

## Physically Treated Cellulose and Its Use in Composites

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The present study investigates the influence of ultrasonic treatment on cellulose and its subsequent application as a filler in composite materials. The treated cellulose was initially characterised using Fourier transform infrared spectroscopy (FTIR) and thermal analysis to evaluate potential changes in chemical structure and thermal stability induced by ultrasound. Following this characterisation, the treated cellulose was incorporated into polymer composites. The resulting materials were analysed in terms of their rheological and mechanical properties.

Before the ultrasonic treatment, cellulose was pre-soaked in distilled water for 24 hours. The resulting cellulose suspensions, prepared at concentrations of 1 wt.% and 4 wt.%, were subjected to ultrasonication for 4 hours using a Bandelin Electronic DT 106 sonifier operating at a frequency of 35 kHz. Following sonication, the treated cellulose was separated by filtration and subsequently dried at 105 °C for 48 hours. Fig. 1 presents the sedimentation profiles of 1 wt.% and 4 wt.% cellulose suspensions, both untreated and ultrasonically treated, after 24 hours. In the untreated suspensions (Fig. 1a and 1c), visible phase separation and sediment formation are observed, indicating poor colloidal stability and aggregation of cellulose particles. In contrast, the ultrasonically treated suspensions (Fig. 1b and 1d) exhibit significantly enhanced dispersion stability, with a more homogeneous and turbid appearance and minimal sedimentation. The observed improvement in colloidal stability can be attributed to the ultrasonic treatment, which likely reduced the particle size and disrupted agglomerates through cavitation and shear forces generated during sonication.

Fig. 3 shows the FTIR spectra of the cellulose samples U1/Cellulose and U4/Cellulose. The spectra obtained demonstrate the presence of the characteristic absorption bands of cellulose. The predominant broad band in the 3333  $\text{cm}^{-1}$  region corresponds to the valence vibrations of the O-H groups. After ultrasonication, there is a decrease in the intensity of this band, indicating a decrease in the bound water content or a change in the arrangement of the hydrogen bonds. A similar trend can be observed for the 1637  $\text{cm}^{-1}$  band, which is related to flexural vibrations of bound water. Another band observed at 2892  $\text{cm}^{-1}$ , belonging to the valence vibrations of the C-H bonds, does not show significant shifts, but its intensity slightly decreases, which may be related to minor changes in the structure of the cellulose chain. In the 1428 and 1365  $\text{cm}^{-1}$  regions, which are related to the deformations of  $\text{CH}_2$  groups and C-H bonds, a decrease in intensities was observed, which may indicate disordered ordering in the crystalline regions. The most intense band in the 1027  $\text{cm}^{-1}$  region, attributed to the valence vibrations of the C-O-C bonds, remains pronounced in all samples, confirming the preservation of the basic structure of the polysaccharide chain. In contrast, the band at 896  $\text{cm}^{-1}$ , typical of amorphous cellulose, shows a decrease in intensity, especially for the U4/Cellulose sample. This phenomenon may be related to an increase in the proportion of crystalline regions after prolonged ultrasonic exposure. Fig. 4 shows the TG curves of untreated and ultrasonically treated cellulose samples, which reveal similar degradation profiles. This indicates that ultrasonic treatment does not significantly affect the thermal degradation behaviour of cellulose. The rheological and mechanical properties of rubber composites containing both untreated and ultrasonically treated cellulose are illustrated in Fig. 5-10. The torque-time curves demonstrate that the incorporation of cellulose (RC) results in an augmentation of torque in comparison to pure rubber (R), indicative of elevated viscosity and filler-matrix interaction. This effect is further enhanced in composites with ultrasonically treated cellulose (U1/RC and U4/RC), particularly for U4/RC, suggesting improved dispersion and interfacial compatibility due to ultrasound treatment. Tensile strength decreases significantly upon the addition of cellulose, with RC showing the lowest values. However, ultrasonic treatment slightly improves the tensile strength of U1/RC and U4/RC samples, although this remains lower than that of pure rubber. Similarly, elongation at break decreases in all cellulose-filled samples, indicating increased stiffness and reduced elasticity. All filled samples (RC, U1/RC and U4/RC) show higher Shore A values than R, confirming the reinforcing effect of cellulose and increased material rigidity. The swelling degree in toluene is highest for pure rubber and decreases with increasing cellulose content, being lowest for the U4/RC sample. This indicates reduced solvent uptake and enhanced network density. Ultrasonically treated cellulose at 1 wt.% (U1/RC) shows the highest crosslinking density, which indicates improved filler dispersion and interaction.

This study investigated the effect of ultrasonic physical treatment on the properties of cellulose and rubber composites containing it as a filler. Ultrasonic treatment of cellulose has been shown to enhance its compatibility with the rubber matrix, resulting in composites that exhibit higher hardness, reduced swelling, and marginally improved mechanical properties when compared to composites containing untreated cellulose.