## **Abstract**

Replacing carbon black (CB) with silica in natural rubber (NR) has gathered tremendous attention in automotive industries. Incorporating high dosages of silica in NR by solid-state mixing is indeed a Herculean task. In green tyre technology, abrasion resistance index (ARI) is crucial alongside rolling resistance, wet grip, and heat build-up. A range of carbohydrates was incorporated during mastication as blocking agent to improve dispersion. Consequently, in this essence, sorbitol has remarkably improved the key properties. A significant improvement in tensile strength, reinforcement index, ARI (~9 %), HBU (heat build-up) (5 to 8 unit), and rolling resistance (8 to 9 % reduction) were evident in sorbitol treated NR compound, even at higher loadings of silica. On contrary, a bio-mimicking approach was adopted herein through incorporation of sorbitol into NR latex, creating protein-carbohydrate interaction. These had led to a significant improvement in key properties of the NR vulcanizates like, >15 % reduction in Tan  $\delta$  at high temperature, besides improved dispersion. The protein and phospholipid ends were cleaved by respective enzymes to understand and decouple the adverse effects owing to the respective constituents on dispersion. The molecular dynamics study reveals faster convergent of RMSD (root-mean-square deviation) and maximum Rg (Radius of Gyration) values of sorbitol treated latex, confirming a stable confirmation of NR matrix in presence of silica. Subsequently, greater number of hydrogen bonding between the functional groups of protein and sorbitol demonstrates successful blocking efficacy. Likewise, in case of synthetic isoprene rubber, the contribution of phospholipid in significant improvements in silica dispersion has been realized. This has been further tested and validated in DPNR based system. The presence of phospholipid made the compound persistent in terms of its overall performance, especially a significant improvement in Tan  $\delta$  at 0 °C than the other compounds. Consequently, >10 % reduction in rolling resistance (RR), 12-units' reduction in heat build-up (HBU), and >15 % reduction in RR, 24-units reduction in HBU were evident in dry and latex mixing composites, respectively utilizing the attributes of incorporation of prevulcanized gels. Furthermore, the drop in viscosity and diminution in die swell of the NR composites indicates a clear improvement in the processing behaviour of the gel embedded compounds. Overall, the scientific know how's derived from the thesis is envisaged to have a significant impact in sustainable and eco-friendly applications of silica in rubber based compounds for futuristic applications in tyres, energy-efficient conveying systems and to reduce carbon-footprints.

**Keywords:** Green tyre, Natural rubber, Silica, Dispersion, Magic Triangle, Carbon-foot print, Blocking agent, Carbohydrate, Pre-vulcanized gel, Energy efficient rubber, Sustainable.