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Switchable anti-bacterial hydrogel layers with self-cleaning capabilities

Abstract

One of the major challenges for modern implantology is the successful implant integration without future adverse effects. Bacterial colonization of the implant-tissue-interface and the formation of a biofilm specifically needs to be avoided.[1,2] While quaternized ammonium cations (QACs) are known to kill bacteria,[3] their major drawback is potential inactivation due to accumulation of dead bacteria on the surface.[4] In order to avoid this inactivation, zwitterionic polyelectrolytes (PEs) can be introduced, resulting in a desorption of bacterial debris.[5] The goal is to create a system with the ability to switch between the desorption and the bacteria-killing mode.

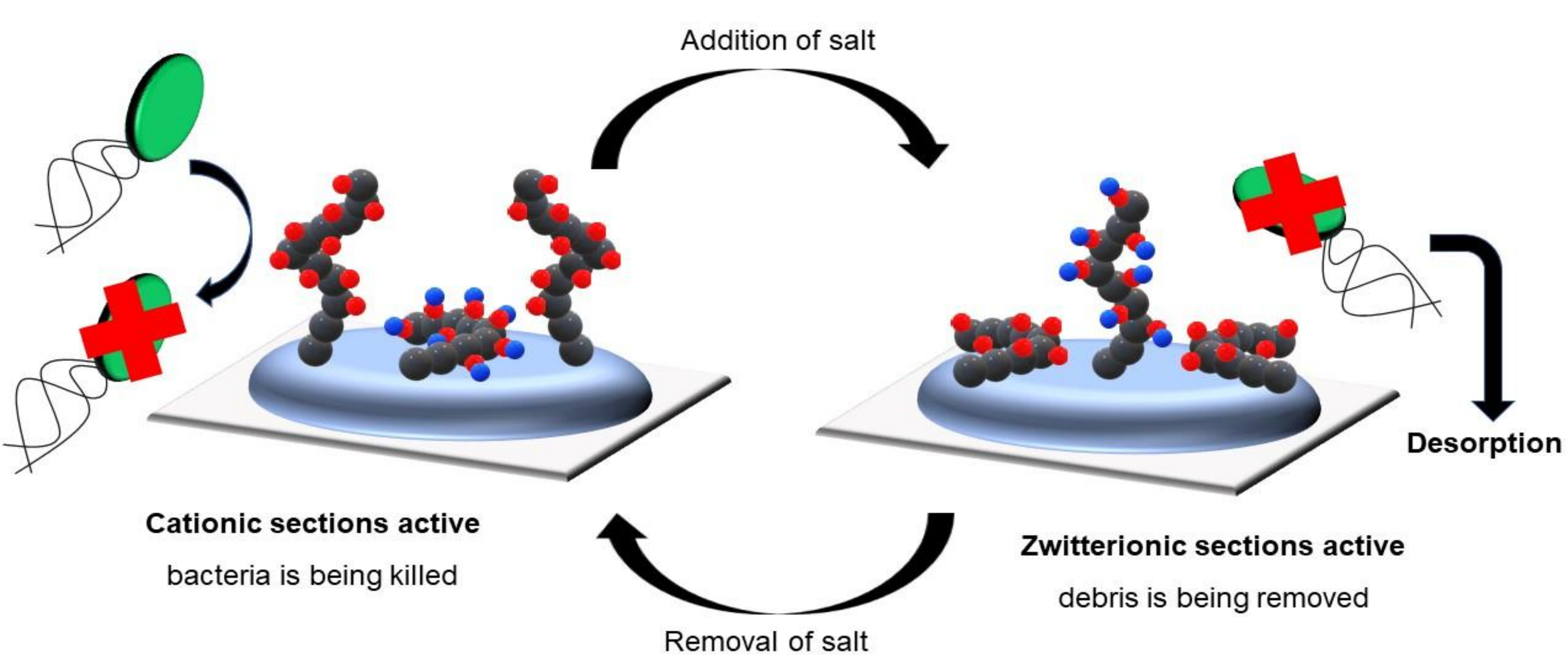
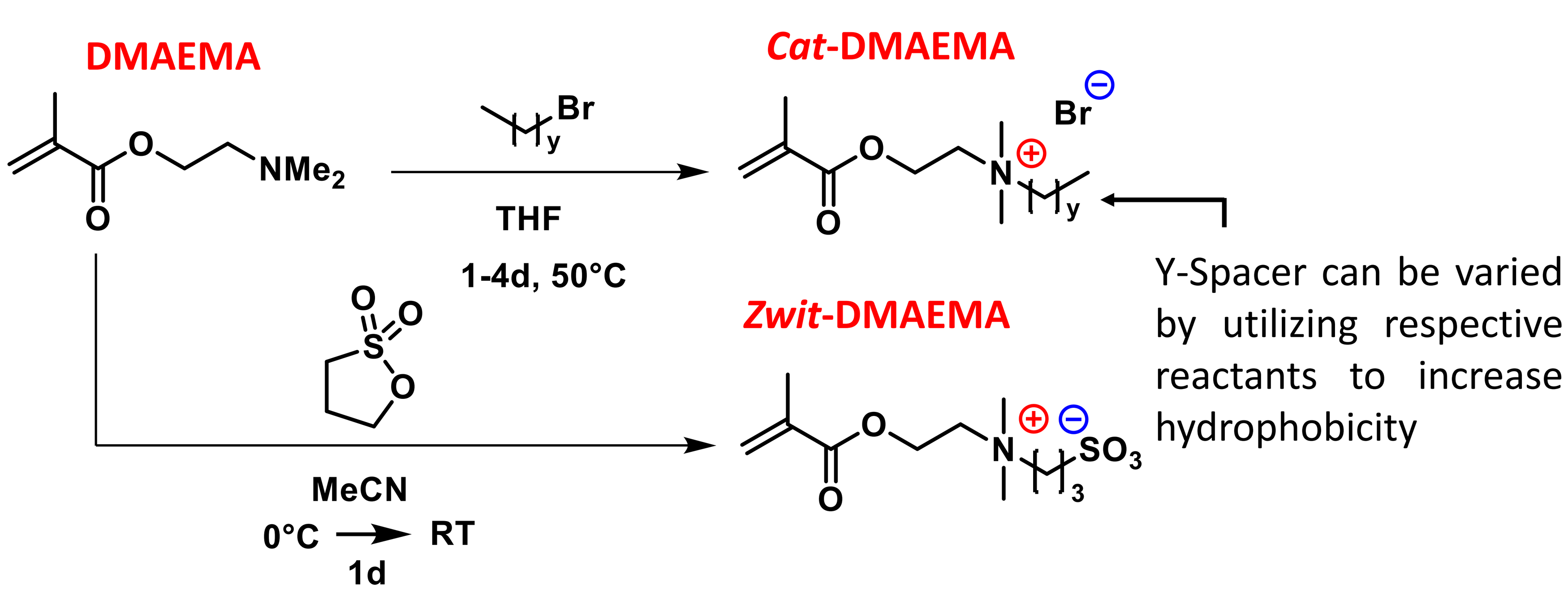


Figure 1: General principle of the switchability of the synthesized network. The gel portion (in blue) of the network will be used as an anchor to help the polymer adhere to the titanium surface (in grey). In environments with low salt concentrations the cationic portion of the network (black-red chain) is active whereas in high salt concentrations the zwitterionic portions (black-red-blue chain) is activated.

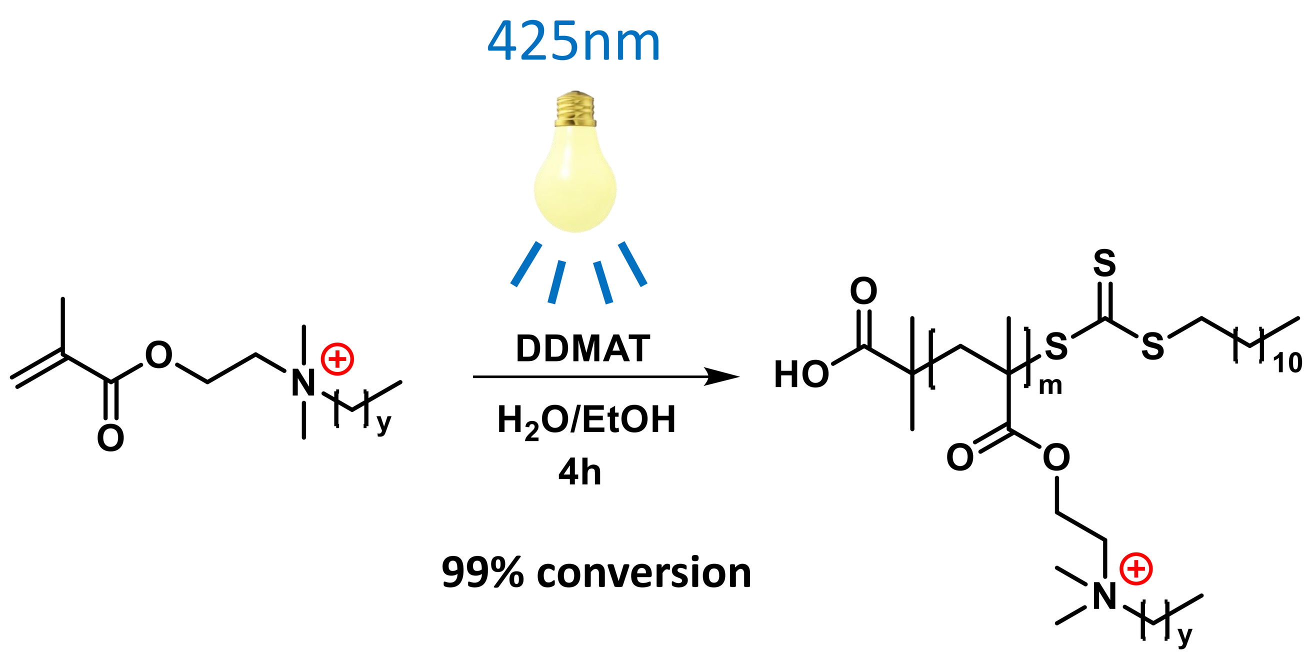
Monomer synthesis



Since the ionic monomers can be isolated as solids and show good solubility for aqueous polymerizations, this pathway is preferred over a post-modification of the respective polymers.

Aqueous photoiniferter RAFT

Classical RAFT approaches utilize elevated temperatures to generate free radicals from initator molecules. However, it is also possible to generate free radicals by irradiation of the CTA with specific wavelengths. No initiator is needed and the polymerization can be conducted at lower temperatures.



Photoiniferter RAFT also enables polymerization for temperature sensitive RAFT agents like CTPA. This can be achieved with green light at a wavelength of 525 nm.

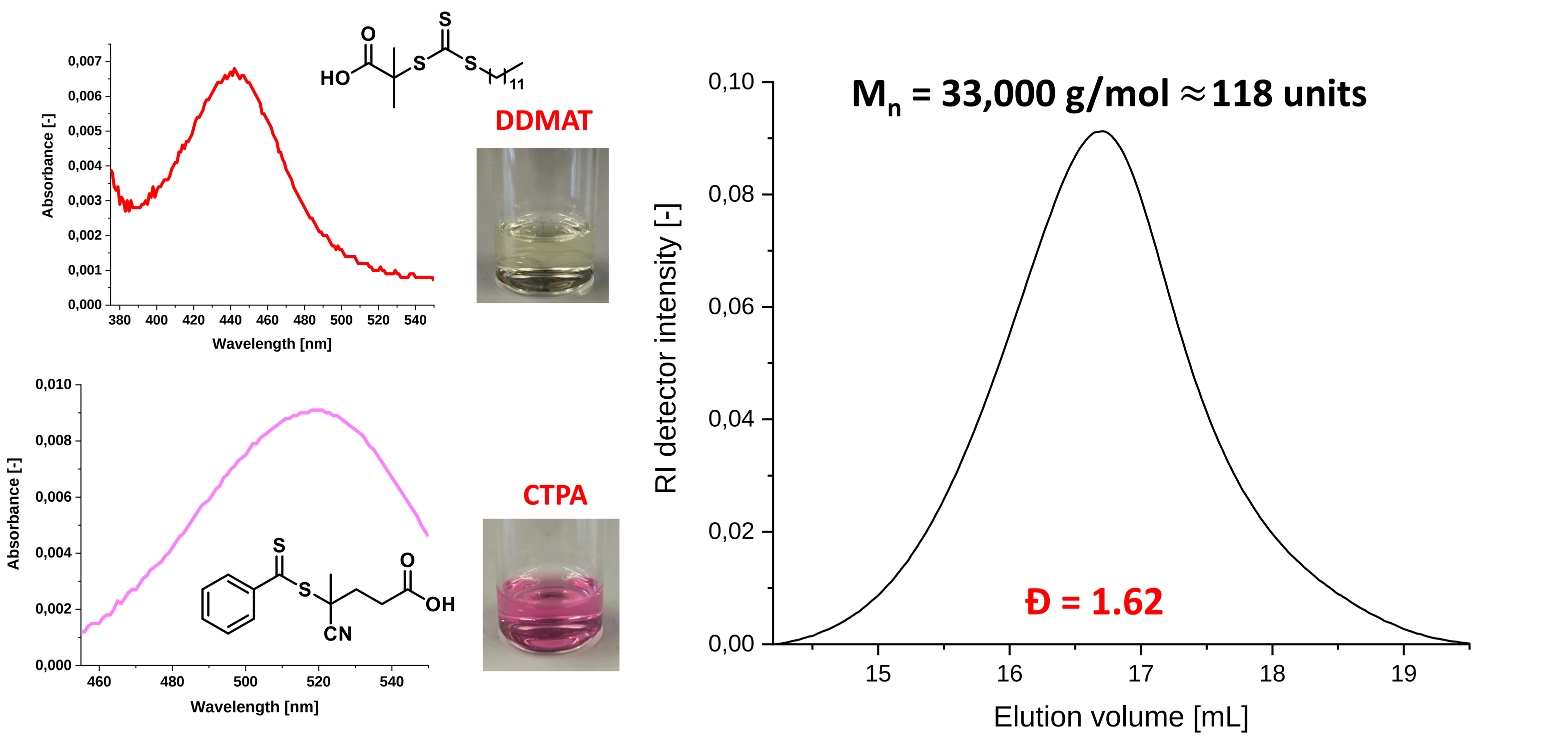


Figure 2: UV-Vis measurements of DDMAT (top left), CTPA (bottom left) RAFT agents and GPC data of a cationic DMAEMA polymer synthesized with DDMAT via photoiniferter RAFT in Water/Ethanol with a ratio of 25:1 monomer:CTA. GPC was measured calibrated with PMMA standards with a low Đ, BHT as an internal standard and with a concentration of 4 mg/ml.

Anti-bacterial experiments

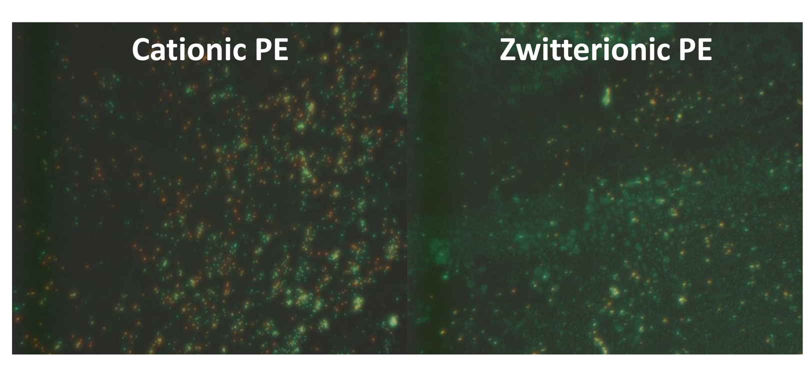


Figure 3: Fluorescence spectroscopy images of gold wafers that were immersed in 0.1 mol solutions of the respective SSMe-modified PEs for 24 h and treated with a solution of E. coli K12 in PBS buffer. All bacteria are stained in green while damaged bacteria are additionally stained in red.

First experiments with self-assembled monolayers of the PEs show the expected responses towards E. coli K12: Many (dead) bacteria on the cationic surface and only few bacteria on the zwitterionic surface.

References
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[3] R. Methling, O. Dückmann, F. Simon, C. Wolf-Brandstetter, D. Kuckling, *Macromol. Mater. Eng.* **2023**, *308*.
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Abbreviations
RAFT: reversible addition-fragmentation chain transfer
DDMAT: 2-(Dodecylthiocarbonothioylthio)-2-methylpropionic acid
CTPA: 4-Cyano-(phenylcarbonothioylthio)pentanoic acid
CTA: Chain transfer agent
QAC: Quaternary ammonium cation
PE: Polyelectrolyte

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