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Flory-Stockmayer analysis on reprocessable polymer networks¹

LINGQIAO LI, XI CHEN, KAILONG JIN, JOHN TORKEKELSON, Department of Chemical and Biological Engineering, Northwestern University — Reprocessable polymer networks can undergo structure rearrangement through dynamic chemistries under proper conditions, making them a promising candidate for recyclable crosslinked materials, e.g. tires. This research field has been focusing on various chemistries. However, there has been lacking of an essential physical theory explaining the relationship between abundancy of dynamic linkages and reprocessability. Based on the classical Flory-Stockmayer analysis on network gelation, we developed a similar analysis on reprocessable polymer networks to quantitatively predict the critical condition for reprocessability. Our theory indicates that it is unnecessary for all bonds to be dynamic to make the resulting network reprocessable. As long as there is no percolated permanent network in the system, the material can fully rearrange. To experimentally validate our theory, we used a thiol-epoxy network model system with various dynamic linkage compositions. The stress relaxation behavior of resulting materials supports our theoretical prediction: only 50 % of linkages between crosslinks need to be dynamic for a tri-arm network to be reprocessable. Therefore, this analysis provides the first fundamental theoretical platform for designing and evaluating reprocessable polymer networks.

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