Thermodynamic regularization based computational framework for strain-induced crystallization

Mahmood Jabareen[1,*], **Vu Nguyen-Xuân**[2], **Mikhail Itskov**[2], **Jean Benoît Le Cam**[3] **and Vu Ngoc Khiêm**[2]

[1]Faculty of Civil and Environmental Engineering Technion, Israel Institute of Technology, Israel
e-mail: cvjmah@technion.ac.il

[2]Department of Continuum Mechanics, RWTH Aachen University, Aachen, Germany

[3]Université de Rennes, CNRS, IPR (Institut de Physique de Rennes) - UMR 6251, Rennes, France

KEYWORDS: *Thermodynamic regularization, Strain-induced crystallization, Loss of ellipticity*

Strain-induced crystallization (SIC) represents a type of phase transition in natural rubbers triggered by deformation. This is evident from the heterogeneities in temperature distribution observed in unfilled natural rubbers subjected to homogeneous uniaxial tension [1], as temperature variations in natural rubber are attributed to SIC. These heterogeneities become pronounced following the onset of SIC, particularly within the stretch ratio range of $4 \lessapprox \Lambda \lessapprox 5.5$ (see Fig. 1). Interestingly, prior to the onset of SIC ($\Lambda_1 \lessapprox 4$) and following the stress upturn ($\Lambda_1 \gtrapprox 6$), the temperature distribution remains homogeneous. This observation further implies that this behavior does not stem from coupling instabilities, as observed in electroactive elastomers, but rather from the loss of ellipticity associated with phase transitions.

In this contribution, we extend the recently developed thermodynamic regularization technique [2] to the context of strain-induced crystallization in natural rubbers and formulate a novel computational framework based exclusively on the laws of thermodynamics and a single thermodynamic potential. The predictions of the computational framework with the four coupled fields (displacement, temperature and two nonlocal interactions) is then demonstrated through multiple numerical benchmarks and comparison with experimental data. Specifically, the average nominal stress, calculated from the ratio of force and initial cross-sectional area, is demonstrated in Fig. 1 (left). In can be seen a good agreement with the experimental data and also there is no stress plateau observed after the onset of SIC due to regularization. The predicted heat source at the center of the specimen is plotted versus time in Fig. 1 (right). It can be observed that, initially, the heat source increases with crystallinity. Near the end of the loading phase, the heat source decreases due to the inflection point in the evolution of crystallinity.

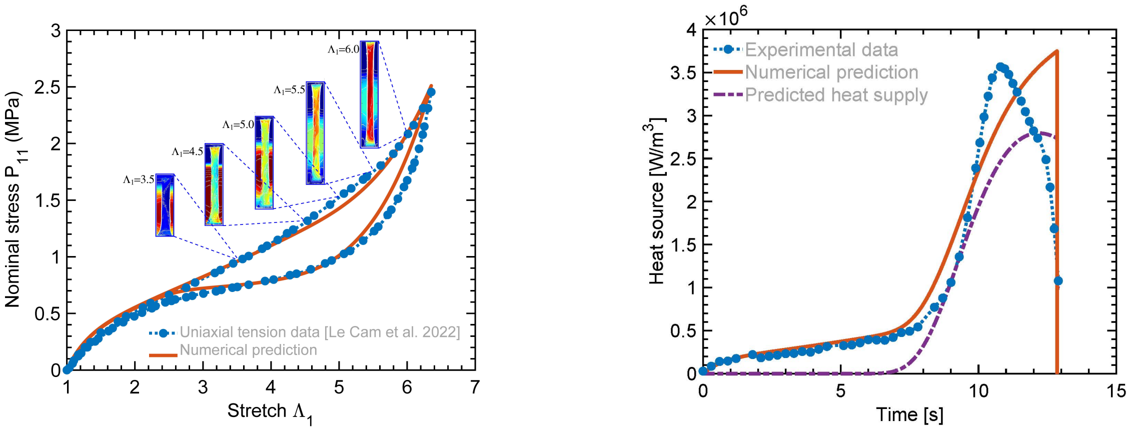

Figure 1: **Left**: Average nominal stress in the uniaxial tension; **Right**: Heat source evolution at the center of the specimen.

[1] J.B. Le Cam, A. Tayeb, S. Charlè, *Polymer* **255**, 125120, 2022. https://doi.org/10.1016/j.polymer.2022.125120

[2] V.N. Khiêm, M. Jabareen, R. Poudel, X. Tang, M. Itskov, *J. Mech. Phys. Solids* **193**, 105874, 2024. https://doi.org/10.1016/j.jmps.2024.105874 2024.