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# Plasma treated activated carbon impregnated with silver nanoparticles for improved antibacterial effect in water disinfection

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## ABSTRACT

A strategy to selectively attach more Ag nanoparticles on the external surface of activated carbon (AC) is being proposed and used as an antibacterial medium for water disinfection. Ag nanoparticles were first synthesized under UV irradiation by reducing silver nitrate with sodium citrate; the latter serving the dual purpose of both a reducing and a capping agent. Transmission electron micrographs show that Ag particles have a mean diameter of 28 nm with a standard deviation of 5 nm. The AC was treated by an oxygen plasma to increase the number of polar functional groups on the surface. This carbon-surface modification treatment increased the number of Ag nanoparticles on the external surface of AC, compared to that inside pores. Fourier transform infra-red and X-ray photoelectron spectroscopy results show that the oxygen plasma treatment leads to an increase in oxygen-containing-functional groups, such as C–O and C=O, from 21.9% to 30.8%. Microbiological investigations by plate assay and shake flask tests confirmed the antibacterial nature of the AC–Ag hybrid, showing an order of magnitude increase in death rate constant from 3.72 to 41.88 h<sup>-1</sup> on plasma treatment (rate constant means rate of loss of viable cell).

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## 1. Introduction

Activated carbon (AC) is widely used as a filtration media in potable water production units for adsorption of organic contaminants [1]. It has a very high adsorption capacity due to its porous microstructure and the chemical nature of the carbon surface [2]. However, in addition, water intended for drinking should be free from fecal and total coliforms [3], which AC cannot ensure. Apart from use of AC, other promising water disinfection technologies are UV disinfection, and reverse osmosis (RO) membrane based water filtration system. UV exposure kills bacteria and virus to a certain extent, but it is not always effective, since either high levels of suspended solids, organic matter or colour reacts with UV radiation, resulting in decreased antibacterial properties [4]. On the other

hand, RO membrane based system is a pressure driven process, in which a semi-permeable membrane rejects dissolved matter and microbes present in the water. However, bio-fouling may decrease membrane flux and shorten membrane life [5,6]. AC based water filter system is still most economical to remove taste, odor and other organic contaminants.

On the contrary, bacteria may actually prefer to adhere on the surface of AC, using the latter as the carbon source [7]. So, it is necessary to prevent bacterial growth on the AC surface. Hence, AC used for water purification should not only have good adsorption capacity, but also good antibacterial activity. In order to kill bacteria and other pathogens, silver (Ag) is used with AC as an antimicrobial agent [8]. Hence, it is a challenge to retain the primary nature of carbon surface for proper removal of contaminants and yet

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ensure antibacterial property via good attachment of Ag to carbon.

One of the most important issues in antibacterial activity of Ag nanoparticles is release of  $\text{Ag}^+$  ions from particles, which is due to its oxidation in presence of water.  $\text{Ag}^+$  release can be controlled to a certain extent by using an appropriate Ag particle size, e.g., particles of 6 nm diameter have higher release (51%), in comparison to 25 nm diameter particles (5.8% release) [9]. A second important issue is aggregation of Ag nanoparticles in the dispersion due to attractive interparticle and surface forces. In order to prevent aggregation and thereby maintain effective antibacterial activity, surfactants and polymers have been widely investigated. However, actual implementations are limited due to the high cost and biocompatibility issues of these additives [10,11]. Other inorganic materials such as  $\text{TiO}_2$ , ZnO, and Cu have also been investigated to understand their antibacterial properties [12–14].

Recent reports in literature have discussed embedding of Ag nanoparticles on various porous carbon platforms – including carbon nanotube, carbon aerogel, and AC – for inactivation of microbes as well as controlled release of Ag [1,15,16]. In these reports, Ag was found to be present on both the external surface of various porous carbon and also inside the pores in these various carbon-forms. However, surface engineering of carbon for selective embedding of Ag nanoparticles only on the external surface of AC is required for efficient use of expensive, metallic Ag. The reason is as follows. Bacteria (like *E. coli*) are of dimension  $1\ \mu\text{m}$  or more, which would not be able to access Ag nanoparticles, whenever the latter are inside AC pores of size less than  $1\ \mu\text{m}$ . So, it is necessary to form Ag nanoparticles on the external surface of AC to facilitate inactivation of microbes in a more effective manner. As Ag nanoparticles have strong interaction with such functional groups – e.g., hydroxyl, carbonyl, carboxyl present on a carbon basal plane [17,18] – a large number of Ag nanoparticles can be embedded strongly on the external surface of AC.

Few attempts have been made in the literature to change the physical and chemical properties of AC for increasing the density of these functional groups. Carbon surface was modified by a wet chemical treatment (e.g., by nitric acid) [19] to introduce carbonyl and carboxyl groups for embedding particles. The disadvantage of this treatment is the resulting structural damage of aromatic planes in both bulk and near surface carbon by nitronium ions [20]. Nitronium ion attack will lead to reduction in adsorption sites for organic contaminants on the carbon surface. Heat treatment, on the other hand, may result in reduction of surface area as well as surface oxygen functional groups [21–23]. In contrast, plasma treatment is an effective method to improve the interfacial adhesion during activation and etching processes [24]. The primary advantage is that plasma treatment enables dry modification on the external surface of AC without altering bulk properties. It is possible to create an environment for oxidative, reactive and inactive reactions by changing the plasma gas.

In this work, we have demonstrated selective engineering of the AC surface by oxygen plasma treatment, so as to have Ag nanoparticles mostly on the external surface of AC. This strategy is expected to achieve improvement in the death-rate

of pathogens (like *E. coli*) in water, not only to make it potable, but also would have significant implications in being able to alter the surface-chemistry of carbon by a simple, solvent-free, purely physical protocol. The latter would be of general interest to a carbon scientist, interested in surface functionalization of carbon for various other applications.

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## 2. Experimental details

### 2.1. Materials and methods

To synthesize Ag nanoparticles, silver nitrate ( $\text{AgNO}_3$ ) was used as the Ag precursor along with trisodium citrate ( $\text{Na}_3\text{C}_6\text{H}_5\text{O}_7$ ). The latter acted both as a reducing agent and a capping agent. Milli-Q grade water (resistivity  $18.2\ \text{M}\Omega\ \text{cm}$  at  $25\ ^\circ\text{C}$ ) was used for Ag nanoparticle synthesis.

### 2.2. Synthesis of Ag nanoparticles in aqueous solution

Ag nanoparticles were prepared by photochemical reduction, as shown schematically in Fig. 1. All experiments were performed in a dark condition to avoid unwanted photochemical decomposition of  $\text{AgNO}_3$ . In a typical synthesis, 70 ml of 0.01 M  $\text{AgNO}_3$  was prepared in a beaker and mixed with 7 ml of 0.1 M  $\text{Na}_3\text{C}_6\text{H}_5\text{O}_7$  at room temperature. The beaker was positioned vertically under UV radiation (365 nm wavelength, 8 W power UV lamp) inside a closed UV chamber for 24 h. The nucleation and growth of Ag nanoparticles were favored by UV irradiation.  $\text{Na}_3\text{C}_6\text{H}_5\text{O}_7$  has three carboxylic groups, out of which two would bind to Ag, leaving the third on the Ag nanoparticle surface free; the latter stabilizing Ag nanoparticles by electrostatic repulsion [25].

### 2.3. Surface modification of AC by oxygen plasma and impregnation with Ag nanoparticles

Dry AC granules with high density polyethylene (HDPE) binder (10% by weight) were received from Filtrix Technologies Pvt Limited, Bangalore and used directly for plasma treatment. The mesh size of AC granules was  $100 \times 325$ , which corresponds to a granule size range between 44 and  $149\ \mu\text{m}$ . The role of HDPE binder is to adhere the AC granules in the form of a cylindrical filter-candle, used as the standard carbon-block in water treatment devices. We first treated the as-received AC with oxygen plasma [Harrick plasma cleaner PDC-32 G, RF (radio frequency) – 13.6 MHz, power – 18 W, pressure – 0.1 torr, plasma treatment time – 10 min] to introduce polar functional groups on the AC surface. Afterwards, 5 g of plasma treated AC samples were crushed lightly and mixed with 80 ml of synthesized Ag nanoparticle dispersion for impregnation. This mixture was stirred for 12 h at 300 rpm. Finally, the impregnated AC–Ag granules were filtered and dried at room temperature.

### 2.4. Characterization

Morphologies of samples were observed in a scanning electron microscope (SEM) (Hitachi S3400N), field emission gun-scanning electron microscope (FEG-SEM) (JSM-7600F) and transmission electron microscope (TEM) (Philips

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