



## Low temperature, rapid solution growth of antifouling silver-zeolite nanocomposite clusters



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### ABSTRACT

Biofouling is a common and pervasive problem which reduces the efficiency of man-made marine structures. Silver-zeolite (AgZ) nanocomposite material is proposed as a promising anti-microfouling agent. Metallic silver nanoparticles were immobilized on silver ion doped ZSM-5 zeolites using a green reducing agent, trisodium citrate. The stable and porous inner structure of ZSM-5 zeolites performs a dual role as a stable size-control template and a reservoir of antimicrobial nanosilver. SEM revealed the globular and cluster-like morphology of the AgZ composites, with a homogenous distribution of silver particles on the surface of the AgZ clusters, while TEM analysis indicated Ag nanoparticles could be detected both on the surface and within the zeolite. UV–visible analysis on AgZ displayed the characteristic surface plasmon resonance absorption maximum for Ag nanoparticles ranging from 408 to 500 nm. Indeed, BET analysis also showed a reduction in surface area of up to 44% with the incorporation of Ag nanoparticles into the zeolite, indicating the formation and growth of Ag within ZSM-5 zeolite. XRD analysis indicated the presence of metallic Ag while the ZSM-5 crystalline framework remained largely intact after the Ag crystal growth process. The AgZ nanocomposites were evaluated for their biofilm inhibition activity against *Halomonas pacifica*, a common marine bacterium implicated in the early stages of biofouling. AgZ loaded with up to 10 wt% Ag reduced biofilm attachment by 81%, and inhibited the growth of marine microalgae *Dunaliella tertiolecta* and *Isochrysis* sp. Overall, results demonstrated the effective anti-microfouling property of AgZ nanocomposites.

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### 1. Introduction

The undesirable colonisation of submerged aquatic surfaces with multiple layers of biological species (e.g. seaweed and barnacles) is termed “biofouling” [1]. It is a common problem in marine-related industries, adversely affecting the efficiency of ship hulls [2], aquaculture cages [3], oil/water pipelines [4,5], and desalination plants [6]. Without appropriate protection in the form of antifouling coatings, the fouling process begins with the formation of a microbial biofilm within the first 24 h of exposure, and culminates in macrofouling by tertiary colonisers within a month [7]. Current antifouling coating technology relies on cuprous oxide-

containing biocides and other booster biocides (e.g. Irgarol, Diuron, copper pyrithione, zinc pyrithione, Sea-nine™ 211, and Zineb) [2]. However, cuprous oxide has been shown to be ineffective against certain species of bacteria and diatoms [8], while harmful levels of Irgarol and Diuron were found in coastal waters, posing threats to non-target marine lifeforms [9,10].

Silver nanomaterials (Ag NM) have been the subject of intense investigation in recent years due to their unique physico-chemical and biological properties. They are becoming increasingly important alternative antimicrobial agents in the medical field due to the emergence of pathogenic microbes with resistance to conventional antibiotics [11]. Antimicrobial Ag NM have been studied as dental resins [12], artificial bone growth supports [13], textiles [14], hydrogels [15], biocompatible polymer films [16], implant/prosthetic materials [17], food packaging [18], household and hospital

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paint coatings [19,20], water treatment systems [21–23], and in sewerage systems [24].

Zeolites are a family of aluminosilicate mesoporous ion exchange materials, with a stable network of hollow channels and pores within the size range of most monoatomic ions [25]. Due to their thermal stability and unique interconnected porous microstructure, natural and synthetic zeolites have been used as templating support materials to host a variety of metallic species including Ag [26], Mg [27], Ni [28], Zn [29], Fe [30], and Ga [31]. Interestingly, silver-containing mordenite is a leading material for the capture and storage of radioactive iodine [32]. Of note, studies of nanoparticle formation in the zeolitic internal structure using in situ pair distribution function (PDF) methods allows the study of the structure and concentration of metallic species supported by the zeolite [33]. All these studies have shown that zeolites are efficient reaction templates to produce composite materials for various technological applications. The porous internal network structure of zeolites provides an ideal and stable template for the formation and growth of nanoparticles with nanometre dimensions. Furthermore, nanoparticles are physically prevented from agglomeration to form larger nanoparticles or micron-sized particles, as they are individually separated within the discrete pores and channels of the zeolite interior.

Previous studies on Ag-zeolite nanocomposite materials proposed an ion exchange process followed by high temperature calcination methods [26,34,35] to produce metallic Ag-zeolite materials. On the other hand, Shameli et al. [36] used sodium borohydride as a room temperature chemical reducing agent to form silver-doped Y-zeolites. Sodium borohydride has been identified as a cause of environmental contamination due to the impact of borohydride anions [37]. In our study, we used trisodium citrate, a green reducing agent which does not pose an immediate catastrophic threat to the environment [37], to form Ag-zeolite nanocomposite materials at a low temperature.

Silver-zeolite nanocomposites possess great potential for marine antifouling applications due to the remarkable antimicrobial property of silver against at least 650 unicellular organisms [38]. Furthermore, zeolites have no known environmental hazards and are regularly used for environmental remediation [39], such as the removal of heavy metals from soils [40] and in the treatment of wastewater [41].

In fact, to the best of our knowledge, there have been very limited studies on adopting silver-zeolite nanocomposites for the evaluation of marine antifouling property. Previous related works include that of De Muynck et al. [42], which studied strategies for the prevention of algal fouling of outdoor terrestrial concrete surfaces using a Cu–Ag zeolite and also Ag nanoparticles. Krishnani et al. [43] reported on the bactericidal activity of silver ion-exchanged zeolite against shrimp pathogenic bacteria and its efficiency in the removal of ammonia. Inbakandan et al. [44] described the antifouling effectiveness of biogenic Ag nanoparticles against bacteria collected from seawater off the coast of Chennai, India. However, reports on Ag-zeolite composite materials against microfouling marine bacteria are still very scarce and difficult to come by.

Hence, in the current study, we synthesised Ag doped-ZSM-5 zeolite nanocomposites using trisodium citrate as a green reducing agent, and studied its anti-microfouling properties. Specifically, we tested its biofilm inhibition activity on a well-known marine bacteria, *Halomonas pacifica* (ATCC 27122), identified as one of the core organisms responsible for marine biofilm formation [45,46].

## 2. Material and methods

The ammonium form of ZSM-5 zeolite was obtained from Zeolyst International, USA. Analytical grade silver nitrate (99.9999%)

and trisodium citrate dihydrate were purchased from Sigma–Aldrich. A marine bacterium, *H. pacifica* (Baumann et al.) Dobson and Franzmann (ATCC® 27122) was purchased from the American Type Culture Collection (ATCC) and cultured in Zobell Marine Broth 2216 (HiMedia Laboratories, India). All materials were used as received. All reagents were prepared with Millipore ultrapure water (18 MΩ/cm).

### 2.1. Synthesis of silver-zeolite (AgZ) nanocomposite

Typically, 1 g of ammonium form-ZSM-5 zeolite was mixed with 20 ml of silver nitrate solution of varying concentrations from 0.1 M to 1.0 M, to produce five samples of silver-zeolite (AgZ) nanocomposite material (refer to Table 1 for sample details). The mixture was magnetically stirred in the dark at room temperature for 2 h in order to induce ion exchange, after which the mixture was separated by centrifugation (4000 rpm, 20 min). The separated Ag ion-zeolite samples were subsequently redispersed in 20 ml of deionised water. 147 ml of trisodium citrate dihydrate solution (concentrations ranging from  $3.4 \times 10^{-3}$  M to 0.034 M) were then added to Ag ion-zeolite samples, in order to reduce the encapsulated Ag ion into the metallic form. The mixture was heated to 80 °C under constant magnetic stirring for 30 min. The AgZ composites were obtained via centrifugation (7000 rpm) and repeatedly washed with excessive deionised water (three times), in order to remove impurities and unreacted precursor reagents. The metallic Ag-zeolite samples were then dried overnight in an oven at 80 °C.

### 2.2. Characterisation of silver-zeolite (AgZ) nanocomposite

The surface plasmon excitation of AgZ nanocomposites was measured using a Varian Cary 50 Conc UV–visible spectrophotometer. Two millilitres of the sample were pipetted into a 10 mm quartz cell and subjected to irradiation at 800–200 nm using a xenon lamp. The surface characteristics of the dried AgZ powder was investigated using an FEI Quanta 400F scanning electron microscope (SEM) under a high vacuum mode, using an accelerating voltage of 20 kV. A backscattered electron detector was used to detect the location of silver particles on the surface of the AgZ samples. The elemental compositions of the samples were evaluated using an Oxford Instruments X-Max with a 20 mm<sup>2</sup> EDX detector. The morphology of the zeolite and AgZ nanocomposites was observed by a JEOL JEM-2100F transmission electron microscope (TEM) at an operating voltage of 200 kV. The crystallinity of the samples was measured using a PANalytical X'Pert PRO X-ray diffractometer, equipped with Cu-Kα radiation ( $\lambda = 0.15406$  nm), in the  $2\theta$  range from 5° to 80°, with a step increment of 0.02°/s. The surface area and pore size of the composites were analysed by nitrogen gas adsorption desorption analysis, using a Micromeritics ASAP 2020 system. The samples were degassed for 2 h at 200 °C before they were analysed for the Brunauer–Emmett–Teller specific surface area ( $S_{\text{BET}}$ ) and the Barrett–Joyner–Halenda (BJH) pore size distribution. Additionally, micropore analysis was conducted using density functional theory (DFT).

### 2.3. Biofilm inhibition assay and antimicrobial assay

To assess the efficiency of AgZ in biofilm inhibition, we used a general static biofilm inhibition assay protocol involving *H. pacifica*, as reported in our previous work [47]. *H. pacifica* is a marine bacterium used as a model biofilm-causing organism in previous antifouling studies [45–47]. In brief, *H. pacifica* was cultured overnight in marine broth, and then subcultured at an optical density at 600 nm ( $OD_{600}$ ) of 0.01 into marine broth along with a suspension of the AgZ samples, to form a sample concentration of

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