Rigidity percolation in dispersions with a structured viscoelastic matrix

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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.

Composite
Adding mineral fillers (CaCO₃: d=0.7 μm) to the binder further confines the asphaltene aggregates (Fig. 2). The inter-particle distance Λ is critical.

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 $\Lambda_c ≈ 0.6 \mu$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.

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.