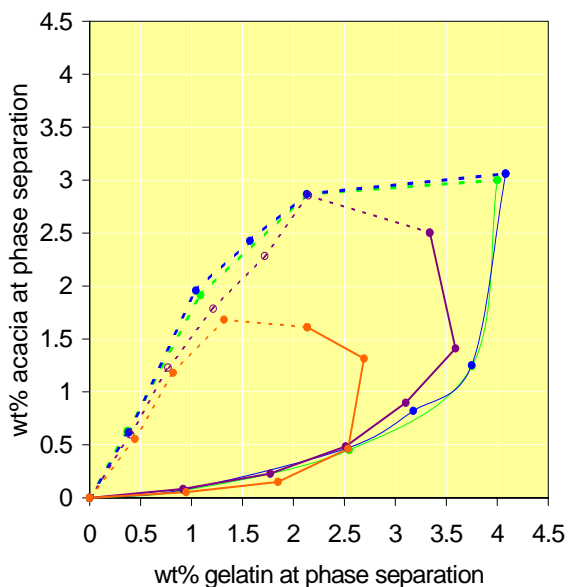




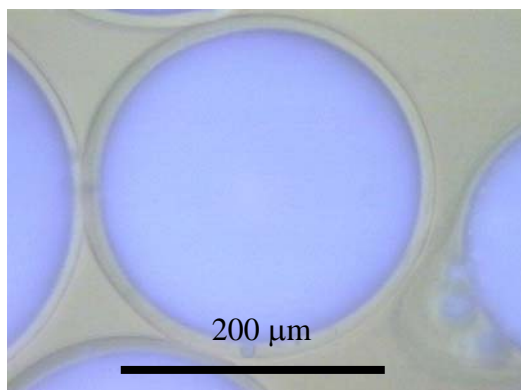
Microencapsulated Non-aqueous Dispersions

Droplets of oil containing finely dispersed nanoparticles can be encapsulated in natural polymers by leveraging three interesting physical chemical properties of certain polyelectrolytes. Firstly aqueous solutions of gelatin form gels on cooling below $\sim 40^{\circ}\text{C}$. Second, gelatin molecules are amphoteric, i.e. negatively charged about $\text{pH} > \sim 5$ and positively charged at $\text{pH} < \sim 4$. And lastly, under certain conditions, two oppositely charged polymers dissolved in water can associate by charge neutralization to form fluid

droplets of a polymer-rich phase called a coacervate. Gelatin and acacia gum mixed in dilute solution at $\text{pH} > \sim 5$ forms a clear single-phase solution. If the pH is lowered below ~ 4 , the gelatin molecules become positively charged, and associate with the negatively charged acacia gum molecules to form droplets of the separate coacervate phase containing the hydrated complex. The phase diagram in figure 1 shows the concentration boundary lines within which the coacervate phase forms from the oppositely charged polymers for four different batches of gelatin (showing the variability in the behavior of different natural materials). The coacervate phase appears for compositions within the boundary lines, while for compositions outside the boundary lines the mixture gelatin / acacia / water remains a single phase. It is of interest to note that the coacervate phase only forms from very dilute solutions ($< 4\%$ w/w) of each polymer, typical of coacervating mixtures.



The liquid coacervate droplets are a few microns in diameter and are clearly visible in an optical microscope. The coacervate droplets are surface active and can engulf particles or droplets of another material that is co-dispersed in the water phase. This is shown clearly in figure 2 where droplets of an oil-based ink formulation comprising a blue dye and white nanoparticles have been engulfed by enough coacervate material that a “green” microcapsule has been formed – both core and shell at this point are liquid. Careful formulation of the oil-based ink, and controlled emulsification into the initial solution of gelatin and acacia gum at pH > 5, ensured that the nanoparticles remained well-dispersed within the oil droplets, that the nanoparticles remained there during the process and did not migrate into the water phase, and prevented water from becoming entrained inside the oil droplets. In this example the core droplet (~200 μm) is much larger than the coacervate droplets that have coated it; droplets several millimeters in diameter can be coated with coacervate to form very large microcapsules. Otherwise droplets of core material can be as small as a few microns or less.



The green microcapsules are made more robust for handling and use by hardening the shell. This leverages the gelling properties of gelatin; the dispersion of the oil droplets and deposition of coacervate was carried out at 40°C. The batch was then cooled to 5°C to gel the walls, and a chemical hardener was added to chemically crosslink the gel.

This type of microcapsule could be used for a variety of purposes – if the nanoparticles dispersed in the oil-phase are paramagnetic, then administered capsules can be traced or concentrated in the body by magnetic means. This could have application in medical imaging or site-specific delivery of a drug. Alternatively, the microcapsules could be used to mask the flavor of unpleasant oily medicines (such as cod-liver oil).



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