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Bouvard

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Rheological and mechanical behaviour of polyethylene-based vitrimers

T. Huang¹, J. Alves², R. Castellani¹, Y. Tillier¹, J.-L. Bouvard¹

¹ Mines Paris, PSL University, Centre for material forming (CEMEF), UMR CNRS, 06904 Sophia Antipolis, France
(tianqi.huang.mines@gmail.com)

² Aliaxis Research & Technology, Omega Park, Building 7, 3 Bd Jean Moulin, 78990 Élancourt, France

The use-driven classification of polymers tends to separate them into two groups, thermoset and thermoplastic polymers, depending on their mechanical and rheological behaviour with temperature. The former usually show better mechanical performances while the latter present interesting reprocessing abilities thanks to the reversibility and thermal dependency of its flowing properties. Recently, an interesting kind of material has emerged, that presents both characteristics: vitrimers. They consist of polymer chains “crosslinked” using thermo-activated labile bonds, forming a dynamic network that allows relaxation and chain mobility at high enough temperatures. A characteristic temperature T_v (or “topology freezing transition temperature”) can be associated with this process, above which the vitrimer will exhibit thermoplastic-like healing and shaping properties, and below which it will show interesting thermoset-like mechanical properties.

In this work, three different polyethylene-based materials are studied: (i) a common HDPE, (ii) a silane-crosslinked polyethylene (PEXb) and (iii) PE-vitrimers¹. Mechanical and flow properties are compared, using rheology and DMA. Measurements highlight how the PE-vitrimer falls between HDPE and PEXb at high temperatures; The variation in vitrimer bond density also refines the behavior from entangled polymer chains to a cross-linked network. Interestingly, the attempt to draw a master curve from the rheological data at different temperatures only works in certain frequency ranges, as highlighted in Figure 1. This seems to highlight different relaxation processes due to the characteristic times and activation energies of the PE-chains and vitrimer network bonds respectively.

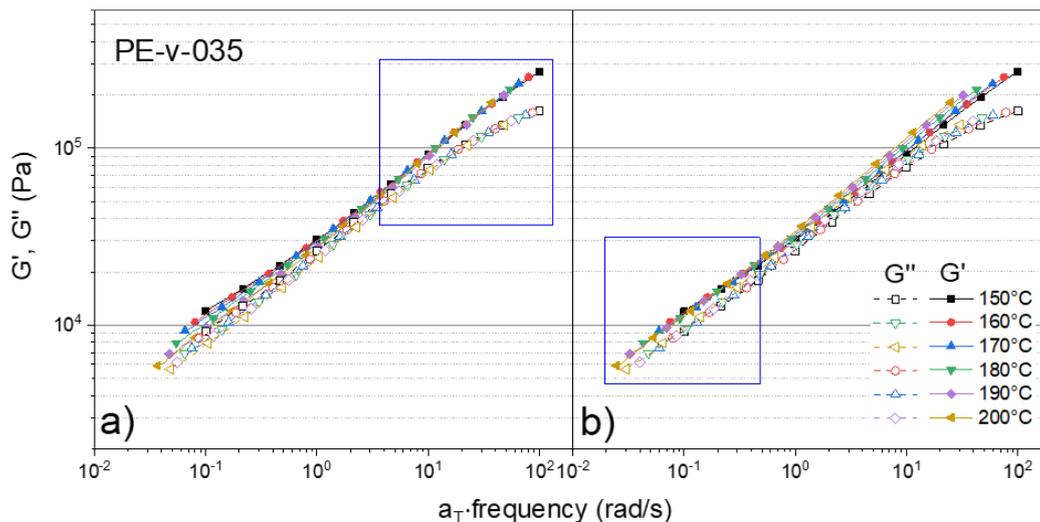


Figure 1: Master curves of G' and G'' for one of the studied PE vitrimers (PE-v-035), from 150°C to 200°C using shifts in a) high-frequency range or b) low-frequency range (blue squares).

¹ M. Maaz, *et al.*, *Macromolecules*. **54**, 2213 (2021).