Dental restorations represent one of the conceptually simplest biomaterials – essentially requiring the restoration of mechanical function in an aesthetically acceptable manner. Yet, despite that conceptual simplicity, an array of complex and often conflicting material, biological, curing and reaction properties necessitate a series of suboptimal solutions. Hundreds of millions of restorations are performed each year and premature failure of these restorations results in the need for a far greater number of restorations, including the occasional removal of additional healthy tooth structure. It is collectively recognized that stresses applied to the material during placement and in use are responsible for several adverse phenomena that may lead to ultimate mechanical failure of dental restorations.

In accordance, here, it is proposed an innovative approach to develop stress-relieving dental materials by incorporating two fundamentally distinct dynamic covalent chemistries (DCCs) at the resin-filler interfaces together with the bulk resin phases of dental composites characterized by high content of particulate matter. The proposed double-dynamic strategy will significantly improve the composites' performance upon the chemical and mechanical behavior compared with current composite systems that utilize a static covalent bond at the interface and in the organic monomer phase.

Within the major goals of this proposal, the project aims at comprehensive characterization, fundamental understanding, optimization and implementation of DCC-based dynamic approaches in composite mixtures capable of allyl sulfide-thiol, disulfide-thiol, (AFT) and thiol-thioester (TTE) exchange reactions, through the activation of either AFT or TTE at the filler-resin interface and the resin itself to eliminate interfacial stress and improve toughness. These altered composite structures are hypothesized to facilitate stress relaxation during (AFT) and after polymerization (TTE), to improve fracture toughness, and to improve lifetimes as indicated by fatigue analysis. Moreover, the ceiling composite properties will be determined and adjusted appropriately for improved reactive group conversions, initially developed stresses and enhanced composites' structural homogeneities. Multiple-phase exchange reactions will be taken advantage of and further extended in time post-polymerization, and overall, a significant enhancement of the mechanical characteristics will be expected. Such composites will resist the typically observed property decline following composite implantation. Based on unambiguous preliminary results that demonstrate the ability to achieve stress relaxation even when only the filler surface is dynamic, it is hypothesized that composites formed with either type of DCC-modified interfaces will represent ideal candidates for implementation in a variety of commercial composite dental restoratives and beyond. Normalized characterization methods for stress relaxation, shrinkage stress, reaction rates, ultimate conversions, mechanical properties, will all be crucial in comprehensive and in-depth assessment and understanding of the interface and resin phase relaxation phenomena.

Ultimately, the overall aim is to establish the value of using DCC-functional interfaces in dental composites for all resin-filler combinations as a disruptive approach to dental composite materials of extended lifetimes, and silica filled composites in general.