

The molecular orientation induced in Styrene Butadiene Rubber (SBR) samples by stretching at room temperature (taken to be 22°C) as a function of filler (silica) and curing agent content (sulphur) has been studied. The molecular orientation was inferred from the orientation function which was determined with the use of infrared dichroism. Initially, the samples were characterized and the characterization tests included hardness, density, and tensile tests which yielded stress - strain curves and tensile strength. Stress relaxation experiments have in addition been carried out to determine the relaxation times ( $\tau$ ) which further characterizes the molecular dynamics of the variously treated SBR samples. The characterization results showed that hardness, density, tensile strength and stiffness increase with increase in filler content in the samples. A similar trend was noted for samples with different amount of curing agent though for density and tensile strength, there was increase up to a certain point before starting to decrease. The stress relaxation tests results for each sample appeared to be described by an exponential function suggesting that there is a tendency for the specimens to acquire an equilibrium state of stress after deformation. Relaxation time ( $\tau$ ) showed an increasing trend with increase in filler content up to a certain point before starting to decrease. The above observations can be attributed to the fact that increasing the concentration of a reinforcing filler such as silica up to a certain critical concentration to a vulcanisate is known to improve hardness, abrasion resistance, tensile strength, and to increase both stiffness and density of the vulcanisate. For samples with different amounts of curing agent, the relaxation time ( $\tau$ ) increases with increase in curing agent content up to a certain point then starts to decrease. This is due to the existence of a critical curing agent concentration above which its (curing agent) influence on ( $\tau$ ) starts to decrease. Higher sulphur contents beyond this critical concentration increases the degree of cross linking to levels in which molecular orientation of the material when in tension is substantially reduced. As a result, the material exhibits inferior tensile properties and has a short relaxation time.

The dependence of "orientation function" (hereby denoted by  $\langle P \rangle$ ) on extension shows a linear relationship for all samples. The slopes of these linear graphs (hereby referred to as Orientation coefficient and denoted by "C") are positive for all the samples indicating that  $\langle P \rangle$  increases with increase in extension. The value of "C" for all the samples decreases with increasing amount of filler and the curing agent content up to a certain point before starting to increase. This can be attributed to the fact that addition of a reinforcing filler such as silica up to a certain critical concentration to a vulcanisate increases the stiffness of the material. As a result, the molecular chains of the material are not easily oriented hence the reason for the decrease in  $\langle P \rangle$  with increase in the filler content. Beyond this critical concentration, the weak filler-filler interaction dominates the SBR composite hence the rate of chain orientation with extension (represented by "C") increases. Increase in the curing agent content up to a certain point in a material leads to increase in the degree of crosslinking. The crosslinks formed restricts the free mobility of the molecular chains. The higher the degree of crosslinking, the higher the restriction of mobility of these molecular chains and hence the reason for the decrease of  $\langle P \rangle$  with increase in the curing agent content up to this critical concentration. Above this concentration, the weak sulphur - sulphur interaction dominates the composite leading to the increase in the rate at which the chains orient themselves with extension hence increasing the value "C".