“Electrically induced healing of a Diels-Alder-CNT polyurethane composite”

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Thermoreversible [4+2] Diels-Alder chemical bonds are widely used in the field of intrinsic self-healing polymers¹². These reversible covalent bonds can be incorporated into polymeric matrices to reform broken bonds upon heating. The most commonly used Diels-Alder moieties are the furan (diene) and the maleimide (dienophile) because of their low temperatures of activation (T_retro-DA = 120°C, T_DA = 50°C, cf. Figure 1).

![Figure 1. Thermoreversible Diels-Alder reaction](image)

In this communication, we introduce a new Diels-Alder based self-healing polyurethane composite containing multi-walled carbon nanotubes (MWNT). The self-healing polyurethane was synthesized through reactive extrusion process (REX). With this technique, a polyurethane based pre-polymer bearing maleimide pendant groups, obtained by polycondensation in bulk between hexamethylene diisocyanate, dodecanediol and a diol bearing a maleimide function, is crosslinked with a difurane coupling agent in order to form thermoreversible networks. This manufacturing process of self-healing polymers by REX offers several advantages to other methods such as the short residence times (synthesis time), the lack of use of solvents, the good dispersion of the MWNT into the polymeric matrix and its scalability into a real industrial process. The MWNT were introduced to promote the healing of the polymer matrix based on the electrical properties of the MWNT: by application of a current,
the temperature of the polymer rises by Joule effect and leads to the necessary mobility and rDA reaction necessary to close cracks. As monitored by IR-camera, the electrically-induced temperature increase is in the range of the retro-Diels-Alder reactions. Moreover a temperature increase at the edge of the crack was observed leading to a very localized crack closure by zipping effect. The healing efficiency of this new polyurethane nanocomposite was attested via scratch test and IR camera, while a rheometer coupled with the FT-IR spectroscopy was used to study the reversibility and the kinetics of the Diels-Alder reactions occurring under heating/cooling cycles.

References
