Rigidity percolation in dispersions with a structured viscoelastic matrix

Published: 01/01/2002

Document Version
Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:
• A submitted manuscript is the author's version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
• The final author version and the galley proof are versions of the publication after peer review.
• The final published version features the final layout of the paper including the volume, issue and page numbers.

Link to publication

Citation for published version (APA):

General rights
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.
• Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
• You may not further distribute the material or use it for any profit-making activity or commercial gain
• You may freely distribute the URL identifying the publication in the public portal ?

Take down policy
If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.
Rigidity percolation in dispersions with a structured viscoelastic matrix

E.D. Sourty¹, M.W.L. Wilbrink¹, W.P. Vellinga², P.J.G. Schreurs², M.A.J. Michels¹ and H.E.H. Meijer²

¹ Department of Applied Physics, Eindhoven University of Technology
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—asphaltene, peptized by the resin constituent of the surrounding 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.

Fig. 1. Schematic representation of the binder internal structure.

Composite
Adding mineral fillers (CaCO₃; \( d = 0.7 \mu m \)) to the binder further confines the asphaltene aggregates (Fig. 2). The inter-particle distance \( \Lambda \) is critical.

Fig. 2. Schematic representation of the composite internal structure

Experimental results
Fig. 3 shows the complex shear modulus \( G^* = G' + iG'' \) measured by dynamic mechanical analysis (DMA) of composites with increased filler content \( \phi \). A transition occurs for \( \phi = \phi_c = 0.09 \) (inter-particle distance \( \Lambda_c \approx 0.6 \mu m \)); \( G^* \) reaches a plateau (\( G'_p \)) 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.

Fig. 3. Master curve for the complex shear modulus measured by dynamic mechanical analysis of composites with increased filler content.

\[
G'_p \propto (\phi - \phi_c)\alpha
\]
\[\alpha = 3.75; \phi_c = 0.09\]

Fig. 4. Storage shear modulus at the plateau \( G'_p \).
\( G'_p \) obeys a scaling law: \( G'_p \propto (\phi - \phi_c)^\alpha \) in agreement with percolation theory (Fig. 4).

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 C₃-symmetrical discoid molecules) with comparable internal microstructure are now being investigated.
- Concurrently, numerical simulation using finite element method is being implemented.