

REVEALING THE CORRELATION BETWEEN STRAIN-INDUCED CRYSTALLIZATION AND LOCAL STRAIN FIELD AROUND THE CRACK-TIP OF NATURAL RUBBER

Thanh-Tam Mai¹, Tomohiro Yasui², Ruito Tanaka², Hiroyasu Masunaga³, Taizo Kabe³, Katsuhiko Tsunoda⁴, Shinichi Sakurai² and Kenji Urayama¹

¹Kyoto University, Kyoto, Japan

²Kyoto Institute of Technology, Kyoto, Japan

³Spring-8, Hyogo, Japan

⁴Bridgestone Corporation, Tokyo, Japan

ABSTRACT

Natural rubber (NR)¹ exhibits strain-induced crystallization (SIC) which is an important feature setting NR apart from other elastomer types. SIC serves as a self-strengthening mechanism, significantly improving the mechanical strength and fracture toughness of the rubber². Specifically, the crystallization that occurs near the crack-tip increases crack-growth resistance which ultimately determines the lifespan of the elastomers³. Indeed, several studies⁴⁻⁷ revealed the occurrence of SIC near the crack-tip of NR using micro-beam wide-angle X-ray scattering (μ -WAXS). In general, the area near the crack-tip has an inhomogeneous strain distribution as characterized by digital image correlation (DIC) technique^{8,9}. However, the correlation between the SIC and strain field near the crack-tip has yet to be fully explored and comprehended¹⁻³.

In this study, we investigate the correlations between the degree and orientation of SIC, and the local strain field around the crack-tip in NR using a combination of μ -WAXS and μ -DIC techniques. We characterize the boundary of the SIC region and the spatial distributions of the crystallinity index and the c-axis of the SIC crystals. By relating the 2D strain field, we elucidate the strain criteria for the occurrence of SIC, and the relationship between the c-axis of SIC crystals and the principal axis of strain tensor.

ACKNOWLEDGEMENTS

This work was partly supported by JST, CREST (grant number JPMJCR2091), Japan.

REFERENCES

1. Huneau, B. Strain-Induced Crystallization of Natural Rubber: A Review of X-Ray Diffraction Investigations. *Rubber Chem. Technol.*, **84** (3), 425–452., 2011. <https://doi.org/10.5254/1.3601131>.
2. Persson, B. N. J.; Albohr, O.; Heinrich, G.; Ueba, H. Crack Propagation in Rubber-like Materials. *J. Phys. Condens. Matter*, **17** (44). 2005. <https://doi.org/10.1088/0953-8984/17/44/R01>.
3. Creton, C.; Ciccotti, M. Fracture and Adhesion of Soft Materials: A Review. *Reports Prog. Phys.*, **79** (4),

- 046601., 2016. <https://doi.org/10.1088/0034-4885/79/4/046601>.
4. Trabelsi, S.; Albouy, P. A.; Rault, J. Stress-Induced Crystallization around a Crack Tip in Natural Rubber. *Macromolecules*, **35** (27), 10054–10061., 2002. <https://doi.org/10.1021/ma021106c>.
 5. Brüning, K.; Schneider, K.; Roth, S. V.; Heinrich, G. Strain-Induced Crystallization around a Crack Tip in Natural Rubber under Dynamic Load. *Polym. (United Kingdom)*, **54** (22), 6200–6205., 2013. <https://doi.org/10.1016/j.polymer.2013.08.045>.
 6. Rublon, P.; Huneau, B.; Verron, E.; Saintier, N.; Beurrot, S.; Leygue, A.; Mocuta, C.; Thiaudière, D.; Berghezan, D. Multiaxial Deformation and Strain-Induced Crystallization around a Fatigue Crack in Natural Rubber. *Eng. Fract. Mech.*, **123**, 59–69., 2014. <https://doi.org/10.1016/j.engfracmech.2014.04.003>.
 7. Demassieux, Q.; Berghezan, D.; Creton, C. Microfocused Beam SAXS and WAXS Mapping at the Crack Tip and Fatigue Crack Propagation in Natural Rubber. *Adv. Polym. Sci.*, **286**, 467–491., 2021. https://doi.org/10.1007/12_2020_79.
 8. Osumi, R.; Yasui, T.; Tanaka, R.; Mai, T.-T.; Takagi, H.; Shimizu, N.; Tsunoda, K.; Sakurai, S.; Urayama, K. Impact of Strain-Induced Crystallization on Fast Crack Growth in Stretched Cis -1,4-Polyisoprene Rubber. *ACS Macro Lett.*, **11** (6), 747–752., 2022. <https://doi.org/10.1021/acsmacrolett.2c00241>.
 9. Schreier, H.; Orteu, J.-J.; Sutton, M. A. *Image Correlation for Shape, Motion and Deformation Measurements*; Springer US: Boston, MA, Vol. **54**, 2009. <https://doi.org/10.1007/978-0-387-78747-3>.