Title:	Ordered Water within the Collapsed Globules of an Amyloidogenic Intrinsically Disordered Protein
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Abstract:	Intrinsically disordered proteins (IDPs) confront the traditional sequence-structure-function paradigm and are associated with important functions and amyloid disorders. Water molecules residing in the vicinity of the polypeptide chain play potentially important roles in directing the course of binding-induced folding and amyloid aggregation of IDP. Here we characterized the nature of water molecules entrapped within the collapsed globules of an amyloidogenic IDP, namely, κ-casein. These globules can undergo further compaction in the presence of an anionic detergent that is capable of diminishing the intrachain repulsion from the positively charged glutamine/asparagine-rich amyloidogenic N-terminal domain comprising 100 residues. Using time-resolved fluorescence spectroscopy, we estimated the longer component of the solvation time to be ∼1.4 ns, which is 3 orders of magnitude slower than that in bulk water and more than an order of magnitude slower than the "biological water" present at the protein surface. Profoundly restrained water within the collapsed IDP globules resembles the ordered water cluster found under nanoconfinement. We suggest that the association of these globules would result in the release of ordered water molecules into the bulk milieu causing an entropic gain that would eventually drive the formation of the key (obligatory) oligomeric intermediates on the pathway to amyloids via nucleation-dependent polymerization.
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