



Special issue dedicated to the memory of Prof Masayoshi Okubo

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Received: 2 February 2022 / Accepted: 3 February 2022 / Published online: 22 February 2022
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Prof Masayoshi Okubo



Prof Masayoshi Okubo passed away on February 20, 2021, in Kobe, Japan, due to lung cancer. He was born in Nishinomiya, Hyogo Prefecture, Japan, on Feb 16, 1947. He completed his undergraduate studies in the Department of Industrial Chemistry at Kobe University, graduating in 1969, and subsequently completed his master's degree from the same university in 1971. He was appointed Research Associate at Kobe University in the laboratory of Prof Matsumoto in 1971. He submitted his thesis and obtained the degree of Doctor of Engineering at Kyoto University in 1976 on the topic of “Studies on polymer emulsion having electrical groups.” After this, he spent 1 year as a postdoctoral research fellow at the Fritz Haber Institute of the Max Planck Society (Germany) working on the topic of the surface tension of polymer solutions. He returned to his position at Kobe University, where he was promoted to Associate Professor

in 1985 and Professor in 1995. Later on in his career, Prof Okubo spent 2 months at the Max Planck Institute of Colloids and Interfaces in 2006, working with Dr Klaus Tauer. He officially retired from his position at Kobe University in 2010, and was appointed Emeritus Professor. Prof Okubo remained active, and took up a position as Distinguished Full Professor at Nanjing Tech University in China from 2016 until 2018. He also established a close research collaboration with Rajamangala University of Technology Thanyaburi in Thailand, where he was appointed Adjunct Professor in 2012.

During his career, Prof Okubo supervised more than 120 graduate students and published 397 peer-reviewed research papers and 50 patents. Prof Okubo has also served as Vice President for the Adhesion Society of Japan and the Society of Polymer Science, Japan. He made enormous contributions to the field of polymer colloids, in particular in regard to design and synthesis of polymer particles of specific and complex morphologies. He received several awards for his work, including the ASJ Award for Outstanding Achievement in Adhesion Science and Technology (The Adhesion Society of Japan) and the SPSJ Award for Outstanding Achievement in Polymer Science and Technology (The Society of Polymer Science, Japan), among many others.

Prof Okubo was a very active member of the IPCG (International Polymer Colloids Group) community—he attended most IPCG conferences between 1989 and 2019. Many of you will have known Prof Okubo for many years—he was an enthusiastic supporter and contributor to the IPCG community, and he will be sorely missed. He was well-known for his extremely clear and informative presentations that were further enhanced by a great sense of humor. He has been an inspiration for younger researchers, and has made a very significant mark in the field of polymer colloids.

Prof Okubo was very passionate about his research—his last paper was accepted for publication in *Langmuir* only four days before his passing on his 74th birthday, and he engaged in discussions to finalize this paper via video chat from his hospital bed.

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Prof Okubo is survived by his wife Tsuneko and their two children and four grandchildren.

This special issue

This is a collection of original papers organized to commemorate the contributions to the field of polymer colloids by Prof Okubo. The field of polymer colloids has existed since the early days of development of emulsion polymerization, and has seen rapid expansion over the decades, significantly so since the advent of reversible deactivation radical polymerization (RDRP) now some 25 years ago. The present collection of papers from a variety of authors in the general area of polymer colloids reflects the diversity and vibrance of the field today.

Several papers focus on various aspects/applications of conventional emulsion polymerization. Lovell and co-workers used ^{13}C NMR spectroscopy to investigate the extent of chain transfer to polymer and branch formation for a series of alkyl acrylates in monomer-starved semi-batch emulsion homopolymerization. The understanding developed is of high relevance for optimization of waterborne pressure-sensitive adhesives. Nagao and co-workers report on the synthesis of monodisperse polymer nanoparticles of diameters less than 50 nm by emulsion polymerization. The key point in their approach to obtain such small particles is the use of a surfactant with a CMC significantly lower than that of a common surfactant. The mechanistic reasons are discussed in terms of radical exit and micelle disappearance in the early stage of the polymerization process. Leiza and co-workers developed various strategies to incorporate the fluorinated monomer perfluorooctyl acrylate (POA) into a waterborne polymeric dispersion, the challenge being the very low water solubility of POA. The approach yielding core-shell particles with the core composed of polyPOA resulted in films with the best anti-corrosion properties. Cunningham and co-workers used emulsion polymerization of methyl methacrylate (MMA) and a small amount of *N,N*-dimethylaminoethyl methacrylate (DMAEMA) with the initiator 2,2'-azobis[2-(2-imidazolin-2-yl)propane]dihydrochloride (VA-044) to synthesize CO_2 -switchable latexes. These latexes exhibited excellent responsive CO_2 behavior featuring rapid aggregation and with complete recovery of particle size on redispersion without requiring high energy mixing.

Ma and Gong and co-workers explored the effect of pore activation on emulsion performance and emulsification efficiency in microporous membrane emulsification. Overall, a set of conditions were identified for production of uniform droplet formation, namely large interfacial tension between the membrane pore outflow and the dispersed phase, high pore activation speed, and large spacing between the mem-

brane pores. Zetterlund and co-workers also report on membrane emulsification in conjunction with miniemulsion polymerization for monomers with varying water solubilities. This technique is a convenient approach for synthesis of well-defined polymer nanoparticles of various monomers in the approximate size range 200–3500 nm.

Other papers described synthesis of polymer particles of more complex morphologies and functionalities. Van Herk and co-workers synthesized polymer-vesicle latex particles using a templated approach based on ATRP and vesicles formed by dimethyldioctadecyl ammonium bromide. Structural features such as nanocapsules and protrusions formed depending on the monomer composition employed in the process. Minami and co-workers prepared elastic/glassy Janus composite particles with snowman-like morphology by seeded polymerization of benzyl methacrylate (BzMA) in the presence of cross-linked silicone particles. The particles featured interesting mechanical properties in the form of a combination of rubber elasticity derived from the silicone/PBzMA sea-island structure and hardness derived from the PBzMA lobe. Hawkett and Nguyen and co-workers also focused on synthesis of Janus particles, employing a one-step batch process using amphiphilic macro-RAFT copolymers as stabilizers followed by use of the resulting polystyrene (PS)-based particles as seeds in a continuous radical emulsion polymerization process of MMA and butyl acrylate. The obtained latex was successfully employed to disperse and encapsulate solid calcite extender. Taniguchi and co-workers synthesized luminescent core-shell polymer particles carrying amino groups for covalent immobilization of enzymes for applications in immunoassays. PS core particles were prepared by miniemulsion polymerization of styrene droplets containing 2-(2-chloropropionyl)ethyl methacrylate and a europium complex. The red luminescence attributed to europium complexes embedded in the core particles was observed upon UV irradiation. Fujii and co-workers report on synthesis of PS particle-stabilized Pickering-type paraffin droplets coated by polypyrrole overlayers using aqueous chemical oxidative polymerization of pyrrole. The thickness of the polypyrrole layer could be adjusted by changing the pyrrole concentration. This work has implications in regard to development of novel material delivery systems that can be controlled by various stimuli. Suzuki and co-workers examined synthesis of solid core-hydrogel shell particles using seeded precipitation polymerization. The focus was on how to control the shell structure via parameters such as the monomer and cross-linker concentrations.

Several papers concern more specific applications of polymer colloids. Bohórquez and Asua conducted a modelling study to increase the understanding of the rheological behavior of polymer latexes, considering how the parameters of particle volume fraction, particle size distribution, and shear rate affect the viscosity of waterborne dispersions stabilized

with conventional nonionic surfactants and polymeric stabilizers. The developed predictive capability has important implications in regard to development of latexes with high solids content and low viscosity. Li and Guo and co-workers report on the fabrication of macro-porous polymer foams featuring thermo-active shape memory employing bio-based poly(lactic acid) (PLA) macro-cross-linker using water-in-oil high internal phase emulsion (HIPE) polymerization. Horák and co-workers synthesized block copolymer consisting of the hydrophilic poly[*N*-(2-hydroxypropyl) methacrylamide] (PHP) and reactive poly[*N*-(2-hydrazinyl-2-oxoethyl) methacrylamide] (PMAH) using RAFT polymerization. Conjugation with doxorubicin (Dox) and/or RGDS targeting peptide and subsequent coating of magnetic γ -Fe₂O₃ nanoparticles afforded hybrid nanoparticles with two targeting features with great potential for targeted drug delivery. Chaiyasat and co-workers developed an antimicrobial spray

for coating of materials based on polymer particles containing quaternary ammonium (QA) and benzophenone (BP) groups on their surfaces using a synthetic strategy involving iodine transfer polymerization. Ahmad and co-workers report on synthesis and application of amine functionalized silica (NH₂-SiO₂)-supported Cu-Ni nanocatalyst. Relative to unsupported Cu-Ni bimetal nanoparticles, the supported NH₂-SiO₂/Cu-Ni nanocatalyst exhibited improved catalytic activity.

We hope that the collection of articles in this special issue shall serve as both a fitting homage to Prof Okubo and his career, as well as stimulus for further research in the field to which Prof Okubo contributed so much for several decades.

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