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Editorial

Theme Issue in Memory to Professor Jiro Tsuji (1927–2022)

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The importance of catalysis is obvious and unquestionable, especially bearing in mind that about 90% of all commercially produced chemical products involve catalysts at some step of their manufacture [1]. The history of the catalysis study has been presented in many fruitful review papers [1–7]. Considering the keywords "catalysis" and "history", more than one thousand papers could be found (e.g., 1300 in the Web of Science on 16 April 2024). Interestingly, the special aspects on the development of catalysis research have been presented in respect not only to the different research fields of catalysis (e.g., organotextile catalysis [8], heterogeneous catalysis [9,10], homogeneous catalysis [11,12], electrocatalysis [13], single-atom catalysis [14], phase transfer [15], solid hydrogen storage [16], plasma catalysis [17], catalytic combustion [18], minerals [19], small molecules [20], transition metals [21], computational study/machine learning [22,23], enzyme [24], polymerization [25], water oxidation [26], photoredox catalysis [27,28], photocatalysis [29], artificial photosynthesis [30], fluid catalytic cracking [31], gasoline automobile catalysis [32,33], nanozymes [34], "the origin of life" [35], applied/industrial catalysis [1,36,37], etc.) and particular catalysts (e.g., Ziegler–Natta catalysts [38]), but also to the specific countries (e.g., Korea [39], Brazil [40–42], Portugal [43], Italy [44], and India [45]), companies (e.g., Ciba–Geigy [46], Roche [47], CF-industries [48], Dow Chemical Company [49], Eastman Chemical Company [50], Leuna Factory [51], and Volkswagen [52]), world regions (Latin America [53]), institutes and consortiums (e.g., Dalian Institute of Chemical Physics [54] and CIRCC in Italy [55]), catalysis societies and their meetings (e.g., The Catalysis Society of Japan (CATSJ) [56], The Swiss Industrial Biocatalysis Consortium (SIBC) [57], and "50 Years of German Catalysis Meetings" [58]), and even the collaboration and friendships between countries (Japan–China [59] and France–Venezuela [60]), etc.

Among various papers, the development of catalysis research in the field of organic chemistry has been well documented [61], as shown by a significant increase in the number of published papers (Figure 1). Many different aspects of organic chemistry have been discussed, including single-atom catalysis [62], hydrogenation [63], C-H functionalization [64], enantioselective catalysis [65], methylation [66], nucleophilic phosphinocatalysis [67], chemistry of alkynes [12], isocyanide-based multicomponent reactions [28], aldol reaction [68], Fischer–Tropsch (FT) synthesis [69,70] (and even the reactors [71]), microwave-assisted synthesis [42], Diels–Alder reaction [72], Suzuki–Miyaura cross-couplings [73], synthesis of nitrile [74], formose reaction [75], π-allylpalladium chemistry [76], palladium-based catalysts [77–80], and nickel catalysts [79].

Obviously the studies on catalysis by famous scientists, including the works by Lavlavel G. lonescu [81], Nikolai Zelinsky [82,83], Heinz Heinemann [84], Paul Sabatier [85], Jean Baptiste Senderens [85], Mikhail Kucherov [86], William Crowell Bray [87], Slobodan Anic [88], Luigi Casale [89], and Gilbert Stork [90,91], have also been acknowledged.

This Special Issue of *Catalysts* is dedicated to Professor Jiro Tsuji, who passed away on 1 April 2022. Jiro Tsuji was born on 11 May 1927, in Shiga, Japan. He received a Bachelor of Science degree from Kyoto University, Japan, in 1951, a M.Sc. degree from Baylor University, USA, in 1957, and a Ph.D. from Columbia University, USA, in 1960 under the supervision

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of Gilbert Stork. Tsuji returned to Japan in 1962 to work as a research associate at Toray Industries, where he began his career in transition metal-catalyzed reactions. In 1974, he moved to the Tokyo Institute of Technology and served as a professor until retirement age of 60 in 1988. Thereafter, he was a professor at Okayama University of Science, Japan, from 1988 to 1996, and then at Kurashiki University of Science and the Arts from 1996 to 1999.

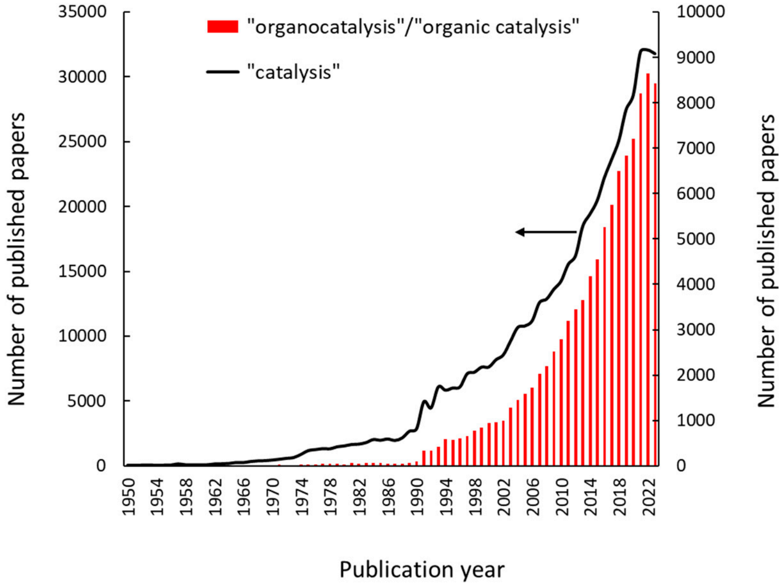

Figure 1. Number of papers published annually on catalysis and organocatalysis (searched in the Web of Science using "catalysis" and "organocatalysis"/"organic catalysis", 27 March 2024).

Professor Jiro Tsuji pioneered the discovery of many transition metal-catalyzed reactions and showed a general idea of developing and applying these reactions in organic synthesis. Among the well-known reactions are several types of Pd-catalyzed ones, such as (i) the substitution of allylic substrates based on the reaction of π-allyl palladium with carbon nucleophiles, discovered in 1965; (ii) reactions of allyl β-keto esters, resulting in allylation, olefin formation, and reduction; (iii) reactions of propargylic substrates; and (iv) the formation of methyl ketones from 1-olefins, based on the Wacker process of ethylene. It is noteworthy that the olefin formation is used as the key step in the industrial synthesis of jasmonate [92]. Other reactions catalyzed by Pd, Ru, and Cu are carbonylation of olefins, dienes, acetylenes, and allyl compounds; decarbonylation of acid chloride and aldehydes; oxidative decomposition of catechol to muconic acid; etc.

Professor Jiro Tsuji focused on the carbon–carbon bond-forming reaction from the very beginning of his research. According to Tsuji, the importance of organic synthesis was taught by professor Gilbert Stork (who he acknowledged in two review papers [90,91]) when he was in the doctoral program at Columbia University.

The significance of the reactions found by Tsuji has been proven by their widespread adoption in academic and industrial laboratories. Consequently, it is not surprising that Tsuji has been honored with the Chemical Society of Japan Award in 1981, the Japanese Medal of Honor with Purple Ribbon in 1994, the Japan Academy Prize in 2004, and the Tetrahedron Prize in 2014. He received the title of honorary professor at Tokyo Institute of Technology on 21 July 2011.

It must be underlined that two reactions were named after him, i.e., the Tsuji–Trost reaction and the Tsuji–Wilkinson decarbonylation reaction. The former (also called the Trost allylic alkylation or allylic alkylation) is a substitution reaction with a palladium catalyst, involving a substrate that contains a leaving group in an allylic position. This work (already mentioned above) was first pioneered by professor Tsuji in 1965 [93] and, then, continued by professor Barry Trost in 1973 with the introduction of phosphine ligands [94]. The latter is a method for the decarbonylation of aldehydes and some acyl chlorides. The name has recognized professor Tsuji, whose team reported as first the use of $RhCl(PPh_3)_3$) catalyst (Wilkinson) for these reactions.

The scientific achievements of professor Jiro Tsuji have resulted in more than 250 scientific papers, cited in more than 8,000 works, and reaching almost 13,000 citations (according to the Web of Science, on 23 April 2024). Obviously, the papers with the highest interests of the scientific community present palladium catalysts in organic synthesis [95–101]. However, also other catalysts have been successfully applied, and the data reported have been highly cited, including osmium tetroxide [102], metal complexes (e.g., rhodium, ruthenium, molybdenum, nickel, palladium, cuprous, titanium, and aluminum [103–108]), and other metals (copper [109,110], bismuth [111], and iron [110]).

This Special Issue of *Catalysts*, in memory of prof. Jiro Tsuji, was announced to acknowledge the magnificent impact of his study on others. In total, eleven papers were published, including three reviews, six research papers, and two communications. It must be pointed out that six of them have been selected as feature papers. The review papers focus on (i) the construction of structurally intriguing π-extended polycyclic heteroaromatics through catalytic coupling reactions [112]; (ii) the coupling reactions using organoborates/Ni and RMgX/Cu reagents [113]; and (iii) the transition metal-catalyzed ring-closing metathesis and coupling reactions [114].

The communications by Sieger et al. and Cusumano et al. present their findings on the Rh-catalyzed addition reaction of nitrogen-containing heterocycles to internal allenes [115] and the origins of enantioselectivity in the Pd-catalyzed decarboxylative allylic alkylation of N-benzoyl lactams [116], respectively.

Among research papers, besides classical organic synthesis approaches, other so-called "green" methods have also been discussed, i.e., using photocatalysis and electrochemistry. The photocatalytic formation of (2S,3S)-S-[(Z)-aminovinyl]-3-methyl-D-cysteine (AviMeCys) has been proposed by Kumashiro et al. [117]. In the case of electrochemistry, oxidative cyclization of ortho-vinyl aniline has been proposed by Hu et al. [118].

The favorite topic of professor Tsuji, i.e., palladium catalysis, has been discussed by Bao et al. [119], presenting a three-component cross-coupling reaction of 2-(trimethylsilyl)phenyl trifluoromethanesulfonate, benzylic/allylic bromides, and 1,1-bis[(pinacolato)boryl]methane. An interesting approach has been proposed by Ito et al. for the preparation of polysubstituted 3-hydroxypyridines from amino acids, propargyl alcohols, and arylboronic acids [120]. Ostrowska et al. proposed the synthetic protocol for palladacycle complexes using a mild base and an environmentally desirable solvent [121]. Finally, the substitution of secondary propargylic phosphates has been carried out by Kobayashi et al. with the use of aryl-lithium-based copper reagents [122].

To conclude, the significant contribution of professor Jiro Tsuji in the fields of organic chemistry and catalysis is unquestionable. It is thought that the research started by Tsuji will inspire others further in the development of new, environmentally friendly synthesis methods for world sustainability.

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