Hemicelluloses represent a largely unutilized resource for future bioderived films in packaging and other applications. However, improvement of film properties is needed in order to transfer this potential into reality. In this context, sepiolite, a fibrous clay, was investigated as an additive to enhance the properties of rye flour arabinoxylan. Composite films cast from arabinoxylan solutions and sepiolite suspensions in water were transparent or semitransparent at additive loadings in the 2.5−10 wt % range. Scanning electron microscopy showed that the sepiolite was well dispersed in the arabinoxylan films and sepiolite fiber aggregation was not found. FT-IR spectroscopy provided some evidence for hydrogen bonding between sepiolite and arabinoxylan. Consistent with these findings, mechanical testing showed increases in film stiffness and strength with sepiolite addition and the effect of poly(ethylene glycol) methyl ether (mPEG) plasticizer addition. Incorporation of sepiolite did not significantly influence the thermal degradation or the gas barrier properties of arabinoxylan films, which is likely a consequence of sepiolite fiber morphology. In summary, sepiolite was shown to have potential as an additive to obtain stronger hemicellulose films although other approaches, possibly in combination with the use of sepiolite, would be needed if enhanced film barrier properties are required for specific applications.