

## Chemical Systems for Life Science

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**Abstract.** The use of dynamic, adaptive materials with feedback control is a tendency of the past decade. Life sciences and medicine require materials with the controlled and responsive assembly of various components on the scales from molecular to macroscopic and even robotics. The main idea of this review is the use of synthetic systems as regulatory networks that facilitate the integration of chemical and biological materials. The synthetic systems, which are inspired by biochemical regulatory networks, help synthetic material to adapt to environment and to interact with living matter cooperatively. The first step in realizing this concept is designing simple model systems. The simplicity means that the system should contain a minimal number of components but should be robust and sustainable to perform the required functions through logic operations and feedback loops. Here we suggest specific examples of robust systems for the selected functionality: compartmentalized signaling cascades, computation with light-induced chemical gradients and advanced biomimetic mixed organic-inorganic materials, and self-regulation in chemical-biological systems. The main challenges for the given examples are discussed, and future prospects of logic operation with chemical systems are provided.

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## REFERENCES

- [1] A. Rosenfeld, P. A. Levkin, *High-Throughput Combinatorial Synthesis of Stimuli-Responsive Materials*, Advanced Biosystems, 2019, vol. 3, art. 1800293.
- [2] V. A. Bolaños Quiñones, H. Zhu, A. A. Solovev, Y. Mei, and D. H. Gracias, *Origami Biosystems: 3D Assembly Methods for Biomedical Applications*, Advanced Biosystems, vol.2, art. 1800230.
- [3] N. V. Ryzhkov, D. V. Andreeva and E. V. Skorb, *Coupling pH-Regulated Multilayers with Inorganic Surfaces for Bionic Devices and Infochemistry*, Langmuir, 2019, vol. 35, no. 26, pp. 8543-8556.
- [4] S. Li, H. Qin, T. Zhang, H. P. Cong and S. H. Yu, *Highly Tough Bioinspired Ternary Hydrogels Synergistically Reinforced by Graphene/Xonotlite Network*, 2018, vol. 14, no. 22, art., e1800673.
- [5] S. Yoshida, H. Ejima and N. Yoshie, *Tough Elastomers with Superior Self-Recoverability Induced by Bioinspired Multiphase Design*, Advanced Functional Materials, 2017, vol. 27, no. 30, art. 1701670.
- [6] C. J. Higginson, K. G. Malollari, Y. Xu, A. V. Kelleghan, N. G. Ricapito and P. B. Messersmith, *Bioinspired Design Provides High-Strength Benzoxazine Structural Adhesives*, Angewandte Chemie International Edition, vol. 58, no. 35, pp. 12271-12279.
- [7] S. W. Thomas, 3rd, R. C. Chiechi, C. N. LaFratta, M. R. Webb, A. Lee, B. J. Wiley, M. R. Zakin, D. R. Walt and G. M. Whitesides, *Infochemistry and infofuses for the chemical storage and transmission of coded information*, Proceedings of the National Academy of Sciences of the United States of America, 2009, vol 106, no. 23, pp. 9147-9150.
- [8] C. Kim, S. W. Thomas, 3rd and G. M. Whitesides, *Long-duration transmission of information with infofuses*, Angewandte Chemie (International ed. in English), 2010, vol. 49 no. 27, pp. 4571-4575.
- [9] D. B. Lawrence, T. Cai, Z. Hu, M. Marquez and A. D. Dinsmore, *Temperature-Responsive Semipermeable Capsules Composed of Colloidal Microgel Spheres*, Langmuir, 2007, vol. 23, no. 2, pp. 395-398.
- [10] S. C. Basak, G. Restrepo and J. L. Villaveces, *Adv. Math. Chem. Appl.*, Vol. 2 (Elsevier Inc., 2016).
- [11] M. L. Peach, A. V. Zakharov, L. Guasch and M. C. Nicklaus, *Chemoinformatics*, Comprehensive Biomedical Physics, 2014, vol. 6, pp. 123-156.
- [12] E. Szymanska, J. Gerretzen, J. Engel, B. Geurts, L. Blanchet and L. M. C. Buydens, *Chemometrics for ion mobility spectrometry data: recent advances and future prospects*, The Analyst, 2016, vol. 141, no. 20, pp. 5689–5708.
- [13] M. S. Cohen, C. Zhang, K. M. Shokat, J. Taunton, *Structural bioinformatics-based design of selective, irreversible kinase inhibitors*, Science, 2005, vol. 308, no. 5726, pp.1318-13121.
- [14] A. Armada-Moreira, B. Thingholm, K. Andreassen, A. M. Sebastião, S. H. Vaz and B. Städler, *On the Assembly of Microreactors with Parallel Enzymatic Pathways*, Advanced Biosystems, 2018, vol. 2, no. 5, pp. 1700244.
- [15] P. L. H. Pham, S. A. Rooholghodos, J. S. Choy and X. L. Luo, *Constructing synthetic ecosystems with biopolymer fluitrodes*, Adv. Biosyst., 2018, vol. 2, no. 3, art. 1700180.
- [16] B. D. Shoener, S. M. Schramm, F. Beline, O. Bernard, C. Martinez, B. G. Plosz, S. Snowling, J. P. Steyer, B. Valverde-Perez, D. Wagner and J. S. Guest, *Microalgae and cyanobacteria modeling in water resource recovery facilities: A critical review*, Water. Res. X, 2019, vol. 2, art. 100024.
- [17] H. Lopes and I. Rocha, *Genome-scale modeling of yeast: chronology, applications and critical perspectives*, FEMS Yeast Research, 2017, vol. 17, no. 5.
- [18] T. Steinbrecher and G. Leubner-Metzger, *Tissue and cellular mechanics of seeds*, Curr Opin Genet Dev., 2018, vol. 51, pp. 1-10.
- [19] K. Le Vay, L. I. Weise, K. Libicher, J. Mascarenhas and H. Mutschler, *Templated Self-Replication in Biomimetic Systems*. Advanced Biosystems, 2019, art. 1800313.
- [20] C. Wolfer, M. Mangold and R. J. Flassig, *Towards Design of Self-Organizing Biomimetic Systems*, Advanced Biosystems, 2019, vol. 3, no. 6. art. e1800320.
- [21] M. Kunzle, M. Lach, T. Beck, *Multi-Component Self-Assembly of Proteins and Inorganic Particles: from Discrete Structures to Biomimetic Materials*, Israel Journal of Chemistry, 2019, vol. 50, no. 10, pp. 906-912.

- [22] R. Roy, "Biomimetic" materials: A potential distortion of materials policies, *Advanced Materials*, 1991, vol. 3, no. 9, pp. 448–451.
- [23] B. A. Blount, T. Weenink and T. Ellis, *Construction of synthetic regulatory networks in yeast*, *FEBS Letters*, 2012, vol.586, no. 15, pp. 2112–2121.
- [24] P. L. Foley and M. L. Shuler, *Considerations for the design and construction of a synthetic platform cell for biotechnological applications*, *Biotechnol Bioeng*, 2010, vol. 105, no. 1, pp. 26-36.
- [25] V. Noireaux and A. Libchaber, *A vesicle bioreactor as a step toward an artificial cell assembly*, *Proceedings of the National Academy of Sciences*, 2004, vol. 101, no. 51, pp. 17669–17674.
- [26] V. Noireaux, Y. T. Maeda and A. Libchaber, *Development of an artificial cell, from self-organization to computation and self-reproduction*, *Proceedings of the National Academy of Sciences*, 2011, vol.108, no. 9, pp. 3473–3480.
- [27] S. Rasmussen and P. F. Stadler, *Protocells* (MIT Press, Massachusetts, 2008).
- [28] J. W. Szostak, *The Narrow Road to the Deep Past: In Search of the Chemistry of the Origin of Life*, Angewandte Chemie International Edition, 2017, vol. 56. no. 37, pp. 11037–11043.
- [29] C. Xu, S. Hu and X. Chen, *Artificial cells: from basic science to applications*, *Materials Today*, 2016, vol. 19, no. 9, pp. 516–532.
- [30] J. Y. Trossat and P. Carbonell, *Synthetic biology for pharmaceutical drug discovery*, *Drug Des Devel Ther.*, 2015, vol. 9, pp. 6285-6302.
- [31] E. V. Skorb, H. Möhwald, *25th Anniversary Article: Dynamic Interfaces for Responsive Encapsulation Systems*, *Advanced Materials*, 2013, vol. 25, no. 36, pp 5029–5043.
- [32] E. V. Skorb and H. Möhwald, "Smart" Surface Capsules for Delivery Devices, *Advanced Materials Interfaces*, 2014, vol. 1, no. 6, art. 1400237.
- [33] E. V. Skorb, H. Möhwald and D. V. Andreeva, *How Can One Controllably Use of Natural ΔpH in Polyelectrolyte Multilayers?*, *Advanced Materials Interfaces*, 2017, vol. 4, no. 1, art. 1600282.
- [34] Y. Altay, S. Cao, H. Che, L. K. E. A. Abdelmohsen and J. C. M. van Hest, *Adaptive Polymeric Assemblies for Applications in Biomimicry and Nanomedicine*, *Biomacromolecules*, 2019, vol. 20, no. 11, pp. 4053-406.
- [35] H. Che, J. C. M. van Hest, *Adaptive polymersome nanoreactors*, *ChemNanoMat.*, vol5, no. 9, pp.1092-1109.
- [36] E. V. Skorb, D. Fix, D. V. Andreeva, H. Mohwald and D. G. Shchukin, *Mesoporous SiO<sub>2</sub> Containers for Corrosion Protection*, *Adv. Funct. Mater.*, vol. 19, no. 15, pp. 2373-2379.
- [37] D. V. Andreeva, D. V. Sviridov, A. Masic, H. Möhwald and E. V. Skorb, *Nanoengineered Metal Surface Capsules: Construction of a Metal-Protection System*, *Small*, 2012, vol. 8, no. 6, pp. 820-825.
- [38] D. V. Andreeva, D. V. Sviridov, A. Masic, H. Möhwald and E. V. Skorb, *Metal Capsules: Nanoengineered Metal Surface Capsules: Construction of a Metal-Protection System*, *Small* , 2012, vol. 8, no. 6, pp. 819.
- [39] N. Srinivas, J. Parkin, G. Seelig, E. Winfree and D. Soloveichik, *Enzyme-free nucleic acid dynamical systems*, *Science*, 2017, vol. 358, no. 6369, art. eaal2052.
- [40] W. Hordijk, M. Steel, S. Kauffman, *The Structure of Autocatalytic Sets: Evolvability, Enablement, and Emergence*. *Acta Biotheoretica*, 2012, vol. 60, pp. 379-392.
- [41] N. Srinivas, J. Parkin, G. Seelig, E. Winfree and D. Soloveichik, *Enzyme-free nucleic acid dynamical systems*, *Science*, 2017, vol. 358, no. 6369, art. eaa2052.
- [42] S. N. Semenov, A. J. Markvoort, W. B. Gevers, A. Piruska, T. F. de Greef and W. T. Huck, *Ultrasensitivity by molecular titration in spatially propagating enzymatic reactions*, *Biophysical Journal*, 2013, vol. 105, no. 4, pp. 1057-1066.
- [43] K. Pardee, A. A. Green, T. Ferrante, D. E. Cameron, A. DaleyKeyser, P. Yin and J. J. Collins, *Paper-based synthetic gene networks*, *Cell*, 2014, vol. 159, no. 4, pp. 940-54.
- [44] K. Montagne, R. Plasson, Y. Sakai, T. Fujii and Y. Rondelez, *Programming an in vitro DNA oscillator using a molecular networking strategy*, *Mol Syst Biol*, 2011,vol. 7, art. 466.
- [45] M. Weitz, J. Kim, K. Kapsner, E. Winfree, E. Franco and F. C. Simmel, *Diversity in the dynamical behaviour of a compartmentalized programmable biochemical oscillator*, *Nat Chem.*, 2014, vol. 6, no. 4, pp. 295-302.
- [46] D. Woods, D. Doty, C. Myhrvold, J. Hui, F. Zhou, P. Yin and E. Winfree, *Diverse and robust molecular algorithms using reprogrammable DNA self-assembly*, *Nature*, 2019, vol. 567, no. 7748, pp. 366-372.

- [47] S. N. Semenov, L. J. Kraft, A. Ainla, M. Zhao, M. Baghbanzadeh, V. E. Campbell, K. Kang, J. M. Fox and G. M. Whitesides, *Autocatalytic, bistable, oscillatory networks of biologically relevant organic reactions*, Nature, 2016, vol. 537, no. 7622, pp. 656–660.
- [48] K. Hasatani, M. Leocmach, A. J. Genot, A. Estévez-Torres, T. Fujii and Y. Rondelez, . *High-throughput and long-term observation of compartmentalized biochemical oscillators*, Chemical Communications, 2013, vol. 49, no. 73, pp. 8090-8092.
- [49] O. Kreft, M. Prevot, H. Mohwald and G. B. Sukhorukov, *Shell-in-shell microcapsules: a novel tool for integrated, spatially confined enzymatic reactions*, Angew Chem Int Ed Engl, 2007, vol. 46, np. 29, pp 5605-5608.
- [50] A. X. Lu, H. Oh, J. L. Terrell, W. E. Bentley and S. R. Raghavan, *A new design for an artificial cell: polymer microcapsules with addressable inner compartments that can harbor biomolecules, colloids or microbial species*, Chem. Sci., 2017, vol. 8, pp. 6893-6903.
- [51] F. He, W. Wang, X. H. He, X. L. Yang, M. Li, R. Xie, X. J. Ju, Z. Liu, L. Y. Chu, *Controllable Multicompartmental Capsules with Distinct Cores and Shells for Synergistic Release*, ACS Applied Materials & Interfaces, 2016, vol. 8, no. 13, pp. 8743–8754.
- [52] H. C. Chiu, Y. W. Lin, Y. F. Huang, C. K. Chuang, C. S. Chern, *Polymer vesicles containing small vesicles within interior aqueous compartments and pH-responsive transmembrane channels*, Angew Chem Int Ed Engl., 2008, vol. 47, no. 10, pp. 1875-1878.
- [53] R. R. Costa, E. Castro, F. J. Arias, J. C. Rodriguez-Cabello and J. F. Mano, *Multifunctional Compartmentalized Capsules with a Hierarchical Organization from the Nano to the Macro Scales*, Biomacromolecules, 2013, vol. 14, no. 7, pp 2403–2410.
- [54] B. Stadler, R. Chandrawati, A. D. Price, S. F. Chong, K. Breheny, A. Postma, L. A. Connal, A. N. Zelikin and F. Caruso, *A microreactor with thousands of subcompartments: enzyme-loaded liposomes within polymer capsules*, Angew Chem Int Ed Engl., 2009, vol 48, no. 24, pp. 4359-4362.
- [55] L. Hosta-Rigau, O. Shimoni, B. Stadler and F. Caruso, *Advanced Subcompartmentalized Microreactors: Polymer Hydrogel Carriers Encapsulating Polymer Capsules and Liposomes*, Small, 2013, vol. 9, no. 21, pp.3573-3583.
- [56] O. Kulygin, A. D. Price, S. F. Chong, B. Stadler, A. N. Zelikin and F. Caruso, *Subcompartmentalized Polymer Hydrogel Capsules with Selectively Degradable Carriers and Subunits*, Small, 2010, vol. 6, no. 14, pp. 1558–1564.
- [57] M. M. Hanczyc, S. M. Fujikawa and J. W. Szostak, *Experimental models of primitive cellular compartments: encapsulation, growth, and division*, Science, 2003, vol. 302, no. 5645, pp. 618-22.
- [58] F. Caschera, T. Sunami, T. Matsura, H. Suzuki, M. M. Hanczyc and T. Yomo, *Programmed Vesicle Fusion Triggers Gene Expression*. Langmuir, 2011, vol. 27, no. 21, pp. 13082–13090.
- [59] G. Agrawal, A. Ulpenich, X. M. Zhu, M. Moller and A. Pich, *Microgel-Based Adaptive Hybrid Capsules with Tunable Shell Permeability*, Chemistry of Materials, 2014, vol. 26, no. 2, pp. 5882–5891.
- [60] S. Seiffert, J. Thiele, A. R. Abate and D. A. Weitz, *Smart Microgel Capsules from Macromolecular Precursors*, Journal of the American Chemical Society, 2010, vol. 132, no. 18, pp. 6606–6609.
- [61] H. Duan, M. Kuang, G. Zhang, D. Wang, D. G. Kurth and H. Möhwald, *pH-Responsive Capsules Derived from Nanocrystal Templating*, Langmuir, 2005, vol. 21, no. 24, pp. 11495–11499.
- [62] S. A. Ulasevich, G. Brezesinski, H. Mohwald, P. Fratzl, F. H. Schacher, S. K. Poznyak, D. V. Andreeva and E. V. Skorb, *Light-Induced Water Splitting Causes High-Amplitude Oscillation of pH-Sensitive Layer-by-Layer Assemblies on TiO<sub>2</sub>*, Angewandte Chemie International Edition, 2016, vol. 55, no. 42, pp. 13001–13004.
- [63] S. A. Ulasevich, N. Brezhneva, Y. Zhukova, H. Mohwald, P. Fratzl, F. H. Schacher, D. V. Sviridov, D. V. Andreeva and E. V. Skorb, *Switching the Stiffness of Polyelectrolyte Assembly by Light to Control Behavior of Supported Cells*, Macromol Biosci., 2016, vol. 16, no. 10, pp. 1422-1431.
- [64] J. Gensel, T. Borke, N. P. Perez, A. Fery, D. V. Andreeva, E. Betthausen, A. H. Muller, H. Mohwald and E. V. Skorb, *Cavitation Engineered 3D Sponge Networks and Their Application in Active Surface Construction*, Advanced Materials, 2012, vol 24, no. 7, pp. 985–989.

- [65] E. V. Skorb, D. G. Shchukin, H. Mohwald and D. V. Sviridov, *Photocatalytically-active and photocontrollable coatings based on titania-loaded hybrid sol–gel films*, Journal of Materials Chemistry, 2009, vol. 19, no. 28, pp. 4931–4937.
- [66] F. Wu, R. Q. Su, Y. C. Lai and X. Wang, *Engineering of a synthetic quadrastable gene network to approach Waddington landscape and cell fate determination*, Elife, 2017, vol. 6, art. e23702.
- [67] A. A. Nikitina, S. A. Ulasevich, I. S. Kassirov, E. A. Bryushkova, E. I. Koshel and E. V. Skorb, *Nanostructured Layer-by-Layer Polyelectrolyte Containers to Switch Biofilm Fluorescence*, Bioconjugate Chemistry, 2018, vol. 29, no. 11, pp. 3793–3799.
- [68] P. S. Gromski, J. M. Granda and L. Cronin, *Universal Chemical Synthesis and Discovery with “The Chemputer.”*, Trends in Chemistry, 2020, vol. 2, no. 1, pp. 4–12.
- [69] S. Kriegman, D. Blackiston, M. Levin and J. Bongard, *A scalable pipeline for designing reconfigurable organisms*, Proceedings of the National Academy of Sciences, 2020, vol. 117, no. 4, pp. 1853–1859.
- [70] W. Cui, J. Li and G. Decher, *Self-Assembled Smart Nanocarriers for Targeted Drug Delivery*. Advanced Materials, 2015, vol. 28, no. 6, pp. 1302–1311.
- [71] V. Linko, A. Ora, M. A. Kostiainen, *Nanostructures as Smart Drug-Delivery Vehicles and Molecular Devices*, Trends Biotechnol., 2015, vol. 33, no. 10, pp. 586–594.
- [72] J. T. Yan, K. Liu, W. Li, H. Shi and A. F. Zhang, *Thermoresponsive Dendronized Polypeptides Showing Switchable Recognition to Catechols*, Macromolecules, 2016, vol. 49, no. 2, pp. 510–517.
- [73] N. Eslahi, M. Abdorahim and A. Simchi, *Smart Polymeric Hydrogels for Cartilage Tissue Engineering: A Review on the Chemistry and Biological Functions*, Biomacromolecules, 2016, vol. 17, no. 11, pp. 3441–3463.
- [74] F. Khan and M. Tanaka, *Designing Smart Biomaterials for Tissue Engineering*, International Journal of Molecular Sciences, 2017, vol. 19, no. 1, art. 17.
- [75] Y. Zhukova and E. V. Skorb, *Cell Guidance on Nanostructured Metal Based Surfaces*, Advanced Healthcare Materials, 2017, vol. 6, no. 7, art. 1600914.
- [76] A. Romeo, T. S. Leung and S. Sanchez, *Smart biosensors for multiplexed and fully integrated point-of-care diagnostics*, Lab on a Chip, 2016, vol. 16, no. 11, pp. 1957–1961.
- [77] V. Scognamiglio, A. Antonacci, M. D. Lambreva, S. C. Litescu and G. Rea, *Synthetic biology and biomimetic chemistry as converging technologies fostering a new generation of smart biosensors*, Biosens Bioelectron., 2015, vol. 74, pp. 1076–1086.
- [78] D. V. Andreeva, I. Melnyk, O. Baidukova and E. V. Skorb, *Local pH Gradient Initiated by Light on TiO<sub>2</sub> for Light-Triggered Modulation of Polyhistidine-Tagged Proteins*, ChemElectroChem, 2016, vol. 3, no. 9, pp. 1306–1310.
- [79] K. G. Nikolaev, V. Maybeck, E. Neumann, S. S. Ermakov, Y. E. Ermolenko, A. Offenhausser and Y. G. Mourzina, *Bimetallic nanowire sensors for extracellular electrochemical hydrogen peroxide detection in HL-1 cell culture*, J Solid State Electrochem, 2018, vol. 22, pp. 1023–1035.
- [80] K. Nikolaev, S. Ermakov, Y. Ermolenko, E. Averyaskina, A. Offenhausser and Y. Mourzina, *A novel bioelectrochemical interface based on in situ synthesis of gold nanostructures on electrode surfaces and surface activation by Meerwein’s salt. A bioelectrochemical sensor for glucose determination*, Bioelectrochemistry, 2015, vol. 105, pp. 34–43.
- [81] G. M. Kehe, D. I. Mori, M. J. Schurr and D. P. Nair, *Optically Responsive, Smart Anti-Bacterial Coatings via the Photofluidization of Azobenzenes*, ACS Applied Materials & Interfaces, 2019, vol. 11, no. 2, pp. 1760–1765.
- [82] D. Snihirova, S. V. Lamaka and M. F. Montemor, *Smart composite coatings for corrosion protection of aluminium alloys in aerospace applications*, In: *Smart Composite Coatings and Membranes: Transport, Structural, Environmental and Energy Applications*, (Woodhead Publishing Series in Composites Science and Engineering, 2016), pp. 85–121.
- [83] M. D. Manrique-Juárez, S. Rat, L. Salmon, G. Molnár, C. M. Quintero, L. Nicu, H. J. Shepherd and A. Bousseksou, *Switchable molecule-based materials for micro- and nanoscale actuating applications: Achievements and prospects*, Coordination Chemistry Reviews, 2016, vol. 308, pp. 395–408.
- [84] N. K. Persson, J. G. Martinez, Y. Zhong, A. Maziz and E. W. H. Jager, *Actuating Textiles: Next Generation of Smart Textiles*, Advanced Materials Technologies, 2018, art. 1700397.
- [85] A. Adamatzky, *Advances in Unconventional Computing, Volume 1: Theory* (Springer, 2017).

- [86] A. Adamatzky, *Advances in Unconventional Computing, Volume 2: Prototypes, Models and Algorithms*, (Springer, 2017).
- [87] G. Ur, J. Axthelm, P. Hoffmann, N. Taye, S. Glaser, H. Gorls, S. L. Hopkins, W. Plass, U. Neugebauer, S. Bonnet and A. Schiller, *Co-Registered Molecular Logic Gate with a CO-Releasing Molecule Triggered by Light and Peroxide*, *J Am Chem Soc.*, vol. 139, no. 14, pp. 4991-4994.
- [88] B. Mosadegh, T. Bersano-Begey, J. Y. Park, M. A. Burns and S. Takayama, *Next-generation integrated microfluidic circuits*, *Lab on a Chip*, 2011, vol 11, no. 17, 2813-2818.
- [89] M. Wehner, R. L. Truby, D. J. Fitzgerald, B. Mosadegh, G. M. Whitesides, J. A. Lewis and R. J. Wood, *An integrated design and fabrication strategy for entirely soft, autonomous robots*, *Nature*, 2016, vol 536, no. 7617, pp. 451-5.
- [90] K. Khoshmanesh, A. Almansouri, H. Albloushi, P. Yi, R. Soffe and K. Kalantar-zadeh, *A multi-functional bubble-based microfluidic system*, *Sci Rep.*, 2015, vol. 5, art. 9942.
- [91] G. T. Hwang, M. Byun, C. K. Jeong and K. J. Lee, *Flexible piezoelectric thin-film energy harvesters and nanosensors for biomedical applications*, *Adv Healthc Mater*, 2015, vol. 4, no. 5, pp. 646-58.
- [92] V. Jella, S. Ippili, J. H. Eom, S. V. N. Pammi, J. S. Jung, V. D. Tran, V. H. Nguyen, A. Kirakosyan, S. Yun, D. Kim, M. R. Sihn, J. Choi, Y. J. Kim, H. J. Kim and S. G. Yoon, *A Comprehensive Review of Flexible Piezoelectric Generators Based on Organic-Inorganic Metal Halide Perovskites*, *Nano Energy*, 2018, vol.57, pp.74-93.
- [93] G. Tan, L. D. Zhao and M. G. Kanatzidis, *Rationally Designing High-Performance Bulk Thermoelectric Materials*, *Chem Rev.*, 2016, vol. 116, no. 19, pp. 12123-12149.
- [94] H. Rodrigue, W. Wang, M. W. Han, T. J. Y. Kim and S. H. Ahn, *An Overview of Shape Memory Alloy-Coupled Actuators and Robots*, *Soft Robotics*, 2017, vol. 4, no. 1, pp 3–15.
- [95] A. Polman, M. Knight, E. C. Garnett, B. Ehrler and W. C. Sinke, *Photovoltaic materials: Present efficiencies and future challenges*, *Science*, 2016, vol. 352, no. 6283, art. aad4424.
- [96] N. V. Ryzhkov, D. V. Andreeva, E. V. Skorb, *Coupling pH-regulated multilayers with inorganic surface for bionic devices and infochemistry*, *Langmuir*, 2019, vol. 35, no. 26, pp. 8543–8556.
- [97] N. V. Ryzhkov, P. Nesterov, N. A. Mamchik, S. O. Yurchenko and E. V. Skorb, *Localization of Ion Concentration Gradients for Logic Operation*, *Frontiers in Chemistry*, 2019, vol 7, art. 419.
- [98] N.V. Ryzhkov, N.A. Mamchik and E.V. Skorb, *Electrochemical triggering of lipid bilayer lift-off oscillation at the electrode interface*, *J. R. Soc. Interface*, 2019, vol. 16, no. 150, art. 20180626.
- [99] Y. Li, T. Zhao, C. Wang, Z. Lin, G. Huang, B. D. Sumer and J. Gao, *Molecular basis of cooperativity in pH-triggered supramolecular self-assembly*, *Nat Commun.*, 2016, vol. 7, art.13214.
- [100] Z. Zhou, F. Guo, N. Wang, M. Meng and G. Li, *Dual pH-sensitive supramolecular micelles from star-shaped PDMAEMA based on  $\beta$ -cyclodextrin for drug release*, *Int J Biol Macromol.*, 2018, vol. 116, pp. 911-919.
- [101] S. Villani, R. Adami, E. Reverchon, A. M. Ferretti, A. Ponti, M. Lepretti, I. Caputo, L. Izzo, *pH-sensitive polymersomes: controlling swelling via copolymer structure and chemical composition*, *Journal of Drug Targeting*, 2017, vol. 25, no. 9-10, pp. 899-909.
- [102] Y. Zhang, P. An and X. Liu, *Bell-shaped sol-gel-sol conversions in pH-responsive worm-based nanostructured fluid*, *Soft Matter*, 2015, vol. 11, no. 11, pp. 2080–2084.
- [103] T. Garnier, A. Dochter, N. T. Chau, P. Schaaf, L. Jierry and F. Boulmedais, *Surface confined self-assembly of polyampholytes generated from charge-shifting polymers*, *Chemical Communications*, 2015, vol. 51, no. 74, pp. 14092–14095.
- [104] A. Dochter, T. Garnier, E. Pardieu, N. T. Chau, C. Maerten, B. Senger, P. Schaaf, L. Jierry and F. Boulmedais, *Directed Self-Assembly of Dipeptides to Form Ultrathin Hydrogel Membranes*, *Journal of the American Chemical Society*, vol. 132, no. 14, pp 5130–5136.
- [105] K. Sadman, Q. Wang, S. H. Chen, D. E. Delgado and K. R. Shull, *pH-Controlled Electrochemical Deposition of Polyelectrolyte Complex Films*, *Langmuir*, 2017, vol. 33, no. 8, pp. 1834–1844.
- [106] X. Zeng, X. Li, L. Xing, X. Liu, S. Luo, W. Wei, B. Kong and Y. Li, *Electrodeposition of chitosan-ionic liquid-glucose oxidase biocomposite onto nano-gold electrode for amperometric glucose sensing*, *Biosensors and Bioelectronics*, 2009, vol. 24, no. 9, pp. 2898–2903.
- [107] E. K. Johnson, D. J. Adams, P. J. Cameron, *Peptide based low molecular weight gelators*, *J. Mater. Chem.*, vol. 21, no. 7, pp. 2024–2027.

- [108] E. K. Johnson, D. J. Adams and P. J. Cameron, *Directed self-assembly of dipeptides to form ultrathin hydrogel membranes*, J Am Chem Soc, 2010, vol. 132, no. 14, pp. 5130-5136.
- [109] P. Kumaraswamy, R. Lakshmanan, S. Sethuraman and U. M. Krishnan, *Correction: Self-assembly of peptides: influence of substrate, pH and medium on the formation of supramolecular assemblies*, Soft Matter, 2017, vol. 13, no. 4, pp. 886–886.
- [110] J. Rodon Fores, M. L. Martinez Mendez, X. Mao, D. Wagner, M. Schmutz, M. Rabineau, P. Lavalle, P. Schaaf, F. Boulmedais and L. Jierry, *Localized Supramolecular Peptide Self-Assembly Directed by Enzyme-Induced Proton Gradients*, Angew. Chem. Int. Ed., 2017, vol. 56, no. 50, pp. 15984-15988.
- [111] H. M. Maltanava, S. K. Poznyak, D. V. Andreeva, M. C. Quevedo, A. C. Bastos, J. Tedim, M. G. S. Ferreira and E. V. Skorb, *Light-Induced Proton Pumping with a Semiconductor: Vision for Photoproton Lateral Separation and Robust Manipulation*, ACS Applied Materials and Interfaces, 2017, vol. 9, no. 28, pp. 24282-24289.
- [112] M. Criado-Gonzalez, L. Corbella, B. Senger, F. Boulmedais and R. Hernandez, *Photoresponsive Nanometer-Scale Iron Alginate Hydrogels: A Study of Gel-Sol Transition Using a Quartz Crystal Microbalance*, Langmuir, 2019, vol. 35, no. 35, pp. 11397-11405.
- [113] S. N. Semenov, A. S. Wong, R. M. van der Made, S. G. Postma, J. Groen, H. W. van Roekel, T. F. de Greef and W. T. Huck, *Rational design of functional and tunable oscillating enzymatic networks*, Nat Chem., 2015, vol. 7, no. 2, pp. 160-165.
- [114] E. Guisasola, A. Baeza, M. Talelli, D. Arcos and M. Vallet-Regi, *Design of thermoresponsive polymeric gates with opposite controlled release behaviors*, RSC Advances, 2016, vol. 6, no. 48, pp. 42510–42516.
- [115] R. A. Ghostine and J. B. Schlenoff, *Ion Diffusion Coefficients Through Polyelectrolyte Multilayers: Temperature and Charge Dependence*, Langmuir, 2011, vol. 27, no. 13, pp. 8241–8247.
- [116] R. Ou, J. Wei, L. Jiang, G. P. Simon and H. Wang, *Robust Thermoresponsive Polymer Composite Membrane with Switchable Superhydrophilicity and Superhydrophobicity for Efficient Oil-Water Separation*, Environ Sci Technol., 2016, vol. 50, no. 2, pp. 906-914.
- [117] K. Uhlig, T. Wegener, J. He, M. Zeiser, J. Bookhold, I. Dewald, N. Godino, M. Jaeger, T. Hellweg, A. Fery and C. Duschl, *Patterned Thermoresponsive Microgel Coatings for Noninvasive Processing of Adherent Cells*, Biomacromolecules, 2016, vol. 17, no. 3, pp. 1110–1116.
- [118] Z. Ahmed, A. Siiskonen, M. Virkki and A. Priimagi, *Controlling azobenzene photoswitching through combined ortho-fluorination and -amination*, Chemical Communications, 2017, vol. 53, no. 93, pp. 12520–12523.
- [119] D. B. Konrad, J. A. Frank and D. Trauner, *Redshifted Azobenzene Photoswitches by Late-Stage Functionalization*, Chemistry, 2016, vol. 22, no. 13, pp. 4364-4368.
- [120] S. Tamesue, Y. Takashima, H. Yamaguchi, S. Shinkai and A. Harada, *Photoswitchable supramolecular hydrogels formed by cyclodextrins and azobenzene polymers*, Angew Chem Int Ed Engl., 2010, vol. 49, no. 4, pp. 7461-7464.
- [121] C. Wang, M. Fadeev, J. Zhang, M. Vazquez-Gonzalez, G. Davidson-Rozenfeld, H. Tian and I. Willner, *Shape-memory and self-healing functions of DNA-based carboxymethyl cellulose hydrogels driven by chemical or light triggers*, Chemical Science, 2018, vol. 35, pp. 7145-7152.
- [122] F. Y. Zhou, B. Feng, T. T. Wang, D. G. Wang, Q. S. Meng, J. F. Zeng, Z. W. Zhang, S. L. Wang, H. J. Yu and Y. P. Li, *Cancer Therapy: Programmed Multiresponsive Vesicles for Enhanced Tumor Penetration and Combination Therapy of Triple-Negative Breast Cancer*, Advanced Functional Materials, 2017, vol. 27, no. 20.
- [123] M. Dobbelen, A. Ciesielski, S. Haar, S. Osella, M. Bruna, A. Minoia, L. Grisanti, T. Mosciatti, F. Richard, E. A. Prasetyanto, L. De Cola, V. Palermo, R. Mazzaro, V. Morandi, R. Lazzaroni, A. C. Ferrari, D. Beljonne and P. Samori, *Light-enhanced liquid-phase exfoliation and current photoswitching in graphene–azobenzene composites*, Nat Commun., 2016, vol. 7, art. 11090.
- [124] J. Luo, S. Samanta, M. Convertino, N. V. Dokholyan and A. Deiters, *Reversible and Tunable Photoswitching of Protein Function through Genetic Encoding of Azobenzene Amino Acids in Mammalian Cells*, ChemBioChem., 2018, vol. 19, no. 20, pp. 2178-2185.
- [125] Q. Bian, W. Wang, G. Han, Y. Chen, S. Wang and G. Wang, *Photoswitched Cell Adhesion on Azobenzene-Containing Self-Assembled Films*, ChemPhysChem, 2016, vol. 17, no. 16, pp. 2503-2508.

- [126] G. L. Nealon, D. H. Brown, F. Jones, G. Parkinson and M. I. Ogden, *An azobenzene-based photoswitchable crystal growth modifier*, CrystEngComm, 2017, vol. 19, no. 9, pp. 1286–1293.
- [127] Y. S. Kim, M. Liu, Y. Ishida, Y. Ebina, M. Osada, T. Sasaki, T. Hikima, M. Takata and T. Aida, *Thermoresponsive actuation enabled by permittivity switching in an electrostatically anisotropic hydrogel*, Nat Mater., 2015, vol. 14, no. 10, pp. 1002-1007.
- [128] A. W. Hauser, A. A. Evans, J. H. Na and R. C. Hayward, *Photothermally reprogrammable buckling of nanocomposite gel sheets*, Angew Chem Int Ed Engl., 2015, no. 54, no. 18, pp. 5434-5437.
- [129] Y. Yang, Y. Tan, X. Wang, W. An, S. Xu, W. Liao and Y. Wang, *Photothermal Nanocomposite Hydrogel Actuator with Electric-Field-Induced Gradient and Oriented Structure*, ACS Appl Mater Interfaces., 2018, vol. 10, no. 9, pp. 7688-7692.
- [130] Y. Zhou, A. W. Hauser, N. P. Bende, M. G. Kuzyk and R. C. Hayward, *Waveguiding Microactuators Based on a Photothermally Responsive Nanocomposite Hydrogel*, Advanced Functional Materials, 2016, vol. 26, no. 30, pp. 5447–5452.
- [131] S. A. Ulasevich, I. Melnyk, D. V. Andreeva, H. Mohwald and E. V. Skorb, *Photomobility and photohealing of cellulose-based hybrids*, EPL (Europhysics Letters), 2017, vol. 119, no. 3, art. 38003.
- [132] S. A. Ulasevich, N. Brezhneva, Y. Zhukova, H. Möhwald, P. Fratzl, F. H. Schacher, D. V. Sviridov, D. V. Andreeva and E. V. Skorb, *Switching the Stiffness of Polyelectrolyte Assembly by Light to Control Behavior of Supported Cells*, Macromolecular Bioscience, 2016, vol. 16, no. 10, pp. 1422–1431.
- [133] D. V. Andreeva, I. Melnyk, O. Baidukova and E. V. Skorb, *Local pH Gradient Initiated by Light on TiO<sub>2</sub>for Light-Triggered Modulation of Polyhistidine-Tagged Proteins*, ChemElectroChem, 2016, vol. 3, no. 9, pp. 1306–1310.
- [134] W. Macyk, G. Stochel and K. Szacilowski, *Photosensitization and the Photocurrent Switching Effect in Nanocrystalline Titanium Dioxide Functionalized with Iron(II) Complexes: A Comparative Study*. Chemistry - A European Journal, 2007, vol. 13, no.20, pp. 5676–5687.
- [135] K. Szacilowski and W. Macyk, *Photoelectrochemical Photocurrent Switching Effect: A New Platform for Molecular Logic Devices*, CHIMIA International Journal for Chemistry, 2007, vol. 61, no. 12, pp. 831–834.
- [136] A. Blachecki, J. Mech-Piskorz, M. Gajewska, K. Mech, K. Pilarczyk and K. Szacilowski, *Organotitania-Based Nanostructures as a Suitable Platform for the Implementation of Binary, Ternary, and Fuzzy Logic Systems*. ChemPhysChem, 2017, vol. 18, no. 13, pp. 1798–1810.
- [137] K. Szacilowski, W. Macyk and G. Stochel, *Light-Driven OR and XOR Programmable Chemical Logic Gates*, Journal of the American Chemical Society, 2006, vol. 128, no. 14, pp. 4550–4551.
- [138] L. Dong, A. K. Agarwal, D. J. Beebe and H. Jiang, *Adaptive liquid microlenses activated by stimuli-responsive hydrogels*, Nature, 2006, vol. 442, no. 7102, pp. 551-554.
- [139] T. Miyata, N. Asami and T. Uragami, *Structural design of stimuli-responsive bioconjugated hydrogels that respond to a target antigen*, Journal of Polymer Science Part B: Polymer Physics, 2009, vol. 47, no. 21, pp. 2144–2157.
- [140] W. L. A. Brooks, G. Vancoillie, C. P. Kabb, R. Hoogenboom and B. S. Sumerlin, *Triple responsive block copolymers combining pH-responsive, thermoresponsive, and glucose-responsive behaviors*, J. Polym. Sci., Part A-1: Polym. Chem., 2017, vol. 55, no. 14, pp. 2309-2317.
- [141] Q. Zhang, J. Colazo, D. Berg, S. M. Mugo and M. J. Serpe, *Multiresponsive Nanogels for Targeted Anticancer Drug Delivery*, Molecular Pharmaceutics, 2017, vol. 14, no. 8, pp. 2624–2628.
- [142] X. Le, W. Lu, H. Xiao, L. Wang, C. Ma, J. Zhang, Y. Huang and T. Chen, *Fe<sup>3+</sup>-, pH-, Thermoresponsive Supramolecular Hydrogel with Multishape Memory Effect*, ACS Applied Materials & Interfaces, 2017, vol. 9, no. 10, pp. 9038–9044.
- [143] C. X. Ma, X. X. Le, X. L. Tang, J. He, P. Xiao, J. Zheng, H. Xiao, W. Lu, J. W. Zhang, Y. J. Huang and T. Chen, *A Multiresponsive Anisotropic Hydrogel with Macroscopic 3D Complex Deformations*. Advanced Functional Materials, 2016, vol. 26, no. 47, pp. 8670–8676.
- [144] D. Morales, I. Podolsky, R. W. Mailen, T. Shay, M. D. Dickey and O. D. Velev, *Ionoprinted Multi-Responsive Hydrogel Actuators*, Micromachines, 2016, vol. 7, no. 6, art. 98.
- [145] X. Zhang and S. Soh, *Performing Logical Operations with Stimuli-Responsive Building Blocks*, Advanced Materials, 2017, vol. 29, no. 18, art. 1606483.

- [146] C. Wei, J. Guo and C. Wang, *Dual Stimuli-Responsive Polymeric Micelles Exhibiting “AND” Logic Gate for Controlled Release of Adriamycin*, Macromolecular Rapid Communications, 2011, vol. 32, no. 5, pp. 451–455.
- [147] B. A. Badeau, M. P. Comerford, C. K. Arakawa, J. A. Shadish and C. A. DeForest, *Engineered modular biomaterial logic gates for environmentally triggered therapeutic delivery*, Nat Chem., 2018, vol. 10, no. 3, pp. 251–258.
- [148] E. R. Ruskowitz, M. P. Comerford, B. A. Badeau and C. A. DeForest, *Logical stimuli-triggered delivery of small molecules from hydrogel biomaterials*, Biomaterials Science, 2019, vol. 7, no. 2, pp. 542–546.
- [149] Y. Dong, M. Liu, H. Zhang and B. Dong, *Reconfigurable OR and XOR logic gates based on dual responsive on-off-on micromotors*, Nanoscale, 2016, vol. 8, no. 15, pp. 8378–8383.
- [150] L. Zhang, H. Zhang, M. Liu and B. Dong, *Reprogrammable Logic Gate and Logic Circuit Based on Multistimuli-Responsive Raspberry-like Micromotors*, ACS Applied Materials & Interfaces, 2016, vol. 8, no. 24, pp. 15654–15660.
- [151] G. Yan, G. Tsekenis, B. Barzel, J.-J. Slotine, Y.-Y. Liu and A.-L. Barabási, *Spectrum of controlling and observing complex networks*, Nature Physics, 2015, vol. 11, no. 9, pp. 779–786.
- [152] K. Drescher, J. Dunkel, C. D. Nadell, S. van Teeffelen, I. Grnja, N. S. Wingreen, H. A. Stone and B. L. Bassler, *Architectural transitions inVibrio cholerae biofilms at single-cell resolution*, Proceedings of the National Academy of Sciences, 2016, vol. 113, no. 14, pp. E2066–E2072.
- [153] J. Dervaux, J. C. Magniez and A. Libchaber, *On growth and form of Bacillus subtilis biofilms*, Interface Focus, 2014, vol. 4, no. 6, pp. 20130051–20130051.
- [154] J. Humphries, L. Xiong, J. Liu, A. Prindle, F. Yuan, H. A. Arjes, L. Tsimring and G. M. Suel, *Species-Independent Attraction to Biofilms through Electrical Signaling*, Cell, 2017, vol. 168, no. 1-2, pp. 200–209.e12.
- [155] A. G. Vecchiarelli, M. Li, M. Mizuuchi, L. C. Hwang, Y. Seol, K. C. Neuman and K. Mizuuchi, *Membrane-bound MinDE complex acts as a toggle switch that drives Min oscillation coupled to cytoplasmic depletion of MinD*, Proceedings of the National Academy of Sciences, 2016, vol. 113, no. 11, pp. E1479–E1488.
- [156] A. Prindle, J. Liu, M. Asally, S. Ly, J. Garcia-Ojalvo and G. M. Suel, *Ion channels enable electrical communication in bacterial communities*, Nature, 2015, vol. 527, no. 7576, pp. 59–63.
- [157] A. Joesaar, S. Yang, B. Bogels, A. van der Linden, P. Pieters, B. Kumar, N. Dalchau, A. Phillips, S. Mann and T. F. A. de Greef, *DNA-based communication in populations of synthetic protocells*, Nat Nanotechnol., 2019, vol. 14, no. 4, pp. 369–378.
- [158] G. J. Melen, S. Levy, N. Barkai and B. Z. Shilo, *Threshold responses to morphogen gradients by zero-order ultrasensitivity*, Mol Syst Biol., 2005, vol. 1, art. 2005.0028.
- [159] N. E. Buchler and F. R. Cross, *Protein sequestration generates a flexible ultrasensitive response in a genetic network*, Mol Syst Biol., 2009, vol. 5, art. 272.
- [160] C. C. Govern and A. K. Chakraborty, *Signaling Cascades Modulate the Speed of Signal Propagation through Space*, PLOS ONE, 2009, vol. 4, no. 2, art. e4639.
- [161] S. N. Semenov, A. J. Markvoort, T. F. de Greef and W. T. Huck, *Threshold Sensing through a Synthetic Enzymatic Reaction-Diffusion Network*, Angew. Chem. Int. Ed., 2014, vol. 53, no. 31, pp. 8066–8069.
- [162] W. Xiong and J. E. Ferrell, Jr., *A positive-feedback-based bistable ‘memory module’ that governs a cell fate decision*, Nature, 2003, vol. 426, no. 6965, pp. 460–465.
- [163] B. N. Kholodenko, *Nat Cell-signalling dynamics in time and space*, Nat Rev Mol Cell Biol., 2006, vol. 7, no. 3, pp. 165–76.
- [164] N. Aubert, C. Mosca, T. Fujii, M. Hagiya and Y. Rondelez, *Computer-assisted design for scaling up systems based on DNA reaction networks*, Journal of The Royal Society Interface, 2014, vol. 11, no. 93, pp. 20131167–20131167.
- [165] V. K. Vanag, D. G. Miguez and I. R. Epstein, *Designing an enzymatic oscillator: Bistability and feedback controlled oscillations with glucose oxidase in a continuous flow stirred tank reactor*, The Journal of Chemical Physics, 2006, vol. 125, no. 19, art. 194515.
- [166] M. M. Wrobel, T. Bansagi, Jr., S. K. Scott, A. F. Taylor, C. O. Bounds, A. Carranza and J. A. Pojman, *pH wave-front propagation in the urea-urease reaction*, Biophys J., 2012, vol. 103, no. 3, pp. 610–615.

- [167] Y. Rondelez, *Competition for Catalytic Resources Alters Biological Network Dynamics*, Physical Review Letters, 2012, vol. 108, no. 1, art. 018102.
- [168] S. G. J. Postma, D. te Brinke, I. N. Vialshin, A. S. Y. Wong and W. T. S. Huck, *A trypsin-based bistable switch*, Tetrahedron, 2017, vol. 73, no. 33, pp. 4896–4900.
- [169] A. Goldbeter, D. Gonze, G. Houart, J. C. Leloup, J. Halloy and G. Dupont, *From periodic behavior to chaos in biological systems*, IFAC Proceedings Volumes, 2001, vol. 39, no. 8, p. 321.
- [170] E. Fung, W. W. Wong, J. K. Suen, T. Bulter, S. G. Lee and J. C. Liao, *A synthetic gene-metabolic oscillator*, Nature, 2005, vol. 435, no. 7038, pp. 118-22.
- [171] K. Kovacs, R. E. McIlwaine, S. K. Scott and A. F. Taylor, *An organic-based pH oscillator*, J Phys Chem A, 2007, vol. 111, no. 4, pp. 549-551.
- [172] M. J. Rust, J. S. Markson, W. S. Lane, D. S. Fisher and E. K. O'Shea, *Ordered phosphorylation governs oscillation of a three-protein circadian clock*, Science, 2007, vol. 318, no. 5851, pp. 809-12.
- [173] B. Novak and J. J. Tyson, *Design principles of biochemical oscillators*, Nat Rev Mol Cell Biol., 2008, vol. 9, no. 12, pp. 981-91.
- [174] J. Stricker, S. Cookson, M. R. Bennett, W. H. Mather, L. S. Tsimring and J. Hasty, *A fast, robust and tunable synthetic gene oscillator*, Nature, 2008, vol. 456, no. 7221, pp. 516-519.
- [175] J. Kim and E. Winfree, *Synthetic in vitro transcriptional oscillators*, Mol Syst Biol., 2011, vol. 7, art. 465.
- [176] J. E. Ferrell, Jr., T. Y. Tsai and Q. Yang, *Modeling the cell cycle: why do certain circuits oscillate?*, Cell, 2011, vol. 144, no. 6, pp. 874-85.
- [177] T. Fujii and Y. Rondelez, *Predator–Prey Molecular Ecosystems*, ACS Nano, 2013, vol. 7, no. 1, pp. 27-34.
- [178] A. S. Wong, S. G. Postma, I. N. Vialshin, S. N. Semenov and W. T. Huck, *Influence of Molecular Structure on the Properties of Out-of-Equilibrium Oscillating Enzymatic Reaction Networks*, Journal of the American Chemical Society, 2015, vol. 137, no. 38, pp. 12415–12420.
- [179] A. S. Y. Wong, A. A. Pogodaev, I. N. Vialshin, B. Helwig and W. T. S. Huck, *Molecular Engineering of Robustness and Resilience in Enzymatic Reaction Networks*, J. Am. Chem. Soc., 2017, vol. 139, no. 24, pp. 8146-8151.
- [180] A. Meister and M. E. Anderson, *Glutathione*, Annu Rev Biochem., 1983; vol. 52, pp. 711-760.
- [181] L. K. Moran, J. M. Gutteridge and G. J. Quinlan, *Thiols in cellular redox signalling and control*, Curr Med Chem., 2001, vol. 8, no. 7, pp. 763-72.
- [182] L. Qian and E. Winfree, *Scaling up digital circuit computation with DNA strand displacement cascades*, Science, 2011, vol. 332, no. 6034, pp. 1196-1201.
- [183] L. Qian, E. Winfree and J. Bruck, *Neural network computation with DNA strand displacement cascades*, Nature, 2011, vol. 475, no. 7356, pp. 368-372.
- [184] Y. J. Chen, N. Dalchau, N. Srinivas, A. Phillips, L. Cardelli, D. Soloveichik and G. Seelig, *Programmable chemical controllers made from DNA*, Nature Nanotechnology, 2013, vol. 8, no. 10, pp. 755–762.
- [185] Z. Xie, L. Wroblewska, L. Prochazka, R. Weiss and Y. Benenson, *Multi-input RNAi-based logic circuit for identification of specific cancer cells*, Science, 2011, vol. 333, no. 6047, pp. 1307-1311.
- [186] M. B. Miller and B. L. Bassler, *Quorum sensing in bacteria*, Annu Rev Microbiol., 2001, vol. 55, pp. 165-99.
- [187] G. Sirasani, L. Tong and E. P. Balskus, *A Biocompatible Alkene Hydrogenation Merges Organic Synthesis with Microbial Metabolism*Angew. Chem. Int. Ed., 2014, vol. 53, no. 30, pp. 7785-7788.
- [188] S. Wallace, E. E. Schultz and E. P. Balskus, *Using non-enzymatic chemistry to influence microbial metabolism*, Curr Opin Chem Biol., 2015, vol. 25, pp. 71-79.
- [189] S. Wallace and E. P. Balskus, *Interfacing Microbial Styrene Production with a Biocompatible Cyclopropanation Reaction*, Angew. Chem. Int. Ed., 2015, vol. 54, no. 24, pp. 7106-7109.
- [190] J. Liu, S. H. J. Chan, T. Brock-Nannestad, J. Chen, S. Y. Lee, C. Solem and P. R. Jensen, *Combining metabolic engineering and biocompatible chemistry for high-yield production of homo-diacetyl and homo-(S, S)-2,3-butanediol*, Metabolic Engineering, 2016, vol. 36, pp. 57–67.
- [191] R. Peri-Naor, T. Ilani, L. Motiei and D. Margulies, *Protein–Protein Communication and Enzyme Activation Mediated by a Synthetic Chemical Transducer*, Journal of the American Chemical Society, 2015, vol. 137, no. 30, pp. 9507–9510.

- [192] R. Peri-Naor, L. Motiei and D. Margulies, *Artificial signal transduction therapy: a futuristic approach to disease treatment*, Future Medicinal Chemistry, 2015, vol. 7, no. 16, pp. 2091–2093.
- [193] J. Cao, C. Zhou, G. Su, X. Zhang, T. Zhou, Z. Zhou and Y. Yang, *Arbitrarily 3D Configurable Hygroscopic Robots with a Covalent-Noncovalent Interpenetrating Network and Self-Healing Ability*, Adv. Mater., 2019, vol. 31, art. e1900042.
- [194] A. Henson, J. M. Gutierrez, T. Hinkley, S. Tsuda and L. Cronin, *Towards heterotic computing with droplets in a fully automated droplet-maker platform*, Philos. Trans. A Math. Phys. Eng. Sci., 2015, vol. 373, no. 2046, art. 20140221.
- [195] S. J. Park, M. Gazzola, K. S. Park, S. Park, V. Di Santo, E. L. Blevins, J. U. Lind, P. H. Campbell, S. Dauth, A. K. Capulli, F. S. Pasqualini, S. Ahn, A. Cho, H. Yuan, B. M. Maoz, R. Vijaykumar, J. W. Choi, K. Deisseroth, G. V. Lauder, L. Mahadevan and K. K. Parker, *Phototactic guidance of a tissue-engineered soft-robotic ray*, Science, 2016, vol. 353, no. 6295, pp. 158-162.
- [196] K. W. Moored, F. E. Fish, T. H. Kemp and H. Bart-Smith, *Batoid Fishes: Inspiration for the Next Generation of Underwater Robots*, Marine Technology Society Journal, 2011, vol. 45, no. 4, pp. 99–109.
- [197] K. Y. Ma, P. Chirarattananon, S. B. Fuller and R. J. Wood, *Controlled flight of a biologically inspired, insect-scale robot*, Science, 2013, vol. 340, no. 6132, pp. 603-607.
- [198] J. S. Koh, E. Yang, G. P. Jung, S. P. Jung, J. H. Son, S. I. Lee, P. G. Jablonski, R. J. Wood, H. Y. Kim and K. J. Cho, *Jumping on water: Surface tension-dominated jumping of water striders and robotic insects*, Science, 2015, vol. 349, no. 6247, pp. 517–521.
- [199] N. V. Ryzhkov, N. Brezhneva and E. V. Skorb, *Feedback mechanisms at inorganic–polyelectrolyte interfaces for applied materials*, Surface Innovations, 2019, vol. 7, no. 3-4, pp. 145-167.
- [200] Y. Lanchuk, A. Nikitina, N. Brezhneva, S. A. Ulasevich, S. N. Semenov and E. V. Skorb, *Photocatalytic Regulation of an Autocatalytic Wave of Spatially Propagating Enzymatic Reactions* Chemcatchem, 2018, vol. 10, no. 8, pp. 1798-1803.
- [201] E. V. Skorb, D. V. Andreeva, A. P. Raiski, N. A. Belyasova, H. Mohwald and D. V. Sviridov, *Titanium dioxide-assisted photocatalytic induction of prophages to lytic cycle*, Photochemical & Photobiological Sciences, 2011, vol. 10, no. 12, pp. 1974-1978.
- [202] A. Prindle, J. Liu, M. Asally, S. Ly, J. Garcia-Ojalvo and G. M. Süel, *Ion channels enable electrical communication in bacterial communities*, Nature, 2015, vol. 527, no. 7576, pp. 59-63.
- [203] J. Gensel, T. Borke, N. P. Pérez, A. Fery, D. V. Andreeva, E. Betthausen, A. H. E. Müller, H. Möhwald and E. V. Skorb, *Active Surfaces: Cavitation Engineered 3D Sponge Networks and Their Application in Active Surface Construction*, Advanced Materials, vol. 24, no. 7, art. 984.