ABSTRACT

The subject of the presented work is the embedding of self-healing functions into the technically important elastomers EPDM, SEBS and NBR. During service in technical parts, these materials are exposed to mechanical load, UV radiation or liquids and gases, promoting the ageing of the polymers. Occurring damages, usually starting from the size of micro-cracks, may spread rapidly and uncontrolled, leading to failure even before the designated interval for maintenance.

Within this work, three different strategies leading to autonomous repair of the polymeric materials are introduced and evaluated. In extrinsic healing concepts, one- and two-component adhesive systems are microencapsulated and embedded into the polymeric matrix. As an alternative extrinsic approach, blends of the technical compound and a second polymeric component are investigated. In addition, intrinsic self-healing systems are studied. The matrix polymers are modified with reversible cross-links such as hydrogen bonds and ionic groups.

It is shown that filled microcapsules, implemented to introduce an extrinsic self-healing function, do not withstand the manufacturing process of elastomeric compounds. However, the synthesis and incorporation of loaded SiO₂ carrier particles is successful. Yet, in the case of damage, these capsules do not necessarily break apart upon impact due to the insufficient linkage to the matrix.

Reproducible healing results can be quantified for SEBS and EPDM polymers in blend systems with highly adhesive secondary polymeric components such as polyisobutylene or boron networked polydimethylsiloxane. The healing effectiveness depends on the nature and the quantity of the additive. NBR based polymers are modified with functional groups forming hydrogen bonds (urazole modification), or by the ionization of carboxylated side groups. These polymers show a pronounced, in some cases, complete self-healing behavior. In addition, due to the reversible cross-links, the Young's modulus and the stress at break are increased and the recovery behavior is improved. The resulting materials may be utilized for different technical applications with no further covalent cross-linking.

Viscoelastic properties and network parameters are obtained by step-strain, hysteresis and oscillatory shear experiments and interpreted according to prevailing polymer molecular theories.

As an improvement to common mechanical self-healing testing procedures, using rectangular or dumbbell shaped specimens, a tensile testing method applying cylindrical test samples is presented. By the use of cylindrical testing geometries in a uniaxial tensile test, homogeneous strain states within the tested sample are obtainable and, especially for soft materials, an inhomogeneous deformation may be excluded.