On the use of carbon nanotubes in prototyping the high energy density Li-ion batteries

M.S. Yerdauletov[1,2,3], *M.V. Avdeev* [2,1], *O.I. Ivankov* [2], *F.S. Napolskiy* [1], *V.A. Krivchenko*[1]

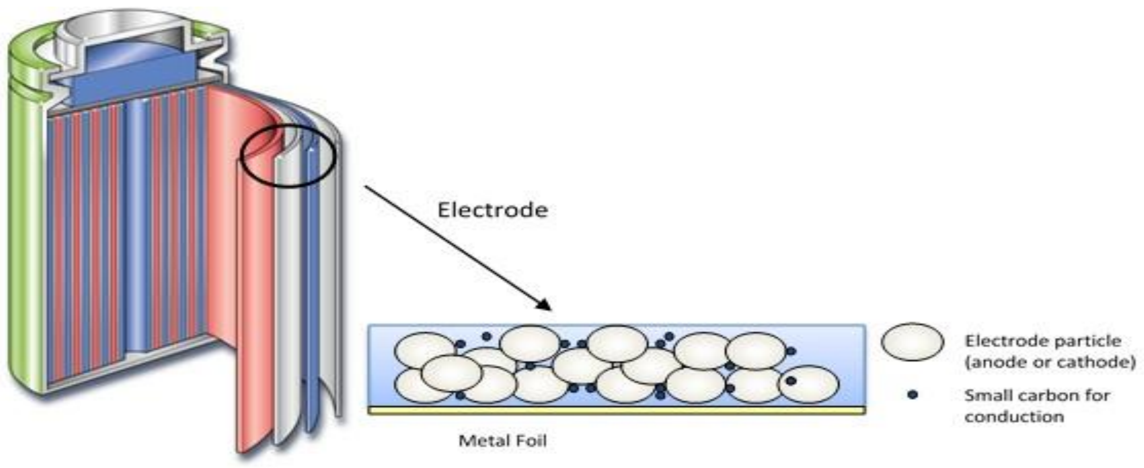
[1] **Dubna State University, Dubna, Moscow Region, Russia**
[2] **Frank Laboratory of Neutron Physics, Joint Institute for Nuclear Research, Dubna, Moscow oblast, 141980 Russia**
[3] **L.N.Gumilyov Eurasian National University, 010008, Astana, Kazakhstan**

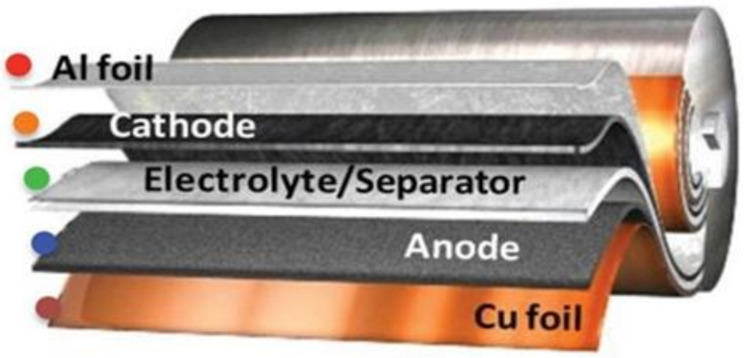
Aim of the work: to study the effect of the microstructure of positive electrodes and improving the specific parameters of lithium-ion batteries

Tasks solved within the framework of the work

1) Study of the influence of the microstructure of the positive electrodes of chemical current sources on their functional characteristics
2) Development of the technology for forming electrode composite materials with different microstructure using synthesized conductive additives and various polymer binders.
3) Study of the microstructure of lithium-ion batteries by small-angle neutron scattering.
4) Use neutron / X-ray radiography and tomography to study the macrostructure of lithium ion batteries.
5) Investigation of electrochemical characteristics of positive / negative electrodes with different compositions and different microstructure. Determination of resistance to cycling at different charging / discharging powers.
6) Study of the influence of the type of electrolyte on the functional characteristics of the electrodes.

The relevance of research

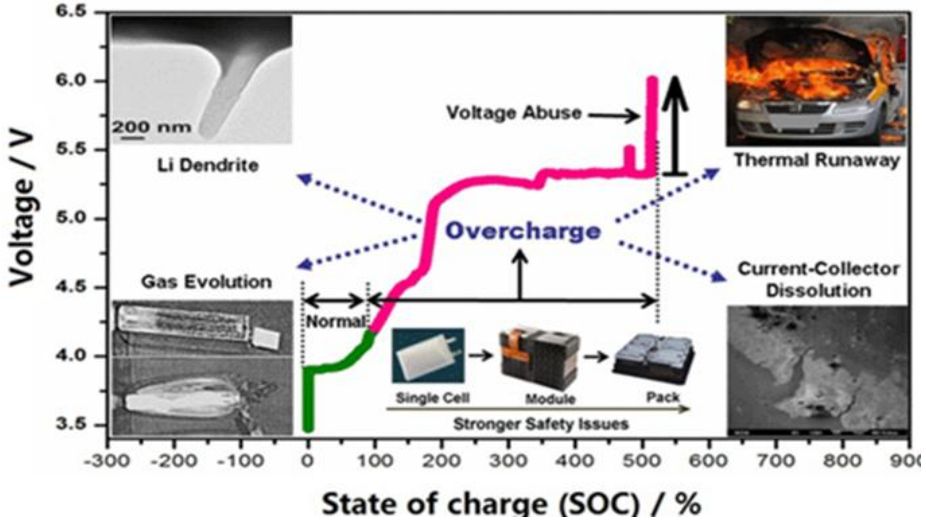




1) An urgent task is to improve the existing, search and develop new energy-intensive and efficient rechargeable chemical current sources.

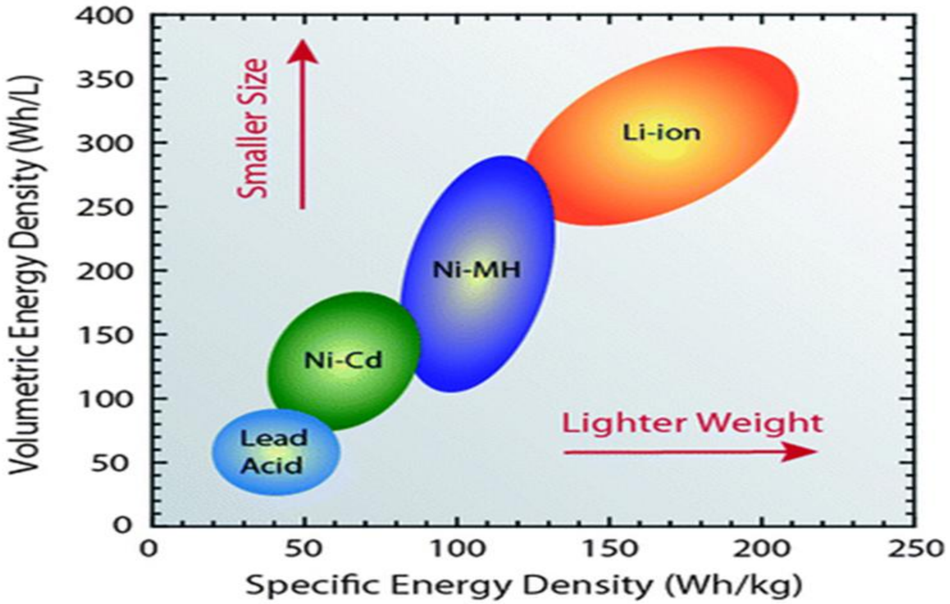
2) The creation of rechargeable chemical current sources with increased specific characteristics is usually associated with the development of new electrode materials that provide a higher reversible specific capacity, a large potential difference and operate at higher discharge / charge currents.

Practical significance

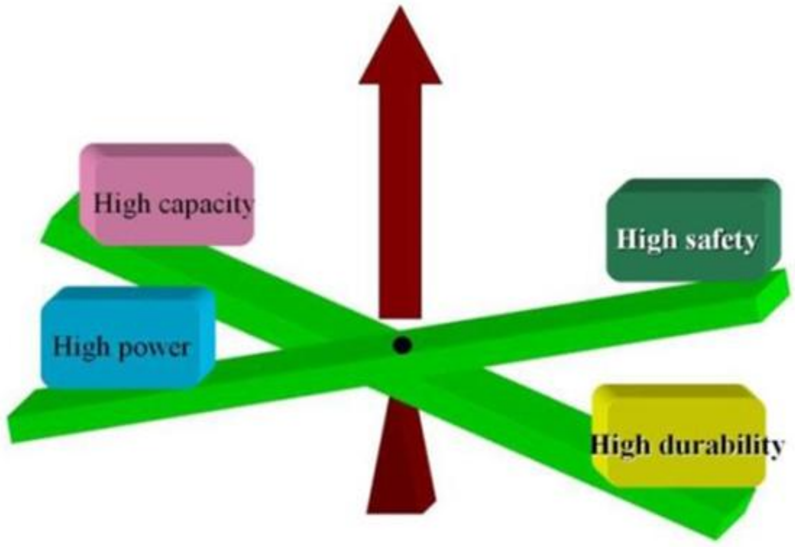


Jianwu Wen, Yan Yu, and Chunhua Chen
Mater. Express 2, 197-212 (2012)

1) The industrial production of modern batteries (primarily lithium-ion), however, is faced with an equally important problem of controlling the microstructure of the applied electrode coatings, without optimization of which there can be a significant loss in the specific energy and / or power of devices.



https://www.epectec.com/batteries



Scientific novelty

1) Fundamentally new scientific and technical solutions for the formation of efficient cathodes will be based on the use of nanostructured conductive additives based on graphene derivatives and modified carbon nanotubes

2) The use of new cathode materials on the example of V2O5

3) The use of polymeric binders with different structures. (gel polymers)

4) Optimization of the microstructure of cathodes for lithium-sulfur and sodium-ion batteries, respectively. These systems can be considered as the next step in the evolution of electrochemical energy storage.

P.G.Bruce et al. // *Nature Materials 11*(1), 2011, 19–29

Electrode preparation

1. Polymer binder Solef5130 dissolving in NMP
2. Conductive additive C45, CNT
3. Active material LiFePO4
   - Mixing on High-shear overhead mixer for 24 hours
4. Electrodes were prepared by dr.blade casting onto 20μm Al-foil
5. Electrochemical tests were carried out in 2032-type coin cells

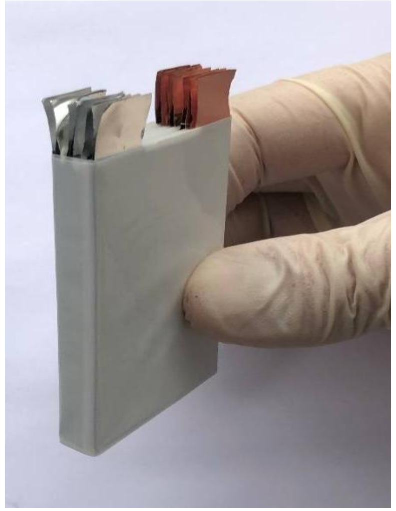


Fig 1. Electrode stack

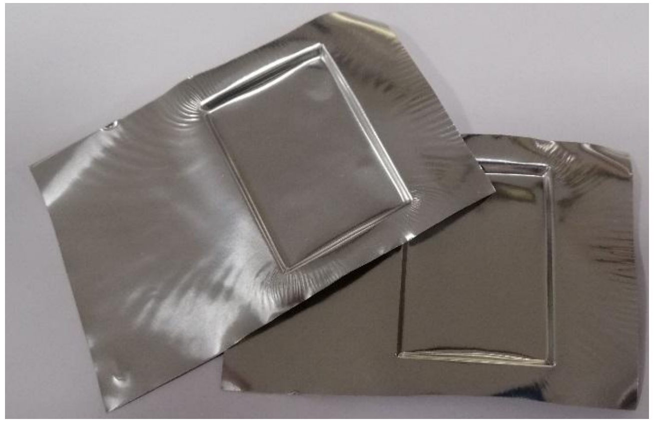


Fig 2. pouch cell case parts

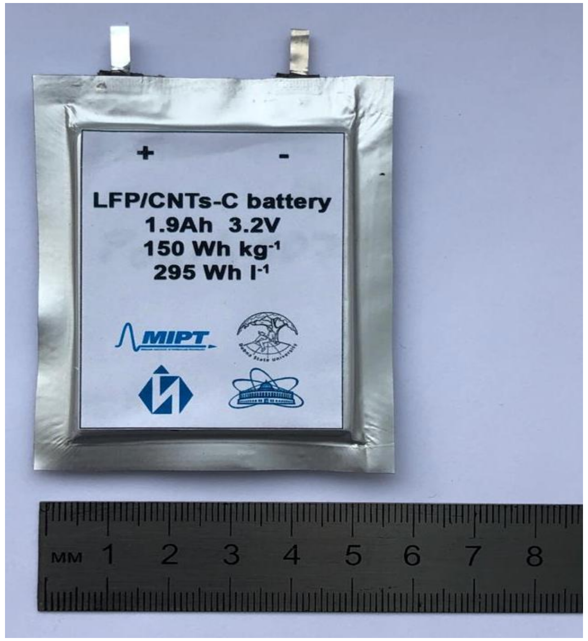


Fig 3. pouch cell after the formation and re-sealing

Main Results

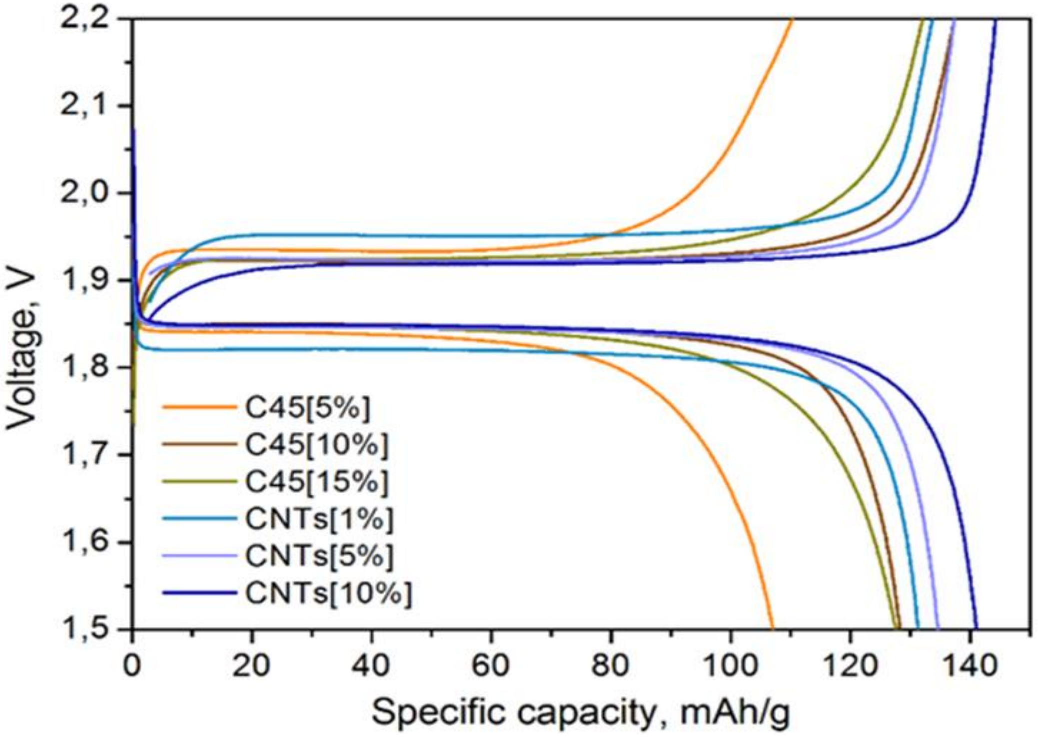


Fig 4. Charge and discharge current is 50mA per g LFP. Specific capacitance calculated per gram of LFP in the cathode structure

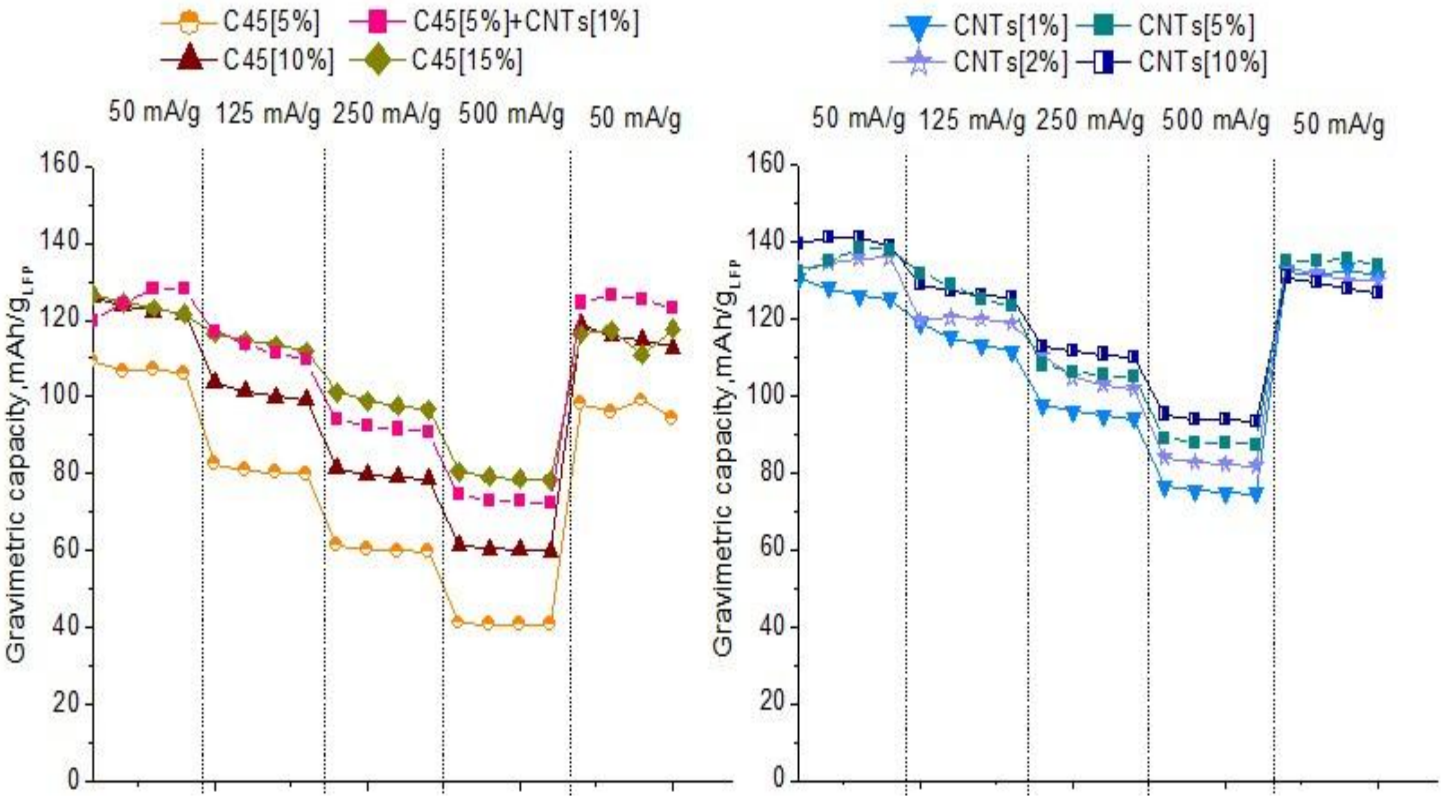


Fig 5. Dependences of the gravimetric capacity on the number of cycles at different discharge currents for cathodes with different mass content of C45 and CNTs

Main Results

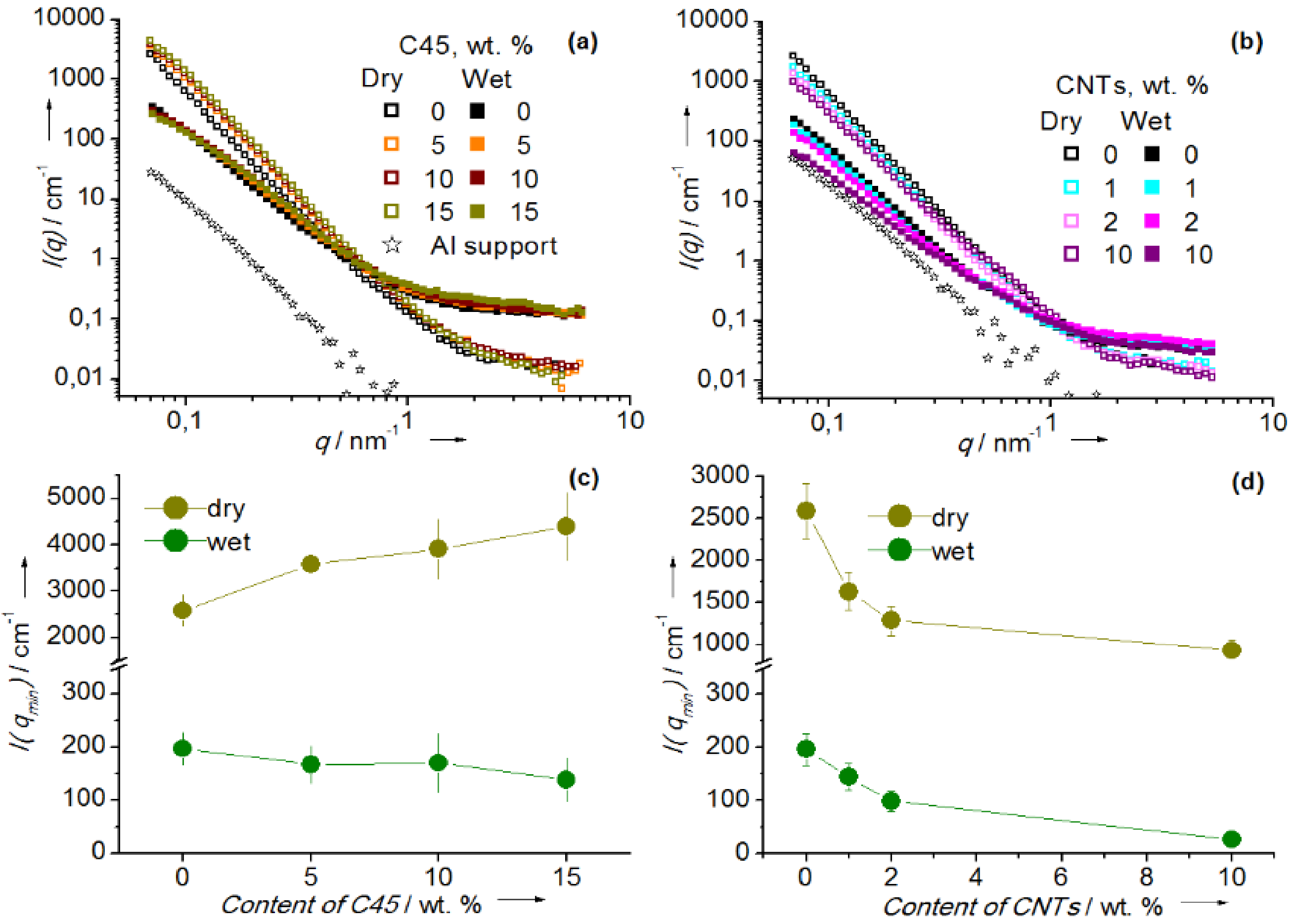
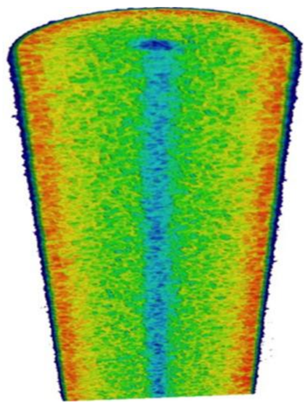


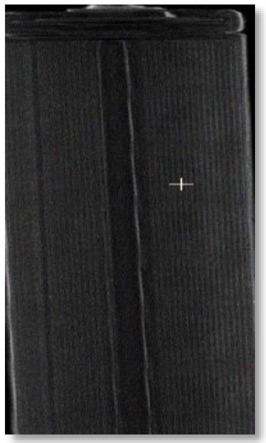
Fig 6. Small-angle neutron scattering curves measured (YuMO setup, IBR-2) for dry and wet (d-electrolyte) electrodes with different mass content of C45 (a) and CNTs (b). Asterix – scattering from Al substrate. Data are calibrated on the thickness of the electrode layer. 0% of conductive additive corresponds to scattering from "LFP matrix". Dependence of forward scattering intensity (I at minimal q) on content of C45 (c) and CNTs (d): data are corrected on the scattering from Al substrate.

Future Plans

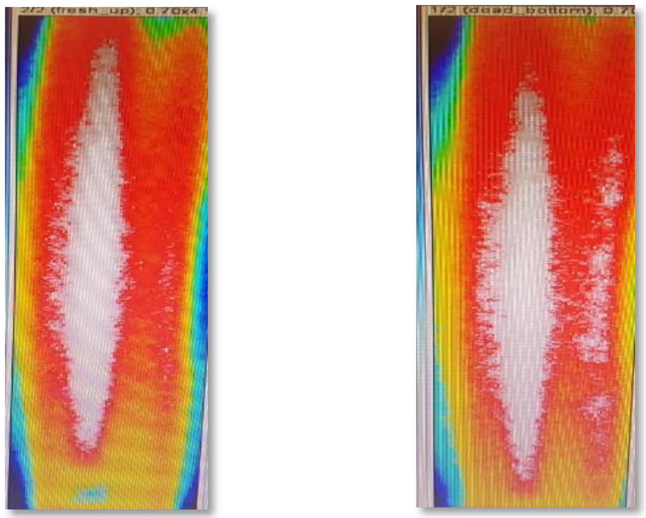
Neutron picture



3D neutron tomography



X-ray radiography



Neutron picture of Coin cell with 0% and 100% capacity

Conclusion

1) The electrodes with 1% of the CNTs demonstrate a higher specific capacity compared with electrodes containing 5-15% of carbon black

2) SANS measurements revealed that the CNT network embedded in the electrode layer provides its greater wettability by an electrolyte compared to carbon black used as conductive additive. This results in better electrode C-rate performance

3) All the carbon structures used in the experiments provides kinetic transport of lithium ions into the crystals of active material

4) The technological aspects of fabrication of high areal capacity LFP-based electrodes using the carbon nanotubes as conductive additives were considered

The main scientific achievements of the team

The results of the research activities of the team members are reflected in a series of publications in top-rated international publications.

List of major publications over the past 5 years:

1. **«On the Use of Carbon Nanotubes in Prototyping the High Energy Density Li-ion Batteries», Filipp Napolskiy, et.al, 2020, Energy Technology, DOI: 10.1002/ente.202000146 (Impact factor – 3.4).**
2. **«In Situ XPS Studies of Solid Electrolyte Electroreduction Through Graphene Electrode», Inozemtseva Alina I., et.al, 2020, Journal of the Electrochemical Society, DOI: 10.1149/1945-7111/aba370 (Impact factor – 3.66).**
3. **«Crumpled Nitrogen-doped Graphene Wrapped Phosphorus Composite as a Promising Anode for Lithium-Ion Batteries», Jiao X., et.al, 2019, ACS applied materials & interfaces, DOI: 10.1021/acsami.9b08915 (Impact factor – 8.7).**
4. **«Direct patterning of reduced graphene oxide/graphene oxide memristive heterostructures by electron-beam irradiation», Kapitanova O.O., et.al, 2019, Journal of Materials Science and Technology, DOI: 10.1016/j.jmst.2019.07.042 (Impact factor – 5).**
5. **«Small-angle neutron scattering studies of pore filling in carbon electrodes: mechanisms limiting lithium–air battery capacity», Tatiana K. Zakharchenko, et.al, 2019, Nanoscale, DOI: 10.1039/C9NR00190E (Impact factor – 6.9).**
6. **«Extended Limits of Reversible Electrochemical Lithiation of Crystalline V2O5», Daniil M. Itkis, et.al, 2019, ChemElectroChem, https://doi.org/10.1002/celc.201801638 (Impact factor –4.15).**
7. **«Modified carbon nanotubes for water-based cathode slurries for lithium–sulfur batteries», Olesya O. Kapitanova, et.al, 2019, J.Mater.Research, DOI: 10.1557/jmr.201 (Impact factor -2.5).**
8. **«Gaining cycling stability of Si and Ge negative Li-ion high areal capacity electrodes by using carbon nanowall scaffolds», Mironovich K.V., et.al, 2017, Journal of Materials Chemistry A, DOI: 10.1039/c7ta03509h (Impact factor – 11.3)**
9. **«Self-assembled nanoparticle patterns on carbon nanowall surfaces», Suetin N.V., et.al, 2016, Physical Chemistry Chemical Physics, DOI: 10.1039/c6cp01638c (Impact factor –3.6)**
10. **«Electrochemical Modification of Electrodes Based on Highly Oriented Carbon Nanowalls», Krivenko A.G., et.al, 2015, Russian Journal of Electrochemistry, DOI: 10.1134/S1023193515100079 (Impact factor –2.17)**
11. **«Enhancement of the Carbon Nanowall Film Capacitance. Electron Transfer Kinetics on Functionalized Surfaces», Natal'ya Semenovna Komarova, et.al, 2015, Langmuir, DOI: 10.1021/acs.langmuir.5b00391 (Impact factor –3.7)**
12. **«Oxygen Reduction by Lithiated Graphene and Graphene-Based Materials», Kataev Elmar Yu, et.al, 2015, ACS Nano, DOI: 10.1021/nn5052103 (Impact factor –14.5)**

Grants and awards :

1. Diploma of the II degree of an international scientific conference «Science and Education-2017» Astana, 2017
2. I Award in the nomination "Scientific and technical applied work" OIAI for young scientists and specialists for 2017, Dubna, Russia
3. Diploma of the II degree of the international conference Falling Walls LAB Dubna, Dubna, Russia, 2018
4. II Prize in the nomination "Scientific and technical applied work of JINR for young scientists in 2018"
5. The best speaker at the VII conference of young scientists and specialists "Alushta-2018" Alushta, Crimea
6. Diploma of the II degree of the international conference Falling Walls LAB Dubna, Dubna, Russia,2019
7. Received a grant from AYSS at JINR for young scientists and specialists for 2020 for young researchers.

Scientific and technological cooperation

The successful conduct of our research activities is based on close cooperation with leading international research centers and manufacturing enterprises.

Joint Institute for Nuclear Research

Lomonosov Moscow State University

SCIENTIFIC RESEARCH AND DESIGN AND TECHNOLOGICAL INSTITUTE OF ELECTROCARAL PRODUCTS. JSC "NIIEI"

MIPT
MOSCOW INSTITUTE OF PHYSICS AND TECHNOLOGY

Institute of Nuclear Physics, Almaty, Kazakhstan

西安交通大學
XI'AN JIAOTONG UNIVERSITY

Thank you for your attention