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Tetrahedron Letters

journal homepage: www.elsevier.com/locate/tetlet

Digest Paper

Surface organic chemistry for application to organic electronics

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ARTICLE INFO

Article history:

Received 29 January 2015

Revised 1 April 2015

Accepted 3 April 2015

Available online xxxxx

Keywords:

Surface functionalization

Self-assembled monolayers

Interfacial chemical reactions

Organic electronics

ABSTRACT

This digest delineates the field of surface organic chemistry and provides representative examples. Surface organic chemistry not only includes the formation of self-assembled monolayers and interfacial chemical reactions on these monolayers, but also involves tuning the physicochemical properties of bulk metal surfaces at the molecular level to achieve a targeted result. Notable synergistic effects for organic electronics can be achieved through lateral and vertical control of the chemical compositions on metal surfaces. Herein, several recent research results in the field are discussed with regard to their utility in electronic applications.

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Introduction

Chemical functionalization of solid surfaces (e.g., glass, gold, silicon oxide, and polymers) has received a great deal of attention in many fields including chemistry, physics, materials science, biotechnology, biomedical engineering, and nanotechnology.¹ The goal of chemical functionalization research is to engineer the interfaces between two different systems, for example, metal systems and organic electronic systems, at the molecular level; this imparts

fundamental understanding of interfacial phenomena for potential applications, such as biomedical technology, heterogeneous catalysis, sensors, energy storages, and organic electronics. In fact, organic chemists have employed solid substrates as heterogeneous catalysts and reaction templates for solid-phase synthesis for a long time.² For instance, peptides and DNA have been widely synthesized in specific alignments on a pore-controlled glass or porous polystyrene beads.³ However, organic chemistry using solid substrates appears to be limited to certain reactions and has not been further extended and developed, presumably because of lack of knowledge in terms of interfacial chemical reactions/interactions and their applications.

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The term, ‘surface organic chemistry’, implies that this field concentrates on interfacial chemical reactions for building molecular structures on surfaces and the interactions of these structures with biological species (e.g., DNAs, proteins, cells, and bacteria) and electrical components (e.g., semiconductors and capacitors).⁴ Therefore, surface organic chemistry is classified as an interdisciplinary research field rather than a discipline for problem solving in classical heterogeneous organic reactions, such as metal-based heterogeneous catalysis. Surface organic chemistry is conveniently divided into three main categories in this review: (1) the formation of organic monolayers (i.e., self-assembled monolayers) on Au and Si/SiO₂ surfaces, (2) chemical transformations of organic monolayers, and (3) applications to organic electronics. A number of excellent reviews covering specific aspects of organic chemistry on metals, metal oxides, and plastics as well as those of interfacial reactions on monolayers have been reported.^{4a,5a,1d,5b,1f,5c} However, there remains lack of systematic and comprehensive reviews of surface organic chemistry for application to organic electronics. Thus, this review focuses on select recent research achievements in the field of surface organic chemistry on two-dimensional surfaces and their application in organic electronics.

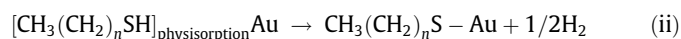
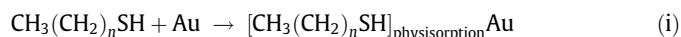
Formation of self-assembled monolayers (SAMs)

In general, self-assembled monolayers (SAMs) are highly ordered molecular assemblies formed spontaneously by the attachment of active surfactants onto a solid surface.^{1b,g} The most representative examples of SAMs are alkanethiolates on Au surfaces (thiol-SAMs) and alkylsilanes on Si/SiO₂ surfaces (silane-

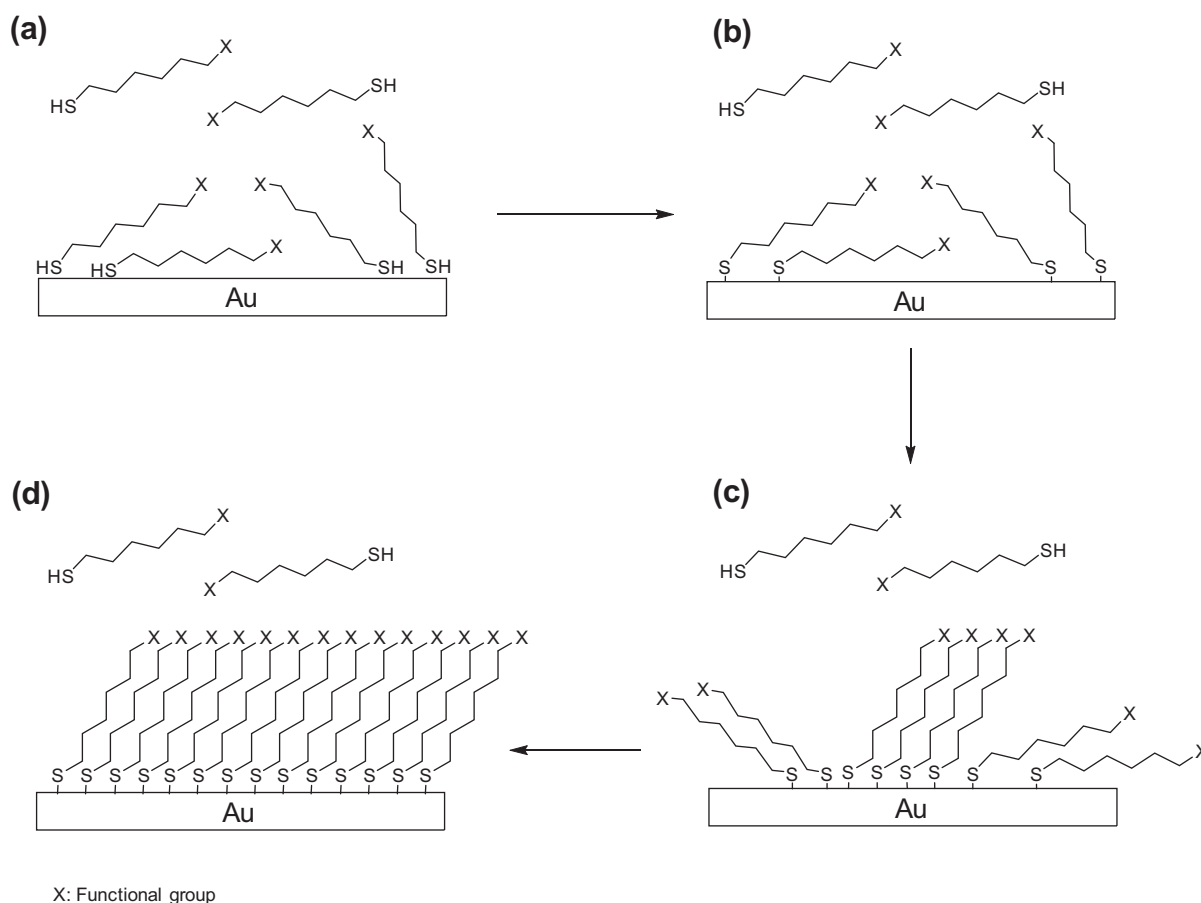
SAMs); these are characterized by the ease of formation of densely packed monolayers and their application versatility, such as in molecular electronics and silicon-based technology. Herein, we focus on the preparation of thiol-SAMs and silane-SAMs from the solution phase, the mechanistic details, and their physical properties. Although the interest in several types of SAMs (e.g., aryl azide- and catechol-based SAMs) is increasing, their discussion is beyond the scope of this review.

Thiol-SAMs on Au surfaces

Although the assembly of alkythiolates, including dialkylsulfides and dialkyldisulfides, on Au(111) surfaces has been widely investigated, the nature and mechanism of the reaction is not yet completely understood.^{1h} Nonetheless, it is assumed that the reaction mechanism can be represented as follows;



where the thiol group is first physisorbed (i) and then chemisorbed (ii) on the gold surface. Furthermore, van der Waals interactions between the alkyl groups also act as a driving force for the formation of monolayers exhibiting highly ordered and densely packed aggregation.^{1g} Scheme 1 depicts the process for the formation of SAMs of alkanethiol on Au(111).^{1h} After each alkanethiol molecule physisorbs onto the gold surface, the thiol group chemisorbs on the surface. As the number of chemisorbed molecules increases, the molecules begin to align themselves upright on the surface



Scheme 1. Schematic depiction of a plausible mechanism for the formation of thiol-SAMs on Au: (a) physisorption, (b) chemisorption of thiol onto Au, (c) nucleation of the erect phase, and (d) formation of monolayers.

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