

## SUBPROJECT 3: Sustainable and recyclable matrix and functional elastomers for I-FRC with hybrid and time-variable crosslinking structures

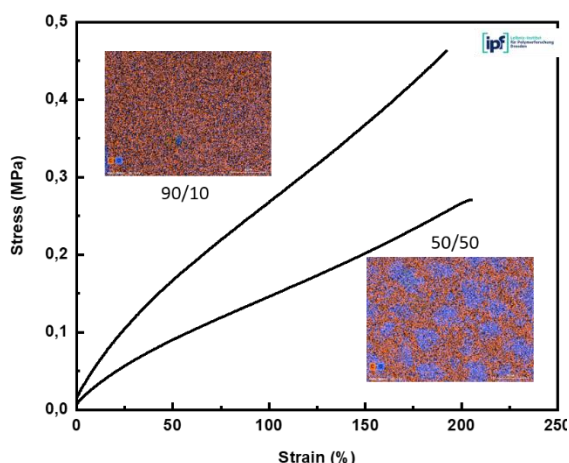
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### Motivation

Aspects of energy- and resource-efficient production, service life performance and recyclability must be considered as early as the design stage of interactive fibre-elastomer composites. This applies in particular to the elastomer materials used both as a matrix and as functional polymer domains in the composite, which represent a major challenge for recyclability as they are usually covalently linked polymer networks. The development and implementation of dynamically cross-linked, thermoreversible or vitrimer-like elastomers based on biogenic polymers in I-FRCs is a promising approach for sustainable I-FRCs. The expected effects on the thermo-mechanical processing and material behaviour, on composite production, boundary layer interaction and composite functionality, especially with regard to long-term properties, are unknown.

### State of the art and previous research

The implementation of various concepts for the realisation of non-covalently cross-linked polymer networks with the aim of producing self-healing and remeltable and thus recyclable elastomers continues to be the subject of intensive research. In addition to H-bridge bonds, ionic interactions, complex formation and dynamic bonding concepts via vitrification are also used [1-3], although the combination of such elastomers with functional or reinforcing textile structures has not yet been researched, and in particular the necessary boundary layer design for phase bonding and long-term performance have not yet been clarified. Our own preliminary work has focussed on the production of thermo-reversibly cross-linked self-healing elastomers based on ionically modified bromobutyl rubbers and reversible ENR networks produced by means of metal complexation and diacid curing [4-7]. While the material behaviour of such elastomers is well qualified for tyre-relevant applications, especially with regard to cracking and wear behaviour, there is no scientifically sound knowledge about the suitability of novel elastomers [8] with temporally and thermally variable cross-linking structures in view of the specific processing, compound formation and functional boundary conditions in I-FRC. This will be the subject of research in SP 3.



SEM-EDX images and stress-strain response of elastomers having liquid isoprene rubber (LIR-403 from Kuraray) and epoxy crosslinker (Polydis 3616 from Struktol) in 90/10 and 50/50 ratio

### Scientific question and project objectives

The main objective of SP 3 in the 3rd cohort is the research and implementation of hybrid and reversible non-covalent cross-linking concepts (e.g. ionic thermoreversible, vitrification) in bio-based three dimensional printable diene rubbers and/or thermoplastic elastomers, which

should be able to be integrated into recyclable I-FRCs. Of central importance is a comprehensive thermo-mechanical characterisation and modelling of the material and processing properties dependent on the instantaneous network structure; the description of the temperature and time-dependent stress relaxation of these novel networks and ultimately the coupling of these adaptive material characteristics to predict the long-term and ageing behaviour of these elastomer components of I-FRCs produced on a sustainable basis. Compared to conventionally covalently cross-linked elastomers, the dynamic network structure of these elastomers requires a thermo-rheologically different processing behaviour during production, so that a suitable process parameter space must be found for composite formation. Finally, suitable characterisation methods and procedures must be researched for the qualification and quantification of the material recyclability of these non-conventional elastomers, which is already necessary at the material level, and implemented in sufficiently descriptive parameters or models – phenomenologically or on the basis of physically motivated approaches taking into account structural evolution – to describe the visco-elastic material behaviour.

There is close cooperation with SP 1, SP 2, SP 8 to define the requirements for the composite formation properties and possible recycling scenarios and with SP 10 to correlate the long-term behaviour characterised at material level with the fatigue behaviour on I-FRC. In cooperation with SP11 also possibilities of 3D printing of filaments of novel functional elastomers will be investigated. With SP 6, an intensive exchange is taking place to implement the thermo-mechanical material behaviour of non-covalently cross-linked elastomers in the constitutive modelling.

## References

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