Experimental protocol for the formation of four different sizes of spherical microdroplets

A microfluidic device composed of polymethylesiloxane (PDMS)-Teflon capillary was used to form microdroplets as shown in Fig. S1. Four different capillary internal diameters (130, 200, 360 and 500 μm) were used to generate four kinds of spherical droplet sizes. The same size of microchannel PDMS chip was used in all experiments. Protein solution (20 mg/mL thaumatin in aqueous solution of 100 mM *N*-(2-acetamido) iminodiacetic acid (ADA) buffer (pH 6.5)), precipitant solution (1.6 M potassium sodium tartrate in aqueous solution of 50 mM 4-(2-hydroxyethyl)-1-piperazineethanesulfonic acid (HEPES) buffer (pH 7.0)), and fluorinated oil (a mixture of 3M™ Fluorinert™ Electronic Liquid FC-40 and 1*H*, 1*H*,

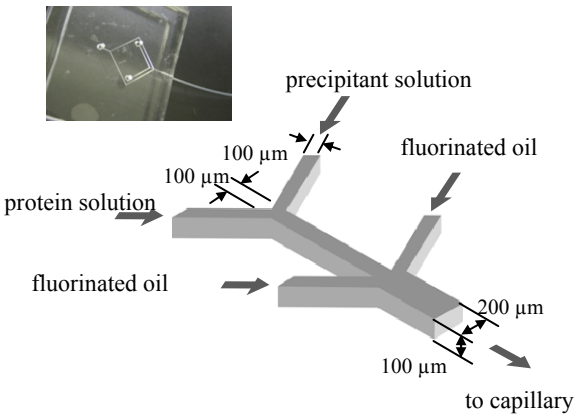

Fig. S1. A photograph of the composite PDMS-Teflon capillary microfluidic device, and a schematic illustration of the channel size and the method of forming microdroplets.

2*H*, 2*H*-Perfluoro-1-octanol (10:1, v/v)) were loaded as shown in Fig. 1. These concentrations were adjusted so as to get the suitable crystallization time both for microdroplet and batchwise process. Protein solution and precipitant solution joined together and form a single droplet in a microchannel. Protein solution and precipitant solution were loaded at the same flow rate, and two fluorinated oil injections were also pumped at the same flow rate. Each flow rate was finely adjusted to obtain spherical microdroplets.