. *J. Chem. Phys.* Vol. 124. P. 104706–104710.
10. Cruz-Chu, E.R., Aksimentiev, A., & Schulten, K. (2009). Ionic current rectification through silica nanopores. *J. Phys. Chem. C.* Vol. 113. P. 1850–1862.
11. Cruz-Chu, E.R., Ritz, T., Siwy, Z.S., & Schulten, K. (2009). Molecular control of ionic conduction in polymer nanopores. *Faraday Discuss.* Vol. 143. P. 47–62.
12. Siwy, Z., Apel, P., Dobrev, D., Neumann, R., Spoor, R., Trautmann, C., & Voss, K. (2003). Ion transport through asymmetric nanopores prepared by ion track etching. *Nucl. Instrum. Meth. B.* Vol. 208. P. 143–148.
13. Fink, D., Vacik, J., Garcia Arellano, H., Muñoz, G.H., Alfonta, L., Fahrner, W.R., Hoppe, K., & Kiv, A. (2014). Biosensors with nuclear tracks and embedded membranes. *Key Eng. Mater.* Vol. 605. P. 83–86.
14. Fink, D., Muñoz, G.H., Vacik, J., & Alfonta, L. (2011). Pulsed biosensing. *IEEE Sen. J.* Vol. 11(4). P. 1084–1087.
15. Fink, D., Kiv, A., Fuks, D., Saad, A., Vacik, J., Hnatowicz, V., & Chandra, A. (2010). Conducting swift heavy ion track networks. *Radiat. Eff. Defects Solids.* Vol. 165. P. 227–244.
16. Fink, D., Hnatowicz, V., & Apel, P.Y. (2004). Transport Processes in Tracks. Transport Processes in Ion Irradiated Polymers. Ed. Fink, D. Springer Series in Materials Science, Vol. 65. P. 128–130, and references therein.
17. Fink, D., Kiv, A., Fuks, D., Munoz, G.H., Cruz, S.A., Fahrner, W.R., & Hoppe, K. (2011). Collective interaction in ion track electronics. *Computer Modelling and New Technologies.* Vol. 15. P. 7–18.
18. Fink, D., Cruz, S., Munoz, G.H., & Kiv, A. (2011). Current spikes in polymeric latent and funnel-type ion tracks. *Radiat. Eff. Defects Solids.* Vol.166. P. 373–388.

19. Mykytenko, N., Fink, D., & Kiv, A. (2015). Computer modeling of ion current pulsations in track-containing foils. *J. Comput. Sci.* Vol. 6. P. 34–39.

АНОТАЦІЯ

МЕХАНІЗМ КОЛИВАНЬ ІОННИХ СТРУМІВ У НАНОТРЕКАХ

У статті наведено опис особливостей пульсацій іонного струму у трекових пристроях. Обговорюються умови, за яких виникають пульсації. Ми розглядаємо різні підходи, що використовуються для пояснення ефектів пульсацій іонного струму. Пульсації іонного струму при проходженні через нанотреки відіграють подвійну роль у практичному застосуванні. У біосенсорних системах ці ефекти суттєво впливають на параметри пристроїв. В інших випадках ці ефекти слугують основою для механізмів їхньої роботи в нових пристроях.

Наші дослідження, а також результати інших авторів, показують, що механізми коливань іонного струму у нанотреках пов'язані з трьома причинами. Перша причина – колективна взаємодія струмів у нанотреках, друга – певна дефектна структура внутрішніх стінок нанотреків, а третя – карбонізація у випадку нанотреків у вуглецевих матеріалах. У статті показано, що знання природи коливань іонного струму у нанотреках дозволяє модифікувати ці ефекти для отримання необхідних параметрів для електронних пристроїв. Ці ефекти особливо важливі при створенні та вдосконаленні сучасних біосенсорів.

Ключові слова: іонні струми, нанотреки, наносенсори, біосенсори.