

March 26-28, 2018
Vienna, Austria

Polym Sci, Volume 4
DOI: 10.4172/2471-9935-C1-009

MECHANISTIC STUDIES OF TRANSITION METAL CATALYZED RADICAL TERMINATION

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Transition metal complexes have proven their utility in macromolecular engineering by catalyzing radical polymerization by atom transfer (ATRP) and also by serving as moderating agents via direct trapping of the growing radical chain to generate organometallic dormant species (OMRP). Certain Cu^I/L and Fe^{II}/L complexes with high ATRP activity have recently been shown to also catalyze radical termination (CRT). The initial interpretation based on work using Cu^I/L has attributed this phenomenon to the generation of hydride intermediates, H-Cu^{II}/L, by β -H atom transfer or β -H atom elimination from the organometallic dormant species, R-Cu^{II}/L. However, more recent

mechanistic investigations with L from a family of modified tris(pyridylmethyl)amines (TPMA) with growing donor power, have supported the idea that R-Cu^{II}/L is the key CRT intermediate, leading to the termination products upon interaction with a second radical chain and without transit through the hydride H-Cu^{II}/L. The intimate mechanism of the R-Cu^{II}/L + R reaction to regenerated Cu^I/L plus termination products, however, is elusive. This presentation will highlight recent experimental and computational investigations that bring light into this matter.

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