The Fourth Green and Sustainable Chemistry Award
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Innovation in Environmentally Benign Organic and Inorganic material Production Using Supercritical Fluids

Recently the matter of primary interest in the field of organic synthesis has been greatly aroused on the importance of efficiency as a practical/economic consideration via "greener" chemistry. Utilization of nontoxic and easily renewable chemicals or catalysts under environmentally friendly conditions is a key aspect that promotes a green synthetic strategy. Furthermore, great concern for green synthesis is the choice of the simplified process. The majority of methods so far used, however, are consisted of several acid-, base-catalyzed, and radical reaction steps using different organometallic compounds and solvents, resulting in potential environmental and biological risks besides the marked decrease in yield.

In order to erase these environmental damaging anxiety, supercritical fluids such as supercritical water (scH_2O) and carbon dioxide ($scCO_2$) should be useful and suitable replacement for organic solvents. ScH_2O and $scCO_2$ are employed for the first time as catalyst as well as medium for organic synthetic reactions and furthermore a supercritical fluid hybrid reaction system incorporated with a microreactor, ionic liquid, or microemulsion, greatly improves chemical reactivities of several reactions.

They found that scH_2O itself possesses Lewis acidity or basicity, besides the Brϕnsted acid-base function. Water is the most environmentally acceptable and inexpensive solvent, and its physicochemical properties can be changed widely in addition to density. Hence our new scH_2O hybrid reaction system incorporated with a microreactor not only can realize one-step manufacture of chemical valuables such as ε -caprolactam and monoterpene alcohols in high yield and high selectivity, even without catalyst, but can be useful for a wide range of acid-catalyzed organic syntheses, thus rendering newly potential applications to environmentally friendly and economically favorable synthesis of various fine chemicals.

Besides, the utilization of carbon dioxide was developed not only as a reaction medium but also a carbon resource by using a $scCO_2$ hybrid reaction system. Especially, "$scCO_2$-ionic liquid hybrid reaction system" for the CO_2 fixation was investigated, and successfully achieved the extremely rapid CO_2 fixation to produce alkylene carbonates and cyclic urethanes with high yields and high selectivities.

Furthermore, as another $scCO_2$ hybrid reaction system, the commercially viable water-in-$scCO_2$ reverse microemulsion system can be invented for the industrial scale synthesis of various quantum size metal nanocrystals. This system can be used as the "nano-reactor system", and the Ag_2S nanocrystals with a mean diameter of 5.9 nm were successfully synthesized in this system.

They have substantiated the first comprehensive method suitable for green chemistry concepts in the synthesis of various kinds of organic or inorganic compounds using supercritical fluids. This new synthetic technology not only can realize the total elimination or the significant minimization of hazards, environment impacts, and generated waste,but implicit in this environmentally responsible concept is a new way of thinking about chemical synthesis and processing, leading to "a dream mass production process" toward the next generation.