### Combining Cationic and Anionic Polymerizations using a Universal Mediator: Enabling Access to Two- and Three-Mechanism Block Copolymers

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ABSTRACT: The combination of multiple polymerization mechanisms and monomer classes to make block copolymers is an ongoing challenge. In particular, the combination of cationic and anionic polymerization mechanisms commonly requires extra compatibilization steps or the use of multi-functional initiators. Herein, we report the use of thiocarbonyl thio compounds (TCTs) as universal mediators to sequentially polymerize ethyl vinyl ether via photocontrolled cationic polymerization (photo-CP) and the thiirane phenoxy propylene sulfide via thioacyl anionic group transfer polymerization (TAGT). Thermal analyses of the resulting block copolymers provide evidence of microphase separation of the blocks. The resulting diblocks can be further chain extended using photoinduced electron/energy transfer reversible addition-fragmentation chain transfer polymerization (PET-RAFT) of N-isopropylacrylamide to make a triblock terpolymer from three different monomer classes incorporated via three different mechanisms without any end group modification steps. The development of this simple, sequential synthesis using a universal mediator opens up new possibilities by providing facile access to diverse block copolymers of vinyl ethers, thiiranes, and acrylamides.

Block copolymers (BCPs) are important and versatile materials because of their unique properties and functions, which are dictated by their composition and microstructure. Do not be a made from a single polymerization mechanism where compatible monomers are added sequentially, but the accessible chemical space is limited to similar monomer classes or to a prescribed addition order to maintain the active chain-end. Ideally, polymer chemists would not be confined to this paradigm, such that disparate monomers could be combined to access an expanded property space. To make this dream a reality, it is necessary to develop new ways to incorporate different polymerization mechanisms (e.g., cationic, anionic, radical) into a single material.

Polymer chemists can combine multiple mechanisms into a single polymer chain through a variety of strategies. Common methods - such as the coupling of two homopolymer chains,5-7 end-group modification,8-21 or the use of multifunctional initiators 22-25 - require additional synthetic steps and/or are limited in the architecture and number of blocks in the final material. The use of a universal mediator that can support multiple mechanisms without modification is an elegant solution to these challenges (Figure 1A). Once synthesized, the mediator can facilitate multiple mechanisms via a simple sequential addition of the respective monomers and reaction conditions. Due to the fact that the mediator stays at the chain end, the user is not limited in the number of blocks accessible. Thiocarbonyl thio compounds (TCTs) have been utilized as universal mediators to combine radical and cationic26-31 as well as radical and anionic

polymerizations<sup>32-37</sup> (Figure 1B), but this strategy has not

yet been applied to the historically more-challenging cationic and anionic mechanisms. Due to the rich literature of cationic<sup>27,38-43</sup> and anionic<sup>32,44,43</sup> homopolymerizations using TCTs, we hypothesized that proper selection of the R and Z groups on the mediator would fill this gap.

Herein, we report a simple multi-pot sequential polymerization that combines photocontrolled cationic polymerization (photo-CP) and thioacyl anionic group transfer polymerization (TAGT) to generate novel BCPs of poly(vinyl

#### A Multi-mechanism BCPs via a universal mediator

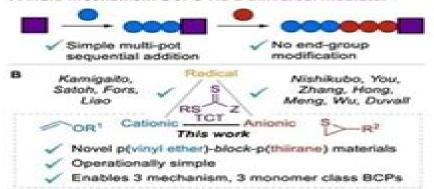


Figure 1. (A) General scheme for the synthesis of BCPs using a universal mediator. (B) Previous TCT universal mediator reports for radical/cationic and radical/anionic and this work on cationic/anionic polymerization.

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