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This manuscript is concerned with expanding the dendritic structural repertoire, through covalent assembly and disassembly in aqueous solutions on poly(ether imine) dendritic macromolecules. The importance of multiple branching through-out the structure to amplify the so-called endo- and exo-receptor properties of dendritic macromolecules has been verified in a number of instances, with special emphasis on larger generations. Multiple branching throughout the structure being an important structural feature of this class of macromolecules, we undertook to expand the structural repertoire to dendritic megamer macromolecules, in aqueous solutions. We further designed the megamers to be reversible, such that a redox stimulus aids to disassemble the megamers to the constituent dendrimer monomer, in aqueous solutions. The kinetics of monomer-megamer assembly was investigated with the aid of light scattering the microscopic techniques. The aggregation behavior of soluble and insoluble components of dendrimer megamer was investigated. Following studies of the assembly to megamers, the disassembly to dendrimer monomer was conducted. The strategy of dendrimer monomer-megamer assembly-disassembly in aqueous solution was further studied towards guest uptake properties. Specifically, water insoluble sudan I was utilized to assess the effect of dendrimer megamer to encapsulate the dye, followed by the release upon disassembly of the megamer to constituent dendrimer monomer. Thus, the work is taken through a planned synthesis, studies of the kinetics of monomer-megamer assembly-disassembly and the endo-receptor properties of the megamer. The studies are performed with three generations of water soluble poly(ether imine) dendrimers, thereby allowing a comparison of dendrimer monomer-megamer assembly-disassembly processes.