Executive Summary

Prof. Christoph Weder, Université de Fribourg Project "Bio-Inspired Mechanically Responsive Polymer Nanocomposites"

This project sought to develop smart materials, which change their mechanical properties upon exposure to a pre-defined stimulus. This behavior is useful for many applications and the investigation of the underlying mechanistic aspects is of fundamental interest. The materials design was inspired by sea cucumbers, which can change the stiffness of their skin on command. This is achieved through a nanocomposite architecture in which rigid nanofibers reinforce a soft matrix. The stiffness is varied by changing the interactions between the fibers via a binding peptide, which is produced (stiff state) or removed (soft state). The goals of this project were to create artificial materials that mimic this architecture and mechanism and to develop an understanding for the structure-property relationships of such materials.

The first bio-inspired nanocomposites studied in this project were prepared by compounding soft polymers and rigid, rod-like cellulose nanocrystals (CNCs) isolated from natural resources. On account of hydrogen-bonding between the hydroxyl groups that are naturally present on their surface, the CNCs form a reinforcing network within the polymer matrix, causing the materials to be very stiff. Upon exposure to water, however, the CNC network is disrupted, which results in a dramatic softening. Based on this general design approach, a range of new materials were designed, prepared, and investigated.

A first set of materials was studied in the context of cortical microelectrodes, which allow electrical contacts with the brain and are potentially useful for the treatment of neurological deficits. The functionality of experimental electrodes is known to decrease over time, possibly due to the mechanical mismatch between the stiff implant and the soft brain tissue, which leads to neuron degeneration and foreign body encapsulation. To confirm this hypothesis, and with the goal to develop better microelectrodes, water responsive, mechanically adaptive polymer nanocomposites were developed, which are initially highly rigid, but soften considerably upon exposure to physiological conditions. In collaboration with partners at Case Western Reserve University (Cleveland OH, USA) it was shown through in-vivo studies that implants based on such adaptive materials cause significantly lower neuroinflammatory and neurodegenerative responses than rigid reference materials and promise to be a valuable platform for the design of intra-cortical devices. Building on these results, we introduced physiologically responsive materials that also release an antiinflammatory drug that combats the acute trauma response caused by the implantation. Invivo results show that the combination of the two mechanisms - softening of the implant and drug release - indeed leads to synergistic effects. Noting that the ability to deliver light to specific locations within the brain using optogenetic tools has opened up new possibilities in the field of neural interfacing, we also developed a first generation of physiologically responsive optical fibers that can be inserted into the brain to activate or mute neurons using photosensitive proteins. Based on the encouraging data from this project, the development of smart materials for cortical interfacing is continuing.

The principle used in the above-described materials was extended to create shape-memory nanocomposites in which a pre-programmed shape change can be triggered upon exposure to water, as well as mechanically adaptive materials that change their properties quite selectively in a specific pH range. Such specificity was achieved by surface-modification of the CNCs with specific binding motifs.

Taking this approach to the next level, we introduced light-responsive, mechanically adaptive materials in which CNCs were modified to feature photo-reactive surface groups. The latter can be used to introduce chemical bonds between the CNCs upon exposure to ultraviolet light. We demonstrated that this design can be used to create nanocomposites which stiffen upon exposure to light. The concept was further extended to create optically healable polymer nanocomposites, in which damages can be easily healed through exposure to UV light.

Overall, the research conducted in this project has afforded a range new materials with intriguing functions and contributed significantly to the understanding of stress-transfer mechanisms at play in these nanocomposites.

The activities supported by this SNSF grant have supported 4 PhD students (2 graduated, 2 in progress). The SNSF support was leveraged by significant funding from three industrial partners with whom specific application ideas were/are investigated. The results were disseminated through so far ca. 25 peer-reviewed papers and book chapters and form the basis of 3 patent applications. The project also nucleated 2 CTI projects that complemented the NRP-funded activities and which resulted in additional papers (2 to date) and patent applications (1 to date).