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Rigidity percolation in dispersions with a structured viscoelastic matrix

E.D. Sourty\textsuperscript{1}, M.W.L. Wilbrink\textsuperscript{1}, W.P. Vellinga\textsuperscript{2}, P.J.G. Schreurs\textsuperscript{2}, M.A.J. Michels\textsuperscript{1} and H.E.H. Meijer\textsuperscript{2}

\textsuperscript{1} Department of Applied Physics, Eindhoven University of Technology
\textsuperscript{2} Materials Technology, Faculty of Mechanical Engineering, Eindhoven University of Technology

Introduction

Carbon-rich refinery residues mixed with mineral particles can lead to composite materials with bulk mechanical properties comparable to those of concrete. Understanding the microstructure mechanisms involved would set ways for improving existing and developing novel strong composites.

Material

Binder

The binder (bitumen) is regarded as a colloidal dispersion of agglomerated stacks of highly planar aromatic molecules—\textit{asphaltene}, peptized by the resin constituent of the surrounding \textit{maltene} phase (Fig. 1). Agglomeration may lead to self-association processes building up fractal structures. Depending on the aggregate compactness, binder may be of the sol-type or gel-type.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig1.png}
\caption{Schematic representation of the binder internal structure.}
\end{figure}

Composite

Adding mineral fillers (CaCO\textsubscript{3}; \(d=0.7\) μm) to the binder further confines the asphaltene aggregates (Fig. 2). The inter-particle distance \(\Lambda\) is critical.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig2.png}
\caption{Schematic representation of the composite internal structure}
\end{figure}

Experimental results

Fig. 3 shows the complex shear modulus \(G^\ast = G^' + iG^''\) measured by dynamic mechanical analysis (DMA) of composites with increased filler content \(\varphi\). A transition occurs for \(\varphi > \varphi_c = 0.09\) (inter-particle distance \(\Lambda_c = 0.6\) μm); \(G^\ast\) reaches a plateau \((G_{p}^\ast)\) at high temperature / low frequency (left side of the graph). Asphaltene aggregates may bridge the inter-particle ligaments: percolation of a stress-carrying network arises (Fig. 2, right). As the maltene phase becomes purely viscous, the percolating network takes on the mechanical behaviour of the composite.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig3.png}
\caption{Master curve for the complex shear modulus measured by dynamic mechanical analysis of composites with increased filler content.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig4.png}
\caption{Storage shear modulus at the plateau \(G_{p}^\ast\).}
\end{figure}

\(G_{p}^\ast\) obeys a scaling law: \(G_{p}^\ast \propto (\varphi_c - \varphi)^{\alpha}\) in agreement with percolation theory (Fig. 4).

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig4.png}
\caption{Storage shear modulus at the plateau \(G_{p}^\ast\).}
\end{figure}

Conclusion and further work

The unique mechanical behaviour of composites based on binder with internal self-organisation and colloidal structure (here bitumen) is attributed to the percolation of a stress-carrying network interlinking filler particle and agglomerated stacks of highly planar molecules (here asphaltene).

Ongoing work includes:
- Synthetic binders (dispersion of H-bonded C\textsubscript{3}-symmetrical discoid molecules) with comparable internal microstructure are now being investigated.
- Concurrently, numerical simulation using finite element method is being implemented.