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Photobiosynthesis of metal/graphene nanocomposites: new materials for water desalination and purification

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ABSTRACT

Ultra-pure water, free from metals and micro-organisms, has been easy obtained from Red seawater (Jeddah, KSA) by plasmonic-graphene hybrid nanocomposites that green synthesized using Baker's yeast (*Saccharomyces cerevisiae*) extract under visible light irradiation. Gas chromatography mass spectroscopy (GC/MS) analysis of aqueous yeast extract indicates the presence of significant biomolecules; butane-2,3-diol, glucose, undecanoic acid, and indole-3-acetic acid that can act under visible light as reducing and capping agent for the growth of (Ag, Au, and Ag/Au) plasmonic nanoparticles and graphene-based plasmonic nanocomposites. These nanocomposites able to absorb sunlight very strongly and converting it very efficiently into heat energy that enhancing the seawater evaporation. Bimetallic Ag/Au/reduced graphene-oxide nanocomposites show the highest gain of temperature and highest stability after three times of recycling processes. These eco-friendly nanocomposites are smart alternatives in water desalination and purification technique by taking the advantages of their excellent photothermal conversion, its energy-saving, cost-effective, and ease recyclability.

Keywords: Metal nanoparticles; Graphene; Yeast; Photosynthesis; Water desalination and purification; Photothermal

1. Introduction

Exploring new technologies of water treatment and water desalination has attracted an increasing interest [1]. The simple solar technology is still of main type for water treatment and purification and its design improvements have made to increase its efficiency [2]. Recently, nanotechnology-based water desalination technology has received attention [3–5], carbon nanotubes (CNTs) [6], and graphene sheets [7] that applied in water treatment and pharmaceutical products treatment. The metal nanoparticles able to concentrate the toxic compounds to part-per-billion (ppb) and sub-ppb levels that can meet the water quality standards and health care [8]. Therefore, due to the

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increasing importance of nanoparticles in environmental, health care, water treatment, and water desalination sectors, several reports attempted to design nanomaterial possessing novel and significantly enhanced physical and chemical properties [9].

The synthesis of metal nanoparticles using chemical methods leads to the presence of some toxic components adsorbed on the surface that may have adverse effects in their applications [10]. Hence, the need to develop environmentally benign nanoparticles has growing interest [11]. Biological extracts have been used for the synthesis of metal nanoparticles by adopting simple procedures, involving the process of reduction in metal ions using these extracts as a source of reducing agents either intracellularly or extracellularly [12–14]. The biological source has been used as nanomanufacturing as a green method of nanoparticles (NPs) preparation.

Intracellular synthesis of magnetite nanocrystallites is an example of naturally synthesized metal nanoparticles by biological processes [15]. Similarly, intracellular CdS nanocrystallites synthesized when certain yeast challenged with toxic metals such as cadmium [16]. Pooley reported that silver nanoparticles through silver leaching in bacteria to accumulate silver sulfide within their membrane [17]. However, the extracellular synthesis of metal nanoparticles using culture supernatants of micro-organisms was reported earlier [18–20]. The main features in this type of synthetic protocols are the preparation of nanoparticles in large quantities and easy downstream processing [21]. This has led to development of a variety of synthetic methods for better control of morphology and size. The ability to control various physical properties (size and shape) in addition to include functionalized groups on the surface has made nanoparticles attractive to search for newer applications [22–24].

Herein, we report green synthesis of (Ag, Au, or Ag/Au)/reduced graphene-oxide nanocomposites. The presented synthetic method could be useful in providing a low-cost preparation method of compatible, and stable graphene-based nanocomposites from Baker's yeast (*Saccharomyces cerevisiae*). Also, studying the applications of these composites in water desalination is based on increasing the solar evaporation of water.

2. Experimental

2.1. Preparation of aqueous extract Baker's yeast sample

Active Baker's yeast (5 g) is dissolved in 500 mL of distilled water and then heated to 90˚C for 2 h, and then filtered with 50-μm pores filter paper. Keeping this aqueous extract at 5˚C for the next preparations.

2.2. Photobiosynthesis of Ag, Au, and Ag-Au nanoparticles

For silver nanoparticles, 50 mL of $AgNO_3$ solution (1 mM) is added to 50 mL of the yeast extract in a quartz vessel. The vessel reaction is irradiated with halogen lamp for 30 min. The color of solution is changed to brownish-yellow that indicated to the silver nanoparticles formation.

For gold nanoparticles, 50 mL of $HAuCl_4$ solution (1 mM) is added to 50 ml of the yeast extract in a quartz vessel. The vessel reaction is irradiated with halogen lamp for 30 min. The color of solution is changed to pink that indicated to the gold nanoparticles formation.

For Ag-Au bimetallic nanoparticles: 15 mL of $AgNO_3$ (1 mM) and 35 mL of $HAuCl_4$ (1 mM) solutions are added to 50 mL of the yeast extract in a quartz vessel. The vessel reaction is irradiated with halogen lamp for 30 min. The color of solution is changed to violet that indicated to the nanoparticles formation.

2.3. Photobiosynthesis of Ag/reduced graphene-oxide, Au/reduced graphene-oxide, Ag/Au/reduced graphene-oxide nanocomposites

For silver nanoparticles: graphene-oxide powder (20 mg) is prepared using Hummer method [25] sonicated in 50 mL of $AgNO_3$ (1 mM) solution then added to 50 mL of the yeast extract in a quartz vessel. The vessel reaction is irradiated with halogen lamp for 30 min.

For gold nanoparticles: graphene-oxide powder (20 mg) sonicated in 50 mL of $HAuCl_4$ (1 mM) solution is then added to 50 mL of the yeast extract in a quartz vessel. The vessel reaction is irradiated with halogen lamp for 30 min.

For Ag-Au bimetallic nanoparticles: graphene-oxide powder (20 mg) sonicated in a solution of 15 mL of $AgNO_3$ (1 mM) and 35 mL of $HAuCl_4$ (1 mM) is then added to 50 mL of the yeast extract in a quartz vessel. The vessel reaction irradiated with halogen lamp for 30 min. All the samples are characterized by transmission electron microscopy (TEM) technique and UV–vis absorption spectrometer.

2.4. Seawater desalination experiment

After formation of nanocomposites, the samples are dried at 70˚C in vacuum oven and then kept in desiccator for 3 h. Ten milligrams of all the samples are added to 100 mL of red seawater (Jeddah-KSA) in a simple distillation system under irradiation with halogen lamp from 0 to 5 h (see Fig. 1). The metal/graphene samples are recovered and collected

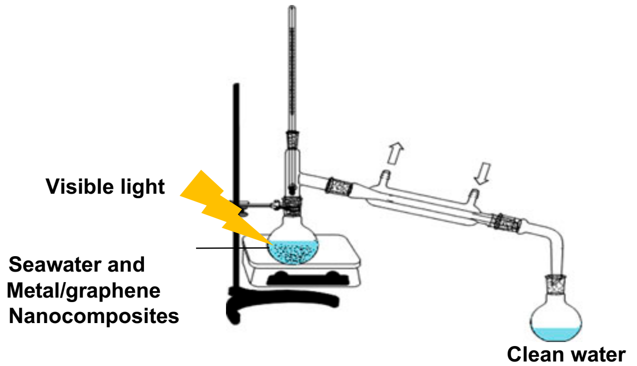

Fig. 1. Showing the water desalination and purification setup using simple distillation system.

on a Buchner funnel and washed with water (10 mL), MeOH (10 mL), acetone (10 mL), and Et_2O (10 mL). The recovered samples are then used in another desalination process of seawater.

The physical, chemical, and microbiological tests like pH, salts measurement, total dissolved salts, heterotrophic plate count, and total hardness tests for the collected water samples are carried out in the laboratory of general organization for greater Cairo water supply, Egypt.

3. Results and discussion

Firstly, it was necessary to know the yeast extract components. Gas chromatography–mass spectrometry (GC-MS) analyses of the culture supernatants show the electron impact mass spectra of butane-2,3-diol, glucose, undecanoic acid, and indole-3-acetic acid. The recorded retention times were 12.84, 18.05, 23.39, and 30.47 min, respectively. Butane-2,3-diol resulted from reducing acetone by butanediol dehydrogenase [26] (see Fig. 2 and Table 1).

3.1. Green synthesis of nanocomposites through the photo reduction of metal ions using Baker's yeast extract

When the metal ions (e.g. $AgNO_3$, $AuCl_3$) are added to aqueous solution of Baker's yeast extract and then allowed to react under visible light, the metal ions are reduced extracellularly to metal nanoparticles (Fig. 3). Fig. 3(a) shows the absorption spectra of the formed Ag, Au, and bimetallic Ag/Au nanoparticles. Fig. 3(b) and (c) shows the absorption spectra of Ag and Au nanoparticles growth as a function with irradiation time, respectively. The characterization TEM images of bimetallic Ag/Au, Ag, and Au nanoparticles are shown in Fig. 3(d)–(f), respectively. The average size of the synthesized nanoparticles is 21.0 ± 1.7 nm for Ag nanoparticles and 13.0 ± 0.9 nm for gold nanoparticles (the total count of NPs = 100 particles).

In our photo-reduction study, photosensitization of butane-2,3-diol and indole-3-acetic acid can be carried out [27]. The reduction in noble metal ions to nanoparticles catalyzed through oxidation of glucose to gluconic acid by providing electrons [28]. The aqueous

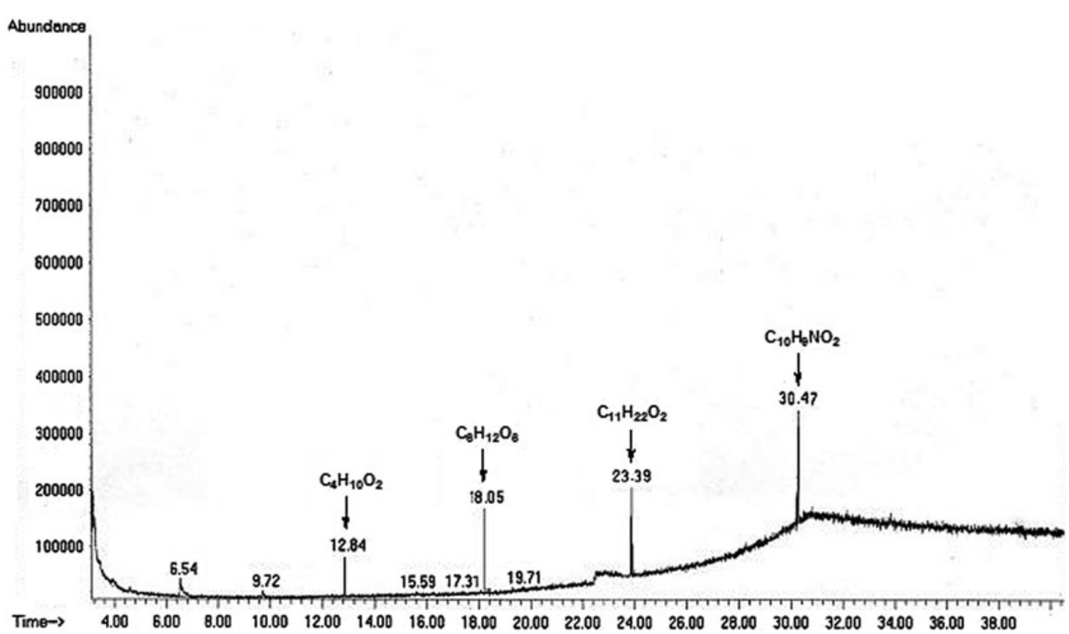

Fig. 2. Showing gas chromatography–mass spectrometry (GC/MS) analysis of aqueous Baker's yeast extract.

Table 1
Gas chromatography–mass spectrometry (GC-MS) data of aqueous Baker's yeast extract

Component m/z @ RT	Compound
90.08 @ 12.84	Butane-2,3-diol
180.14 @ 18.05	Glucose
186.21 @ 23.39	Undecanoic acid
175.16 @ 30.47	Indole-3-acetic acid

Note: m/z = molecular ion and RT = retention time.

solution of the yeast extract under visible light acts as reducing and capping agents. Nanoparticles could be stabilized also by the presence of van der Waals forces between the oxygen and negatively charged groups present in the molecular structure of the butane-2,3-diol or indole-3-acetic acid, and the positively charged groups that surround the surface of NPs according to the chemical equation in Fig. 4.

The same protocol was used to prepare graphene-based metal nanocomposites after reducing the metal ions to metal(0) and graphene oxide to reduced graphene oxide in the presence of aqueous yeast extract under visible light irradiation (Fig. 5).

Our results furnish facile method for green synthesis of metal (Ag or Au) nanoparticles and graphene-based nanocomposites (Ag/G, Au/G, and (Ag/Au/G) using yeast extract under sterile aqueous conditions, which have offered a potential application in water treatment and desalination. The average size of the formed particles decreased in the presence of reduced graphene oxide due to the reduced graphene oxide that reduces the growth rate of the particles and reduces the size [29,30]. The average size of Ag nanoparticles decreased from 21.0 ± 1.7 to 15.0 ± 1.4 nm in Ag/reduced graphene-oxide sample and the average size of Au nanoparticle also decreased from 13.0 ± 0.9 to 9.0 ± 1.1 nm for Au/reduced graphene oxide.

3.2. Water desalination using the synthesized nanocomposites

The Surface Plasmon Resonance, unique property of plasmonic nanoparticles leads to the photothermal property. The strongly absorbed light is converted to heat quickly via a series of nonradiative processes which can be considered a transfer of optical energy to thermal energy as the production of strong

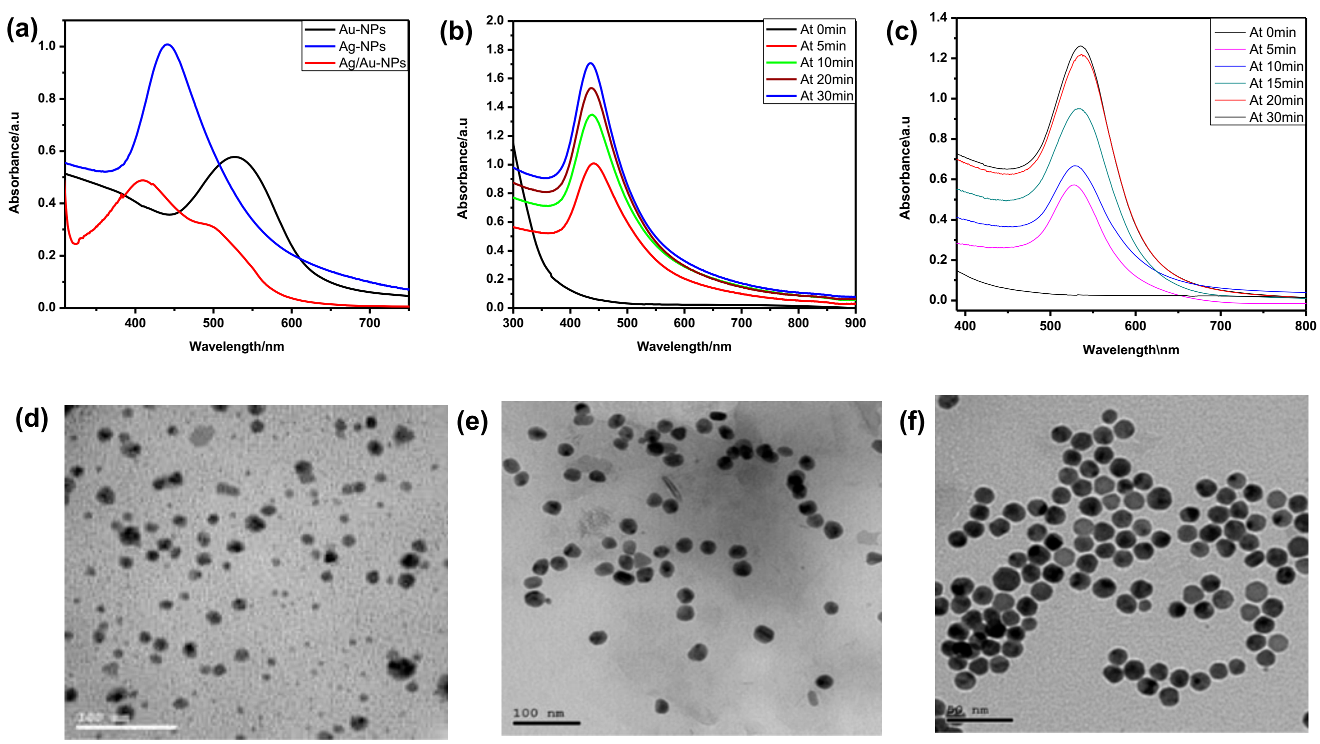

Fig. 3. Showing the absorption spectra of metal (Ag, Au, Ag/Au) nanoparticles (a), Ag NPs (b) and Au NPs (c). TEM images for the growth, Ag/Au NPs (d), Ag NPs (e) and Au NPs (f).

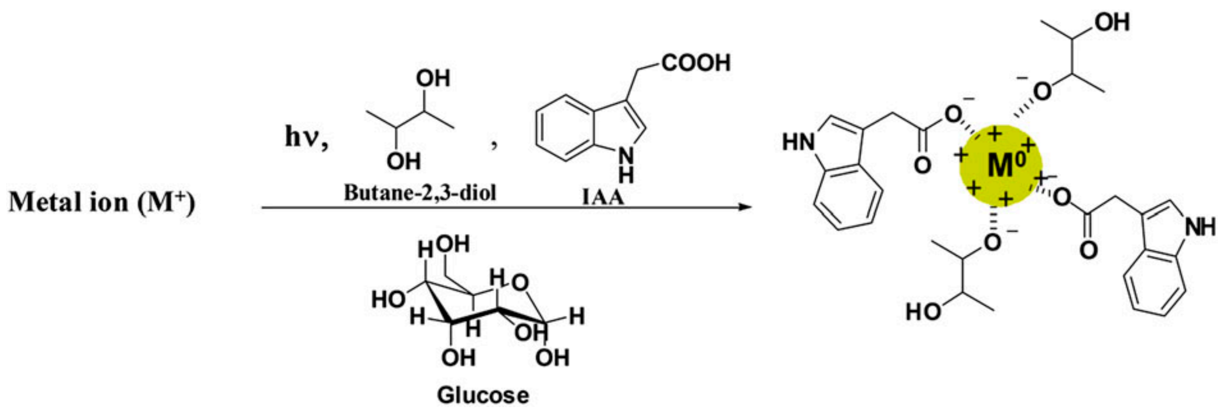

Fig. 4. The nature of the interaction between the charged NPs with butane-2,3-diol and indole-3-acetic acid.

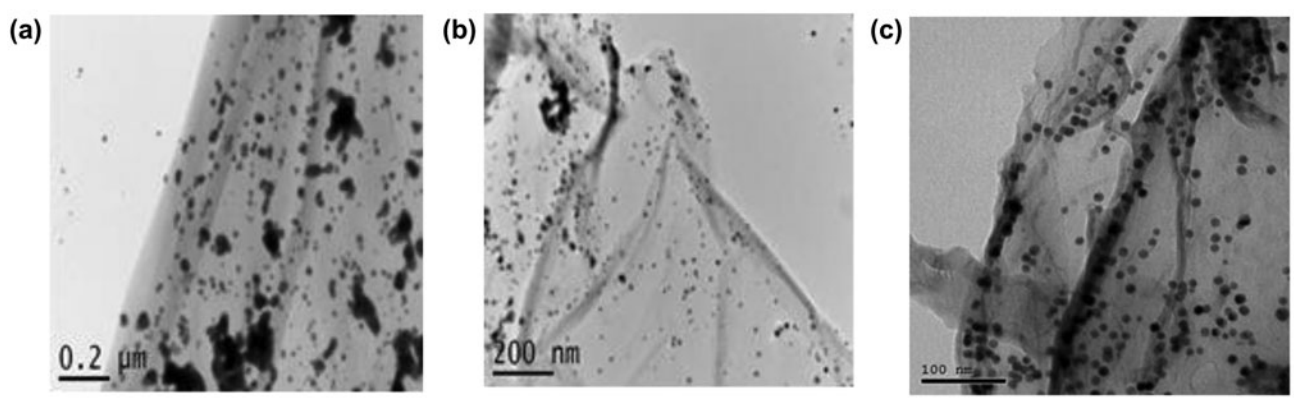

Fig. 5. TEM images showing the growth of Ag/Au/reduced graphene-oxide nanocomposites (a), Ag/reduced graphene-oxide nanocomposites (b) and Au/reduced graphene-oxide nanocomposites (c).

electromagnetic fields on the particle surface and thus consolidated all the radiative properties such as absorption and scattering [31]. This efficient conversion of photons into heat made plasmonic materials as new materials' own photothermal effect. Researchers show a great interest to use plasmonic materials in different applications like cancer diagnosis and photothermal therapy in the last 15 years [32]. On the other hand, graphene is a good photo-absorber especially in the near IR region and convert this energy to heat as very good photothermal material. Moreover, graphene has the great thermal conductivity ever compared to other materials which exposed high thermal conductivity (5,300 W/mK) at room temperature [33]. Therefore, graphene-plasmonic hybrid nanocomposites are expected to have greater photothermal properties more than plasmonic nanoparticles or graphene themselves. Thus, we test their effect in seawater or brackish water distillation as a photothermal application. The process of seawater desalination and purification by evaporation and condensation is known as "water distillation". This work includes the use of plasmon properties of the formed nanoparticles and the thermal capacity of graphene-based plasmonic nanocomposites to condense and transfer electromagnetic radiation of visible light to enough thermal energy to evaporate water.

As shown in Fig. 6(a), the gain in temperature with time is much greater in case of bimetallic Ag/Au/reduced graphene-oxide nanocomposites than other materials as temperature within the experiment (120 min) reaches 99℃, while in case of the nearest competitor, Au/graphene nanocomposites temperature reaches to 97℃ after 300 min. Meanwhile, other water samples show lower gain in the value of temperature, blank sample (seawater without nanocomposites) had the surrounding temperature that did not exceed 35℃ (no purified water after 5 h for blank sample). Samples of seawater that treated with metal nanoparticles had lower gain of temperature in

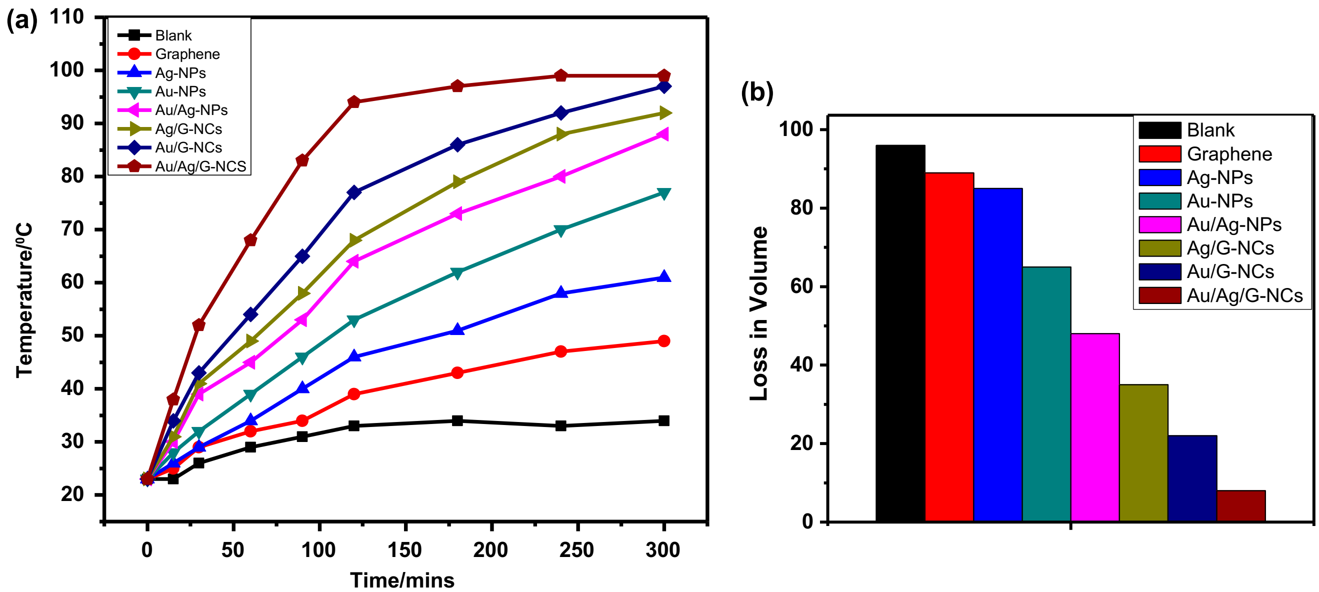

Fig. 6. (a) Showing the gain in temperature of the seawater that treated with different plasmonic nanoparticles and graphene-based plasmonic nanoparticles that photo-biosynthesized by using Baker's yeast extract comparing to bank sample for red seawater collected near Jeddah-KSA and (b) the loss in volume of the treated samples after 5 h of irradiation to visible light.

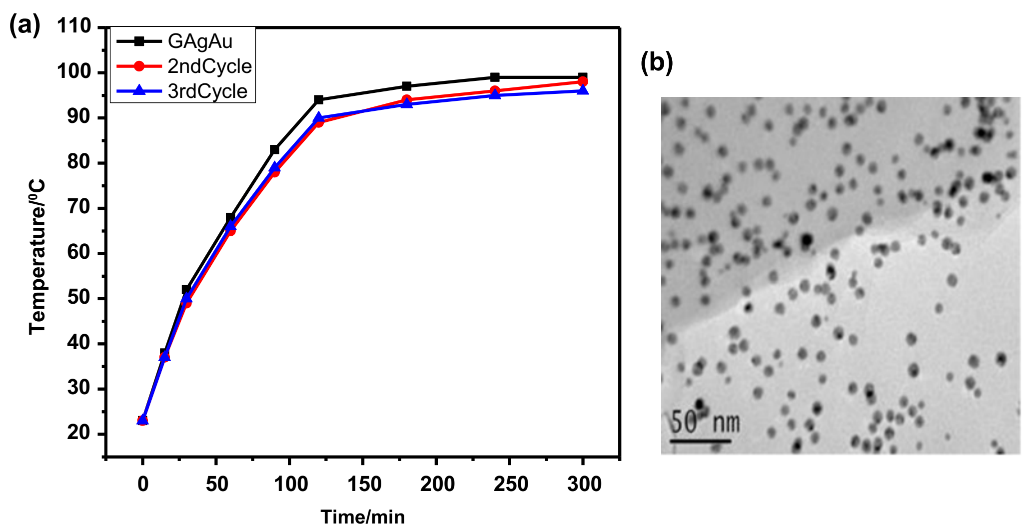

Fig. 7. Showing the gain in temperature of the seawater that treated with Ag/Au/reduced graphene-oxide nanocomposites for three cycles of water distillation experiment (a) and (b) is TEM image of the Ag/Au/reduced graphene-oxide nanocomposites after the third cycle which showing high stability of this composite.

comparison with graphene/plasmonic nanocomposites, which agree with the loss in volume of seawater showed in Fig. 6(b). This performance confirms that graphene/plasmonic nanocomposites have much greater thermal effect on seawater due to the heat transfer between plasmonic materials and graphene, which is excellent light-heat absorber and efficient thermal capacitor. Also, because of the decrease in size of the plasmonic (Ag and Au) in the presence of graphene which increase the surface area, plasmonic/graphene hybrid composites have high temperature gain than the plasmonic materials alone. The temperature gain of plasmonic materials slightly increased when the highly efficient thermal capacitor

Table 2
Showing the difference between the pure water that obtained using water desalination and purification process in the presence of the prepared nanocomposites (Ag/Au/reduced graphene-oxide sample) and Red seawater, tap water, and market water

Water quality	Red sea water	Tap water	Nova bottled drinking water	Metal/graphene desalinated water
pH	8.2	6.7	7.4	7
Ca	386 ppm	55 ppm	11 ppm	0 ppm
Na	14,310 ppm	68 ppm	17 ppm	0 ppm
Mg	742 ppm	32 ppm	6 ppm	0 ppm
K	210 ppm	11 ppm	1.3 ppm	0 ppm
Cl	22,219 ppm	108 ppm	19 ppm	0 ppm
HCO_3	146 ppm	31 ppm	26 ppm	0 ppm
SO_4	3,115 ppm	36 ppm	26 ppm	0 ppm
F	–	1 ppm	1 ppm	0 ppm
NO_3	15 ppm	6 ppm	3 ppm	0 ppm
HPC	>6,500 CFU/ml	32 CFU/ml	20 CFU/ml	0 CFU/ml
TDS	42,840 ppm	290 ppm	120 ppm	0 ppm
Total hardness	1,320 ppm	85 ppm	41 ppm	0 ppm

Note: TDS = total dissolved salts and HPC = heterotrophic plate count.

graphene is added. Fig. 6 displays that the gain in temperature in case of Ag/Au/graphene nanocomposites comparing with other active materials is obviously noticed (Fig. 6(a)). The loss in seawater volume in case of Ag/Au/graphene nanocomposites supports the temperature gain (Fig. 6(b)).

Recycling the Ag/Au/reduced graphene-oxide sample for three times to reusing these composites in water desalination process had tested and it was observed that these nanocomposites show high stability. Fig. 7(b) illustrates TEM image of Ag/Au/reduced graphene oxide after the third cycle and excellent gain of temperature for photothermal action to get pure water (see Fig. 7(a)). The cost of the desalinated water using these environmentally nanocomposites is around 9$/1,000 gallon in comparison with plant cost $7.80/kgal [34] (calculation based on the commercial raw materials that used in the synthesis of these materials in lab. scale that will decrease when we produce these materials in industrial scale). For our future study plan, we are trying to find simple method to collect these composites to reuse many times such as to deposit these materials in the desalination membranes.

The collected water from water distillation process using plasmonic nanoparticles and graphene-based plasmonic nanocomposites tested in comparison with the seawater, tap water, and Nova Bottled Drinking Water (KSA) (see Table 2). From this table, we can note that the obtained water is ultra-pure and free from ions and micro-organisms in comparison with the other samples of water.

4. Conclusion

The present study aims to green synthesis of metal/graphene nanocomposites (e.g. silver/graphene, gold/graphene, silver/gold/graphene) using Baker's yeast extract components under visible light to be used as a low-cost, efficient, and smart alternative in the technology of water desalination and purification. Gas chromatography-mass spectroscopy (GC/MS) analysis of yeast extract indicated the presence of significant biomolecules; butane-2,3-diol, glucose, undecanoic acid, and indole-3-acetic acid that can work under visible light as reducing and capping agent for the growth of metal nanoparticles and graphene-based plasmonic nanocomposites. Ultra-pure water (free from salts and micro-organisms) obtained by exploiting the high photothermal conversion of the photobiosynthesized nanocomposites in a simple distillation system of seawater.

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