

# New Organic-Inorganic Multifunctional Materials Based on Chemistry Similar to Polydopamine

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## Abstract

The creation of a set of hybrid organic-inorganic multifunctional materials has been accomplished through the development of a straightforward one-pot technique carried out in mild conditions. This method is based on the employment of catechol and KIO<sub>4</sub> as oxidising agents along with a chemical imitating polydopamine, 3-aminopropyl-, modified 3-aminopropyl and 3-methylimidazolium-1-trimethoxypropylsilane. The fifteen materials produced by reactions in water at ambient temperature and 70°C were characterised using a variety of methods (nitrogen physisorption, TGA, XPS, 13C and 29Si CP-MAS NMR, IR, pHPZC). The Knoevenagel reaction was selected as a reliable probe to look into the presence of functional groups on the material's surface. Recycling processes were used to examine the catalytic materials that were the most active. The scientific community is very interested in this straightforward method since it paved the way for the creation of new organic-inorganic hybrid materials that can serve a variety of functions.

**Keywords:** *Electrocatalysis; Hydrosilanes; Formamides; Monooxide*

## Introduction

Designing hybrid organic-inorganic materials for organocatalytic and metal-based reactions has received a lot of attention in this area as the continual development of new materials for catalytic applications as well as other reasons is a subject of considerable interest. The so-called mussel-inspired materials based on Polydopamine (PDA) or polydopamine-like polymers, which also show significant promise as catalysts in sustainable chemistry, have received a lot of attention among the multitude of new materials in the last ten years. High catalytic activity, recoverability, and reusability are combined with easy, potentially inexpensive, and environmentally friendly catalytic material production to create catalytic processes that are sustainable. An appealing method in this situation might be to oxidise dopamine or simple catechol-based chemicals to produce polydopamine or polydopamine-like

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molecules. The co-polymerization processes that result in the creation of the appropriate polymeric material necessitate the presence of amine-based derivatives when using simple catechol as an alternative source of the catechol moiety to dopamine. The nature of these materials, which is highly complex, was further studied using 4-methyl catechol and propylamine as model substances. It is understood that primary amines can bind to polydopamine covalently in a manner similar to how rings form in PDA. For the creation of new materials for particular uses, 3-Aminopropyltrialkoxysilane, also known as triethoxysilane (APTES), has been combined with dopamine, tannic acid, or 5,6-dihydroxyindole-2-carboxylic acid. We recently reported a straightforward and inexpensive method for the synthesis of a silica-based material that resembles polydopamine by using the inexpensive catechol and 3-Aminopropyltrimethoxysilane (APTMS), which serves as the amine counterpart, in light of the growing interest in mussel-inspired chemistry. Then, in a very simple manner, we began a deeper inquiry into the creation of novel hybrid organic-inorganic multifunctional materials. In fact, the final material produced by the oxidation-polymerization of catechol, followed by the nucleophilic addition of the amine moiety and hydrolytic polycondensation of the trimethoxysilane group, can exhibit a variety of functional groups, including amine, imine, phenolic hydroxyl, and carbonyl groups. The Knoevenagel reaction was selected as an effective probe to examine the availability of functional groups on the surface of the newly synthesised materials in order to test them. The Knoevenagel reaction is a highly helpful process for the entire synthesis of biologically active natural chemicals and their synthetic counterparts, electroluminescent compounds, for organic solar cells, and in medicinal chemistry, among other applications. It can occur through a variety of routes. It was simple to produce hybrid materials using a one-pot method under calm circumstances. The first stage involved dissolving catechol in 10 mM, pH 9.0, and  $\text{KHCO}_3/\text{K}_2\text{CO}_3$  buffer solution. The oxidant  $\text{KIO}_4$  was introduced with the intention of producing enough o-quinone at ambient temperature in a few moments the solution became quite black. Minutes, after which 2.5 hours were spent stirring. The following step is to The appropriate trimethoxysilane derivative was found in this murky solution for 18 hours while being agitated at  $70^\circ\text{C}$  after being introduced. The following Brown to dark-brown precipitates began to accumulate over time, which easy to filter, wash, and dry, resulting in the hybrid materials. It was simple to produce hybrid materials using a one-pot method under calm circumstances. The first stage involved dissolving catechol in 10 mM, pH 9.0,  $\text{KHCO}_3/\text{K}_2\text{CO}_3$  buffer solution and The oxidant  $\text{KIO}_4$  was introduced with the intention of producing enough o-quinone at ambient temperature In a few moments the solution became quite black. minutes, after which 2.5 hours were spent stirring. The following step is to The appropriate trimethoxysilane derivative was found in this murky solution for 18 hours while being agitated at  $70^\circ\text{C}$  after being introduced. The following Brown to dark-brown precipitates began to accumulate over time, which easy to filter, wash, and dry, resulting in the hybrid materials. The subsequent materials were made using silane A and varying concentrations of  $\text{KIO}_4$  (0.25 and 0.50 equivalent with respect to catechol). In these instances, the oxidation happened quickly. Dopamine oxidation was said to be sped up by the use of periodate as an oxidant. In our case, the mixture quickly turned quite dark after 2.5 hours at ambient temperature. Silane A was then added, and the reaction was heated to  $70^\circ\text{C}$  for 18 hours. More final material was produced by the reaction when 0.50 equivalents of oxidant were used (A2). Because of this, 0.50 equivalents try to create a polymer with fewer branches; the lack of the third methoxy group bound to silicon can lead to the creation of a more linear polymeric structure, assuring the material a larger contact surface and improving the accessibility of the amino functions. Additionally, substituting a methyl group for a methoxy one could make the final product more hydrophobic. The amount of material obtained using this process remained consistent even after the synthesis was repeated with the concentration doubled to produce E2d. Several approaches were used to characterise the hybrid materials, confirming the existence of several functional groups. First, Thermogravimetric Analysis (TGA) was used to characterise the materials. The results are compiled and thermograms are presented in the Electronic Supporting Information (ESI). Around  $100^\circ\text{C}$ , the initial weight loss was noticed and it was caused by the materials absorbing water. A crude indicator of the surfaces' hydrophilicity or hydrophobicity could be the amount of adsorbed water.