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Supramolecular Gels: Materials with Self-Assembled Micro and Nano Fibrillar Networks

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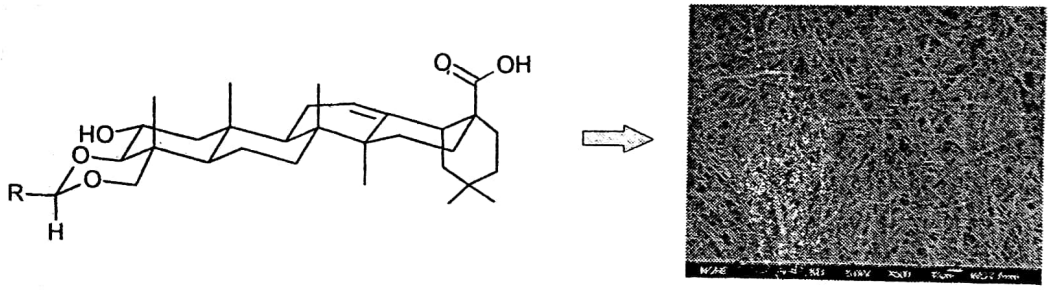
ABSTRACT

Novel classes of supramolecular gelators are synthesized from arjunolic acid which is obtained from *Terminalia arjuna*. Self-assembly properties of the derivatives have been studies in a wide range of organic solvents. The supramolecular assembly of organogels was examined by microscopy. The importance of aromatic ring and carboxylic acid group on gelation are established. The SEM images of the xerogel of gelators reveal deferent types of morphology depending upon the solvent systems and concentration of gelator



Keywords: Terminalia arjuna, Low molecular mass Gelators, Supramolecular assembly, SEM images, micro and nano fibrillar networks.

1. Introduction

Supramolecular gels are formed by the self-assembly of *low molecular mass* organic compounds in a medium have attracted considerable attention in recent years and efficient gelation has been reported with compounds of wide structural diversities.[1-5] The studies in this area have been motivated not only to gain a better understanding of the self-assembly process in a medium but also because of their numerous technological applications.[6] Self-assembly has proved to be a powerful strategy to develop molecularly defined and functional materials. The long range molecular order, obtained by self-assembly of the low molecular mass organic compounds through gelation, has recently been utilized to create self-assembled nanowires. Although there are many definitions of gels, none seems to encapsulate all of their properties. According to Flory, a gel has a continuous structure with macroscopic dimensions that is permanent on the time scale of

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Self-Assembly of Ketals of Arjunolic Acid into Vesicles and Fibers Yielding Gel-Like Dispersions

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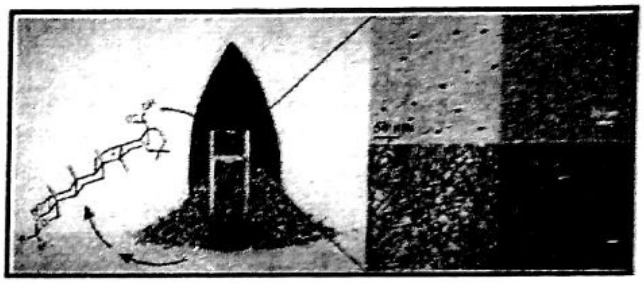
Supporting Information

ABSTRACT: Ten aliphatic and aromatic ketals of arjunolic acid, a renewable, nanosized triterpenic acid which is obtainable from *Terminalia arjuna*, have been synthesized upon condensation with aldehydes. Self-assembly properties of the ketals have been studied in a wide range of organic liquids. With the exception of the *p*-nitrobenzylidene derivative, low concentrations of the ketals self-assemble and form gel-like dispersions in many of the organic liquids examined. The morphologies of the assemblies, studied at different distance scales by optical, electron, and atomic-force microscopies, consisted of fibrillar networks and vesicles which were able to entrap 5(6)-carboxyfluorescein as a guest molecule. X-ray diffractograms indicate that the fibrillar objects are crystalline. A charge-transfer complex was formed from a 1:1 mixture of ketal derivatives with electron-donating and electron-accepting groups, and the 9-anthrylidene derivative in its fibrillar network dimerized upon irradiation. Results demonstrate that subtle changes in the ketal structures can lead to very different aggregation pathways.



INTRODUCTION

Spontaneous self-assembly of low molecular mass organic gelators (LMOGs), molecules leading to gels in liquids, has become an area of intense research interest in recent years because it affords a means to understand more deeply how and why different supramolecular architectures are formed and because 'molecular gels' have many potential and realized technological applications.[1,2] Many of these architectures are in the form of self-assembled fibrillar networks (SAFINs).[2] LMOGs based on naturally occurring species (e.g., amino acids, carbohydrates, fatty acids, steroids, and porphyrins), as well as those produced industrially (e.g., fused-aromatics), have been reported.[2] Plant metabolites, many of which are structurally very complicated, are very attractive candidates as new LMOGs because they are available in renewable supply without extensive synthetic effort. One class of these metabolites is triterpenoids. Their molecular structures are rather rigid, with lengths exceeding 1 nm, and their 30 carbon atoms are joined in very specific ways that include several centers of chirality.[3] The basic structures are complemented by functional groups at different positions and orientations. However, very few studies of the self-assembly of triterpenoids have been reported.[4]

Arjunolic acid, **1**, is a pentacyclic triterpenoid with functional groups (N. B., one carboxy and three hydroxy groups at opposite ends of the triterpenoid backbone) that are amenable to several types of easily accomplished structural modifications. For example, alkyl esters of arjunolic acid are excellent gelators of several organic liquids.[5] The cross sections of individual fibers within their SAFINs were found to be in the nanometer range, twisted in one sense only, and as a result, their gels display intense induced circular dichroism.

The steric dispositions of the hydroxyl and hydroxymethyl groups at C2−C4 of arjunolic acid have enantiomorphic resemblance to those at C3−C5 of the D-glucopyranose skeleton (Figure 1). Molecules with related backbones, such as D-sorbitol, are known to form ketal derivatives (N. B., dibenzylidene sorbitol), which are excellent gelators of a wide

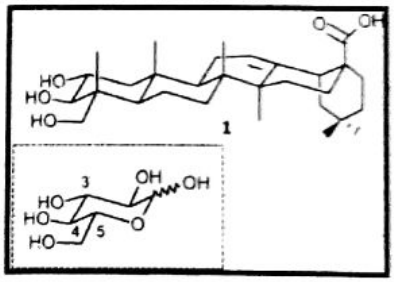


Figure 1. Structure of arjunolic acid, **1**. Inset: Corresponding array of three hydroxy groups in D-glucopyranose in a projection emphasizing the correspondence of the patterns.

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Structural elucidation, Hirshfeld surface analysis and quantum mechanical study of para-nitro benzylidene methyl arjunolate

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ABSTRACT

A benzylidene derivative of arjunolic acid, namely, para-nitro benzylidene methyl arjunolate (**3**) have been synthesized and characterized by X-ray structural studies and the electronic structure was calculated at the DFT level with a detailed analysis of Hirshfeld surface and fingerprint plot facilitating a comparison of intermolecular interactions. The crystal packing of (**3**) exhibits intermolecular O–H···O and C–H···O hydrogen bonds forming linear chains propagating parallel to [1 0 0] and [0 1 0] directions, respectively, which are further linked through C–H···π (arene) bonds to generate two-dimensional framework. Investigation of intermolecular interactions and crystal packing *via* Hirshfeld surface analysis reveals that more than two-thirds of the close contacts are associated with weak interactions. Hirshfeld surface analysis for visually analyzing intermolecular interactions in crystal structures employing molecular surface contours and 2D fingerprint plots have been used to examine molecular shapes. The large HOMO–LUMO energy gap indicates a high kinetic stability for the title compound (**3**).

1. Introduction

Arjunolic acid, a terpenoid, is the major component of the extracts of the heavy wood of *Teminalia arjuna* [1] in the form of a colorless crystalline solid and belongs to the family of combretaceae and is an important medicinal plant found in India [2,3]. Medicinal activities of arjunolic acid and other extraneous constituents of *T. arjuna* have been reported [4,5]. Clinical investigation of *T. arjuna* displayed the benefit in the treatment of coronary artery disease, heart failures and it has also been found to have antibacterial and antimutagenic properties [6]. The extract of the plant have antibacterial activity against periodontopathic bacteria [7]. Terminalia has also been prescribed as an antidysenteric antimicrobial agent and useful in the last phase of AIDS [8]. The arjunolic acid having a rigid lypophilic backbone offers a great opportunity for the construction of molecular receptors, supramolecular architectures, and functional nanomaterials [9].

The properties of crystalline material strongly depend on how the constituent components are organized with respect to one another and a control over this organization directly provides a handle over the functional properties of the material. Crystals are assembled in spontaneous process called self-assembly that proceeds through a series of molecular recognition events. Molecular self-assembly through weak non-covalent forces is the hallmark of biological systems. Among a number of non-covalent forces such as hydrogen bonding [10], C–H···π [11] and π···π [12] interactions, hydrogen bonding and C–H···π interactions have been widely utilized in directing molecular self-assembly for the construction of various supramolecular architectures.

Investigation of the structural stability of compounds by both experimental techniques and theoretical methods is of great interest. With the development of computational methods, theoretical modeling of drug design, functional material design, has become possible [13]. In recent years, density functional theory (DFT) has been favourite in theoretical modeling. Literature search shows that the DFT has a great accuracy in reproducing the experimental values of in geometry, dipole moment, vibrational frequency, etc. [14–18]. During our ongoing phytochemical investigations on this plant, we have isolated the title derivative of arjunolic acid and to investigate the molecular structure, the possibilities for intra- and intermolecular hydrogen bonding in the solid state, the X-ray structure analysis was undertaken along with the DFT calculations to investigate the molecular geometry and electronic structure. The novel chiral para-nitro benzylidene methyl arjunolate derivative (**3**) of arjunolic acid has the potential to be used as a structural framework for molecular recognition in supramolecular chemistry. An investigation of close intermolecular contacts between the molecules *via* Hirshfeld surface analysis is also presented in order to quantify the interactions within the crystal structure.

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SYNTHESIS, CHARACTERIZATION AND ANTIMICROBIAL ACTIVITY OF UNSYMMETRICAL BI-HETEROCYCLIC-AZO-COMPOUND(3-PYRIDYL-AZO - ADENINE)

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Abstract:-
Adenine and its different derivatives have so many antibacterial activities which encouraged us to synthesize unsymmetrical bi-heterocyclic azo compound containing one N-heterocyclic nucleic acid as adenine and other pyridine spacer by azo group. Designed compound of 3-pyridyl azo adenine is to be synthesis by diazotization of 3-aminopyridine and then link with adenine in KOH solution. We search out two types of azo products by chromatographic separation: 2,8-[bis-{(3′-pyridyl)azo}]adenine (**1a**) and 2-[(3′-pyridyl)azo]adenine (**1b**). Synthesized compounds were characterized by elemental analysis, conductance, melting point, IR, UV-Vis and ^{1}HNMR spectral data. Both compounds were tested for antimicrobial activities against some common gram-positive and gram-negative bacteria. One of the synthesized compounds (**1a**) exhibits a little bit antimicrobial activity.

Keywords: Adenine, 3-aminopyridine, diazotization, Antimicrobial activity.

1. Introduction:

Symmetrical bi-heterocyclic compounds linked by azo-imine groups (-N=N-C=N-)[I,II] are found in literature(**Figure-1**) and such type of azo compounds have so many activities compared to other non-heterocyclic azo compounds[III,IV-VI]. Design and synthesis of unsymmetrical azo compounds containing N-heterocyclic rings are very much scarce [VII-VIII]. Nucleic acid related study has so much enthuse as seem to biological activities. Such nucleic acid component adenine and its derivative cover too much application in different fields like, biological activities [IX-XI], agrochemicals activities like: fungicides, insecticides and herbicides [XII] and also work as multimodal ligands [XIII-XV]. Another heterocyclic compound pyridine has too much potentiality in their different form with variable activities such as photo-physical, photochemical, antimicrobial, redox and anticancer [III, XVI-XVII]. The azo-imine group in 3-pyridyl-azo-adenine has controlling power over the two biologically active adenine and pyridine compounds. Synthesis of unsymmetrical azo compound containing biologically active N-heterocyclic rings are great confront to us. Finally, we were distinctly

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ANTIMICROBIAL ACTIVITY OF NEWLY SYNTHESIZED AND CHARACTERIZED MIXED BI-HETEROCYCLIC AZO COMPOUND (3-PYRIDYL-AZO-BENZIMIDAZOLE)

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ABSTRACT:
Azo-imine group containing Pyridine and benzimidazole in mixed bi-heterocyclic azo compound should have activities in biological fields. We are convinced from literature survey of pyridine and benzimidazole derivatives for synthesis of mixed bi-heterocyclic azo compound, 2-[(3′-pyridyl)azo]benzimidazole. Synthesis has been carried out by the reaction between diazonium salt of 3-aminopyridine and benzimidazole in alkaline solution at low temperature. After purification, structure of the newly synthesized compound has been characterized on the basis of IR, UV-Vis, ^{1}HNMR and Elemental analysis. Investigation of invitro anti-microbial activity of synthesized compound was done by well diffusion method against some common gram positive and gram negative bacteria. The successfully synthesized compound exhibited highest to moderate inhibitory effect against Gram-negative bacteria.

KEYWORDS: Pyridine, Benzimidazole, Bi-heterocyclic azo, Antimicrobial activity.

1. INTRODUCTION:

With change in environment microbes can change their activities on living things. Chemists are also deliberately engaged to discover new chemical compounds which are most effective to destroy such type of new microbes. Benzimidazole is one of the most important N-heterocyclic rings containing chemical compound which has valuable diverse activities in biological fields. We have searched out virtual activities of benzimidazole and its derivatives. Benzimidazole or its different derivatives have been successfully used as drugs in different fields, like: Anticancer[Ia-b] and Antidiabatic agents.[II] It has also different biological activities, such as: Antiviral,[III] Antifungal,[IV] Anthelmintic,[Va-b] Antibacterial,[VIa-b] Antagonist,[VII] and Selective Inhibitor.[VIIIa-b] Benzimidazole or its different derivatives also have ability to use as

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Green Chemistry Refers to Pollution Prevention

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Introduction:

Chemistry is the science of matter. Are you afraid of chemistry? Many people are tried to avoid it. But avoid of chemistry is impossible. Because all things such as air around us, the water we must drink, and all living organisms are made with the chemicals. People who try to avoid all things that they regard as chemical may fail to realize that chemical processes are continuously being carried out in their own bodies. It has come to be recognized in recent years, that the science of chemistry is central to addressing the problems facing the environment. Through the utilization of the various sub-disciplines of chemistry and the molecular sciences, there is an increasing appreciation that the emerging area of green chemistry is needed in the design and attainment of sustainable development. A central driving force in this increasing awareness is that green chemistry accomplishes both economic and environmental goals simultaneously through the use of fundamental scientific principles. Recently, a basic strategy has been proposed for implementing the relationships between industry and academic, and hence, funding of the research that constitutes the engine of economic advancement; it is what many schools of economics call the "triple bottom line" philosophy, meaning that an enterprise will be economically sustainable if the objectives of environmental protection, social benefit, and market advantage are all satisfied. This is the challenge for the future of the chemical industry, its development being strongly linked to the extent to which environmental and human needs can be reconciled with new ideas in fundamental research. On the other hand, it should be easy to foresee that the success of environmentally friendly reactions, products, and processes will improve competitiveness within the chemical industry. If companies are able to meet the needs of society, people will influence their own governments to foster those industries attempting such environmental initiatives. Of course, fundamental research will play a central role in achieving these worthy objectives. Green chemistry looks at

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Green Synthesis of Bi-heterocyclic Azo compounds and its Complex formation

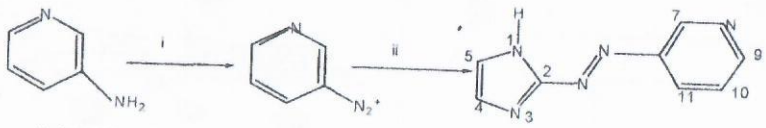
Tanmay Mathur

Introduction

There have been several occasions in the history of Chemistry community focused on goals, developing different reaction types to specific application. Now it is the turn of green chemistry towards sustainable development via pollution prevention and resource conservation. Azo compounds are an important class of organic colorants which consist of a conjugated chromophore azo (-N=N-) group in association with one or more aromatic or heterocyclic systems.[1] Azo function is photochromic, redox active, pH-responsive and their complexes act as efficient molecular switch.[2] Economic synthetic methodologies designed by use of Safer solvents[3] and auxiliaries substances for synthesis of pyridyl-azo-imidazole is a new class of unsymmetric bi-heterocycles azo compounds and have been synthesized by coupling diazonium salt of 3-aminopyridine with imidazole in aqueous Na_2CO_3 solution. A new class of [Ru(CO) (3′-PyaiH)$(PPh_3)_2$] Prepared by solution of [$Ru(CO)_3(PPh_3)_3$] in MeOH was added to the 3′-PyaiH in methanol.

Synthesis of 3′-PyaiH

Pyridylazoimidazole is a new class of unsymmetric bi-heterocycles and have been synthesized by coupling diazonium salt of 3-aminopyridine with imidazole in Na_2CO_3 solution (Scheme 1).

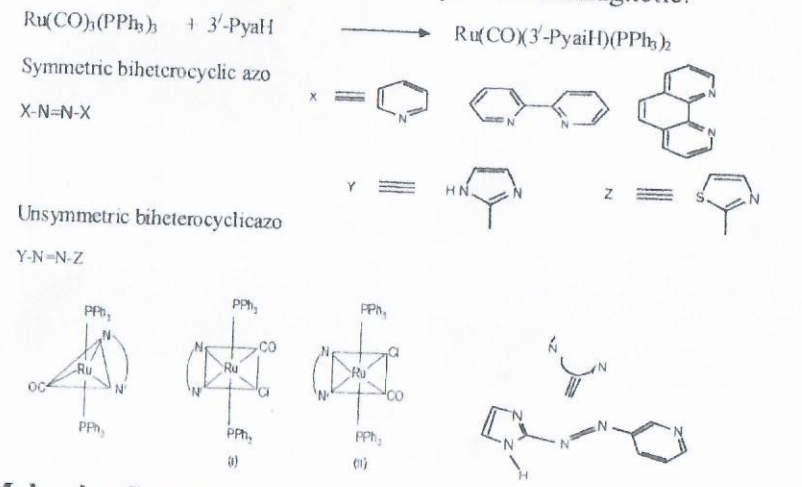


i) $NaNO_2$-HCl, 0-5°C; ii) Imidazole, Na_2CO_3 solution, pH = 6.5-7.5;

Scheme 1

Synthesis of Ru(CO)(3′-PyaiH)$(PPh_3)_2$:

In extremely moisture free condition under dinitrogen atmosphere $Ru(CO)_3(PPh_3)_3$ has been reacted with 3'-PyaiR in MeOH under a reflux of 2-2.5 hrs. The product isolated after filtration has been purified by chromatography process. The complexes are extremely stable and remain unchanged upon exposure to air even in solution for a week. The composition of the complexes, Ru(CO)(3′-PyaiH)$(PPh_3)_2$ has been supported by microanalytical data. The complexes are nonelectrolyte and diamagnetic.



Molecular Structure

i) X-Ray structure of 3′-PyaiH:

Molecular structure of 3′-PyaiH with atom numbering scheme is shown in Figure-1. Both imidazole and pyridyl fragments are

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Environmental Problems: Energy and Carbon dioxide

Dr. Tanmay Mathur

Introduction:

Good scientific theories are said to have 'predictive power'. In other words, armed only with a theory, we should be able to make predictions about a subject. If the theory's any good, the predictions will come true. Here's an example: when the table of elements was proposed, many elements were yet to be discovered. Using the theory behind the Periodic Table, the Russian Chemist Dmitri Mendeleev was able to predict the properties of germanium, gallium and scandium, despite the fact they hadn't been discovered. Increasing industrialization and unsustainable consumption patterns are escalating the environmental problems due to depletion of resources and energy. The unsustainable use of renewable resources and the generation of the toxic materials during industrial operations are creating problems to biodiversity, environment and human health.

Joseph Black, a Scottish chemist and physician, first identified carbon dioxide in the 1750s. Carbon dioxide can be found mainly in air, but also in water as a part of the carbon cycle. A substance so crucial to all life forms would not be likely to cause climate change beyond what naturally occurs in the climate system. The record shows that a warming trend of 0.8°C per year has started 300 years ago; long before the increase in greenhouse gas emissions. Global warming did not cause any severe impact on the human population in the past. However, global cooling has been known to have been a great threat over the past 5000 years. Warmer temperatures that would occur as a result of global warming would only manifest during the night and at lower temperature ranges. Increasing concentrations of carbon dioxide may cause plants to grow more easily. It is also believe that areas lost to drought and other impacts that stop plant growth may become more arable again because of increased atmospheric CO_2 concentrations. Studies by NASA suggest that the earth has become about 6% greener over the last two decades. More plants have grown in warm regions, whereas in colder regions plant growth decreased. The positive impact of global warming should