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GREEN SYNTHESIS: NANOPARTICLES AND NANOFIBRES BASED ON TREE GUMS FOR ENVIRONMENTAL APPLICATIONS

ZIELONA SYNTEZA: NANOCZĄSTKI I NANOWŁÓKNA WYTWORZONE Z NATURALNYCH GUM I ICH ŚRODOWISKOWE ZASTOSOWANIE

Abstract: The recent advances and potential applications of nanoparticles and nanofibres for energy, water, food, biotechnology, the environment, and medicine have immensely conversed. The present review describes a 'green' method for the synthesis and stabilization of nanoparticles and 'green electrospinning' both using tree gums (arabic, tragacanth, karaya and kondagogu). Furthermore, this review focuses on the impending applications of both gum stabilized nanoparticles and functionalized membranes in remediation of toxic metals, radioactive effluents, and the adsorptive removal of nanoparticulates from aqueous environments as well as from industrial effluents. Besides, the antibacterial properties of gum derivatives, gum stabilized nanoparticles, and functionalized electrospun nanofibrous membranes will also be highlighted. The functionalities of nanofibrous membranes that can be enhanced by various plasma treatments (oxygen and methane, respectively) will also be emphasized.

Keywords: tree gums, nanoparticles, green electrospinning, environmental remediation, antibacterial membranes, oxygen and methane plasma treatments

Introduction

In the past few decades, nanoparticles and nanofibres have demonstrated superior performance in numerous applications, including energy, water, the environment, medicine, and health care etc. [1-4]. However, many of the materials and processes currently used for the synthesis of nanoparticles are dependent on non-renewable resources and also generate hazardous wastes. The potential environmental and health impacts of engineered nanoparticles have been comprehensively studied and reported in the literature [5-8]. Green nanotechnology, the combination of nanotechnology and the principles and practices of green chemistry, may hold the key to building an environmentally sustainable society in the near future. Nanomaterials also offer applications to prevent pollution by utilizing certain catalytic processes to create less waste production, sense pollutants in water and the environment, destroy harmful bacteria and viruses and create clean water and a clean

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environment [8-10]. Green chemistry is a set of principles or practises that encourages the design of products and processes that reduce or eliminate the use and generation of hazardous substances [11-14]. Current green nanotechnology practices often involve the use of natural sources, non-hazardous solvents, biodegradable and biocompatible materials and energy-efficient processes in the preparation of nanoparticles [15-17].

Remarkable applications of the use of electrospun nanofibres have recently been reported in many fields such as energy, the environment, water purification, sensor devises, tissue engineering scaffolds, wound dressing, drug delivery, etc. [2, 18]. Electrospinning is one of the most proficient and sophisticated methods for the fabrication of nanofibres on a large scale and with a large surface area to volume ratio, high porosity and stability [19]. The electrospinning process parameters such as system and process variable requirements have been meticulously reported in the literature [20, 21]. In order to develop 'green electrospinning' technology it is necessary to use non-toxic, cheap, and environmental friendly solvents and materials. Compared to the current electrospinning process, which utilises hazardous and corrosive and non-ecological organic solvents, water based solvents and polymers are being developed to produce electrospun nanofibres and membranes, which make the electrospinning processes a more economical and greener technology for environmental protection.

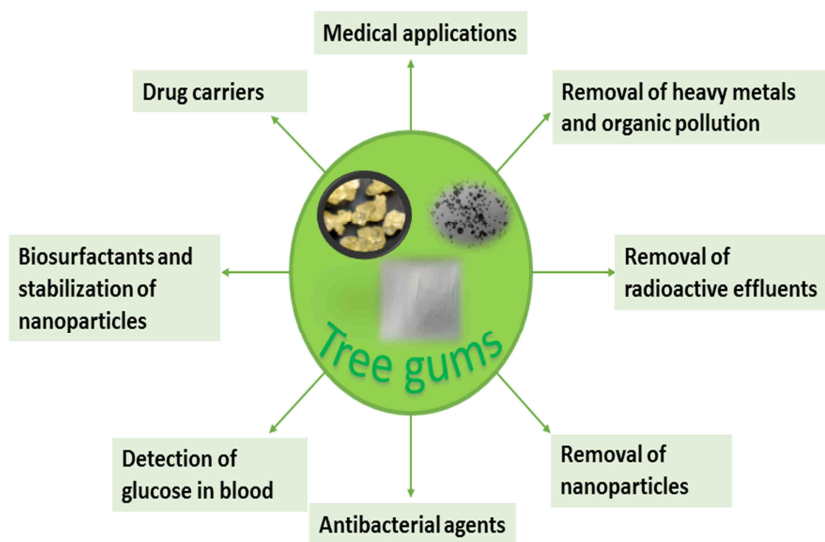


Fig. 1. Non-food applications of tree gums and created with them nanoparticles and nanofibers

In this review, we focus on the green synthesis (fabrication, characterization, and application) of nanoparticles and green electrospinning nanofibres based on tree gum polysaccharides, we also discuss the various fabrication methods using water soluble polymeric blend solutions of gums with polyvinyl alcohol or poly ethylene oxide. Furthermore, the various functionalities of electrospun fibrous membranes, surface modifications and the enhancement of their properties (water contact angle, hydrophobicity/hydrophilicity, fibre diameter, and surface area) before and after various

plasma treatments (oxygen and methane) are examined. This current report further focuses on our own studies and describes the environmental and antibacterial applications of gum-stabilized nanoparticles, functionalized fibres and membranes. Figure 1 shows the schematic model of the applications of tree gums and created with them nanoparticles and nanofibers.

Natural and renewable sources of reducing agents for the synthesis of nanoparticles

Nature has created many indispensable materials required for the fabrication of nanoparticles. Among them, plant extracts (*Salvia officinalis*, *Lippia citriodora*, *Pelargonium graveolens*, and *Punica granatum*), biopolymers (starch, cellulose, chitosan, tree gum polymers), and other natural compounds like vitamins, proteins, peptides (e.g. glutathione), and sugars (e.g. glucose, fructose) are such materials, which provide suitable reducing and surface agents for the nanoparticle synthesis/stabilization [14-17, 22-30]. Plant extracts are regarded as one of the most promising natural reducing agents, such as metabolites (e.g. sugars, alkaloids, polyphenols, phenolic acids terpenoids), and proteins and co-enzymes help to synthesis metal and metal oxide nanoparticles [31-37]. These NPs can be used in biomedical applications due to their production advantages *via* a biosynthetic route, which fashions the defined size, morphology and high chemical purity of NPs [38].

Biopolymers (cellulose and its derivatives, chitosan and its derivatives, alginate, dextran, and tree gums) are another family of natural sources used as reducing and stabilizing agents for metal and metal oxide nanoparticle synthesis [39-58]. Vitamin B₁, Vitamin B₂ (riboflavin), Vitamin C (ascorbic acid), coffee and tea extracts, beet juice, and grape pomace are well-known natural reducing agents or antioxidants used for the synthesis of stable nanoparticles and are comprehensively reported [59-65].

Tree gums - an overview

Gums are hydrocolloids, which are hydrophilic in nature and are found in almost every biosphere on earth; in plants, animals, and many bacteria. They contain a large number of hydroxyl groups, usually arranged in a fairly regular manner along the backbone of the molecule, which allows for the chelation of mono- and divalent cations, thereby cross-linking the hydrocolloid chains together and forming complex macrostructures [66-68]. Hydrocolloids are some of the most well-known polysaccharides, having complex structures with glycosidic bonding. Most gums are heterogeneous polysaccharides with complicated structures and extremely high molecular masses [69-71]. The simplest interactions of hydrocolloids are (as the name suggest) with water and it is this interaction that is key to their use in foodstuffs. Gum hydrocolloids are effective water adsorbents and to a greater or lesser extent may be solubilized by water. Owing to the high number of hydroxyl groups, water is held within the molecular structure by hydrogen bonding and also within the voids created by the complex molecular configuration [72-76]. The important tree exudate gums available on the market are gum arabic (GA), gum karaya (GK), gum tragacanth (GT), kondagogu gum (KG), and gum ghatti (GG). Extensive research has been carried out on various aspects of these tree gum polysaccharides, which includes their availability, molecular weight distribution, chemical structures, and food & non-food

applications [77-80]. None of the three gums are produced in developed countries. They must be imported from developing countries. The chemical composition of these gums is complex and varies depending on the source and its age. Therefore, it is not possible to provide defined structural formulas of these biopolymers. Exudate gums are used in an overwhelming number of applications, especially in the food industry. However, there are also considerable non-food applications [78].

Gum arabic (GA) is a branched, neutral or slightly acidic, complex polysaccharide obtained as a mixed calcium, magnesium, and potassium salt. The backbone consists of 1→3-linked β -D-galactopyranosyl units. The side chains are composed of two to five 1→3-linked β -D-galactopyranosyl units, joined to the main chain by 1→6-linkages. Both the main and the side chains contain units of α -L-arabinofuranosyl, α -L-rhamnopyranosyl, β -D-glucuronopyranosyl, and 4-O-methyl- β -D-glucuronopyranosyl units, the latter two mostly as end-units [81-83]. It is suggested that the high-molecular-weight fraction of the gum consists of large carbohydrate blocks with a molecular mass of approximately $2.5 \cdot 10^5$ Da attached individually to a polypeptide chain. GA is obtained from the stems and branches of *Acacia Senegal* and *Acacia seyal*, and being a branched polysaccharide, it exhibits unique structural, physical and chemical properties [84-87]. Consequently, it is widely used in food and pharmaceutical applications [88-90].

Gum tragacanth (GT) is a complex, highly branched, heterogeneous polysaccharide, naturally occurring as a slightly acidic calcium, magnesium, and potassium salt. It has a molecular mass of approximately $8.4 \cdot 10^5$ Da [91, 92]. The composition of the gum obtained from different *Astragalus* species shows considerable variation. This variability is not surprising, since the genus *Astragalus* is the largest within the *Leguminosae* family. It occurs worldwide in tropical regions and contains around 2,000 species, grouped into more than 100 sub-divisions [93]. GT consists of two fractions; tragacanthic acid or bassorin is insoluble in water, but has the capacity to swell and form a gel, and the other fraction is called tragacanthin and is water-soluble. Both fractions contain small amounts of proteinaceous material and methoxyl groups, the latter being present in higher amounts in the water-soluble fraction [94]. The water-swellaable tragacanthic acid fraction has a high molecular weight and a rod-like molecular shape. The main chain is formed by 1,4-linked D-galactose residues with side chains of D-xylose units attached to the main chain by 1,3 linkages. The water-soluble tragacanthin is a neutral, highly branched arabino-galactan with a spherical molecular shape. Its structure probably consists of a core composed of 1,6- and 1,3-linked D-galactose with attached chains of 1,2-, 1,3-, and 1,5-linked L-arabinose [75, 95]. GT is mainly used in food and pharmaceutical fields.

Gum karaya (GK) is a complex, partially acetylated polysaccharide obtained as a calcium and magnesium salt. It has a branched structure and a high molecular mass of approximately $16 \cdot 10^6$ Da [96, 97]. The backbone of the gum consists of α -D-galacturonic acid and α -L-rhamnose residues. Side chains are attached by 1,2-linkage of β -D-galactose or by 1,3-linkage of β -D-glucuronic acid to the galacturonic acid of the main chain. Furthermore, half of the rhamnose residues of the main chain are 1,4-linked to β -D-galactose units [92, 98, 99]. The chemical composition of gum samples obtained from different *Sterculia* species and from different places of origin was found to be quite similar [96, 100]. Commercial gum karaya contains approximately 13-26% galactose and 15-30% rhamnose, which is considerably higher than the rhamnose content of other commercial exudates gums [100]. However, the protein content of approximately 1% is lower than that

of other exudate gums. Gum karaya contains approximately 40% uronic acid residues and 8% of acetyl groups [98]. Due to the presence of these acetyl groups; native gum karaya is insoluble and only swells in water. Le Cerf et al [97] distinguished three fractions in gum karaya, based on their solubility in water. Only 10% of the native gum was solubilized in cold water, increasing to 30% in hot water. After deacetylation with dilute ammonia, 90% of the native gum dissolves in water. The equivalent weight of the deacetylated-soluble fraction was higher than that of the cold-water-soluble fraction. This indicates that only lower-molecular-weight molecules are able to dissolve in cold water, while deacetylation leads to the solubilisation of material of a higher molecular weight [78, 97].

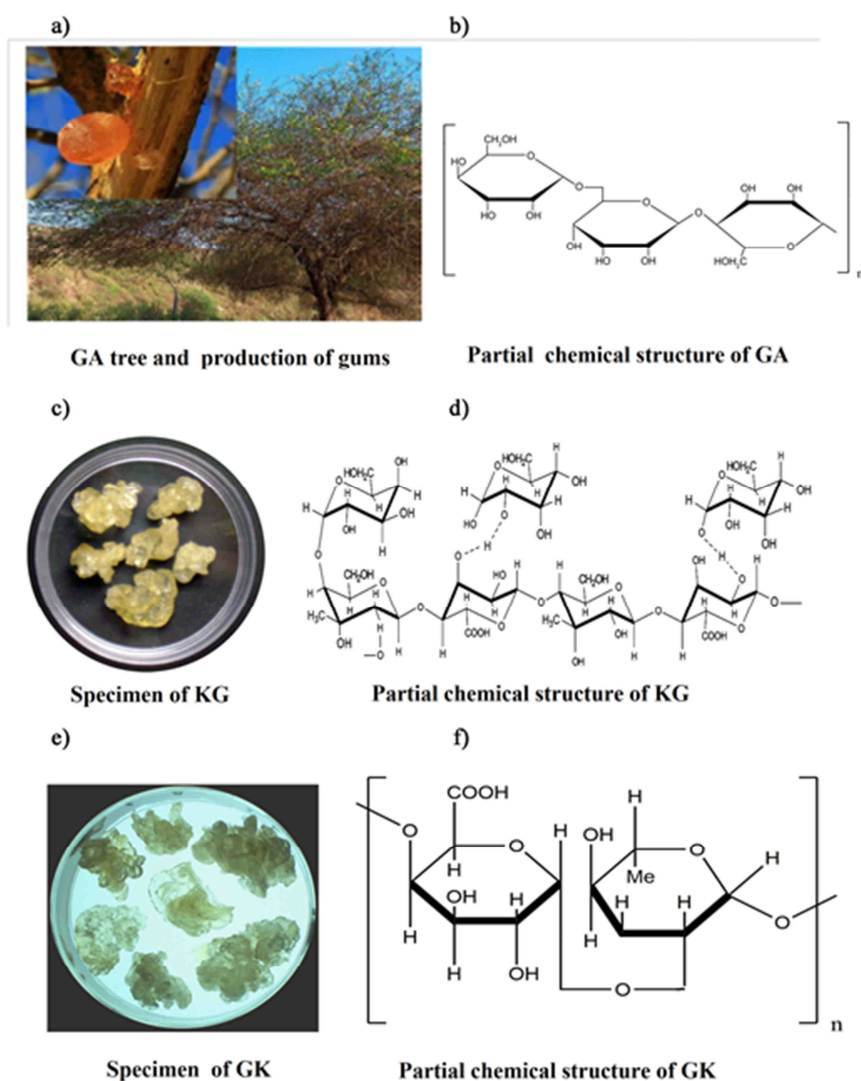


Fig. 2. Specimens of: a) GA, c) KG, and e) GK with their corresponding partial chemical structures (b, d, and f, respectively) [79]

Gum kondagogu (KG) belongs to the species of *Cochlospermum* and family *Bixaceae*. Even though gum karaya, also known as Indian tragacanth, and KG are classified in the same group, there are considerable differences in their physical and chemical properties [101, 102]. Extensive research has been carried out on KG (*Cochlospermum gossypium*), a gum extracted from kondagogu tree, which grows in India, including its morphological, physical and chemical, structural, rheological, pharmaceutical emulsifying properties and its toxicological evaluation as a food additive [79, 80, 102-107]. Furthermore, this gum can also be used as a biosorbent for the removal of toxic metal contaminants from aqueous environments and also as an environmental friendly material for the stabilization and a reducing agent for the synthesis of metal/metal oxide nanoparticles [108-112]. Structural analysis of this biopolymer has shown that it contains sugars such as arabinose, rhamnose, glucose, galactose, mannose, glucuronic acid and galacturonic acid. Based on spectroscopic characterization, the probable structural feature assigned to KG is (1 → 2) β-D-Gal *p*, (1 → 6) β-D-Gal *p*, (1 → 4) β-D-Glc *p* A, 4-O-Me-α-D-Glc *p* A, (1 → 2) α-L-Rha, and (1 → 4) α-D-Gal *p* A [79, 80].

Gum ghatti (GG), an Indian gum, is a non-starch polysaccharide, and the main species is *Anogeissus latifolia* (Combretaceae, Myrtales), a large deciduous tree found in dry areas [113, 114]. GG is used as an emulsifier and thickener in food industries [115-117]. Recently, Deshmukh et al [118] reported a detailed review of the molecular structure, properties, and pharmaceutical applications of GG.

Tree gums as a natural renewable source for the green synthesis of nanoparticles

Tree gums (GA, GK, KG, GT, and GG) have been used as a template for the synthesis and stabilization of various metal (Ag, Au, Pt, Pd, Fe, Cu, Se etc.) and metal oxide (Fe₃O₄, CuO, ZnO etc.) nanoparticles [27, 28, 58, 65, 124, 119-139].

All of the above-mentioned NPs (Table 1) were characterized using a variety of spectroscopic and microscopic analyses, such as UV-visible spectrophotometry, scanning electron microscopy (SEM), transmission electron microscopy (TEM), Fourier transform infrared spectroscopy (FTIR), powder X-ray diffraction (XRD) and energy dispersive spectroscopy (EDS). Vinod et al [131] reported that the colloidal NPs with average sizes of Ag (5.5 ± 2.5 nm), Au (97.8 ± 2.3 nm) and Pt (2.4 ± 0.7 nm) stabilized with KG were found to be stable even after 6 months at room temperature, and the reduction was attributed to the various functional groups (–OH, –COO, –C=O, and CH₃CO–) present in the gum structure. Furthermore, gums have three major parameters for the preparation of NPs and follow the cardinal principles of green chemistry; *i.e.* the environmentally benign solvent medium (water and ionic liquid-based green solvents) for the synthesis of NPs; gum acts as reducing agent due to the presence of many functional groups in the gum structure (–OH, –COO–, –CO, and CH₃CO–) and gums are non-toxic and biodegradable materials for the stabilization of NPs. However, even today, many studies have reported the use of toxic solvents such as organic solvents, and toxic, highly reactive and environmentally and biologically risky reducing agents such as hydrazine, sodium borohydride and dimethyl formide, and capping agents such as EDTA, triethanolamine and tetraethylammonium bromide, which are known to bio-accumulate in the environment and are persistent pollutants, which may have ecological or human health risks, used for the synthesis of NPs [8-10, 17].

Table 1

Synthesis of metal and metal oxide NPs interceded by various tree gum polysaccharides

Tree gums	Type of NPs	Size, morphology and stability of NPs	References
Gum arabic (GA)	Ag	~ 5 nm; face centred cubic structures with crystalline, 5 month stability	[119]
GA	Ag	16.0 \pm 2.0 n.m; FCC, highly stable	[120]
GA	Ag	2-20 nm; spherical shape, single crystalline, highly stable for one month	[121]
GA	Au	6.52 \pm 0.66 nm, spherical, stable for 5 weeks	[122]
GA	Au	5.5 nm, with spherical particles; crystalline	[123]
GA	Au	26.8 \pm 5.3 nm, with FCC structures, spherical, stable even in the presence of NaCl up to 3 M	[27]
GA	Au	15-20 nm, spherical	[124]
GA-Fe ₃ O ₄	Au	2 nm, spherical	[65]
GA	Se	~34.9 nm, with spherical structures	[28]
GA	Cu	~ 3-9 nm, with spherical particles; crystalline structure	[125]
GA	Fe ₃ O ₄	~20 nm, with non-spherical morphology	[126]
Gum karaya (GK)	Ag	12.5 \pm 2.5 nm, spherical particles, crystalline and stable for 6 months	[58]
GK	Ag	4 \pm 2 nm, FCC with crystalline structure	[127]
GK	Ag	7-10 nm, spherical	[128]
GK	Au	7.8 \pm 1.8 nm, spherical, stable for 6 months	[58]
GK	Au	20-25 nm, spherical	[129]
GK	Pt	5.0 \pm 1.2 nm, spherical	[58]
GK	CuO	10.5 \pm 2.4 nm, spherical	[130]
GK	Fe ₃ O ₄	18.5 \pm 3.5 nm, spherical	[58]
Gum kondagogu (KG)	Ag	5.5 \pm 2.5 nm, spherical, FCC, stable for more than 6 months	[131]
KG	Ag	3 nm, spherical, highly stable	[132]
KG	Au	7.8 \pm 2.3 nm, spherical, stable for more than 6 months	[131]
KG	Au	12 \pm 2 nm, nano-crystalline	[133]
KG	Pt	2.4 \pm 0.7 nm, crystalline, stable for more than 6 months	[131]
KG	Fe ₃ O ₄	spherical size iron oxide particles with diameters in the range of 8-15 nm	[134]
Gum tragacanth (GT)	Ag	13.1 \pm 1.0 nm with spherical nanoparticles	[135]
GT	ZnO	55-80 nm, high crystalline nature and single phase of synthesized	[136]
Gum ghatti (GG)	Ag	5.7 \pm 0.2 nm, spherical nanoparticles	[137]
GG	Pd	4.8 \pm 1.6 nm, spherical shape	[138]
GG	Fe ₃ O ₄	35 nm	[139]

Figures 3 and 4 show the various nanoparticles (Ag, Au, Pt, Fe₃O₄, CuO) synthesised using GK and KG.

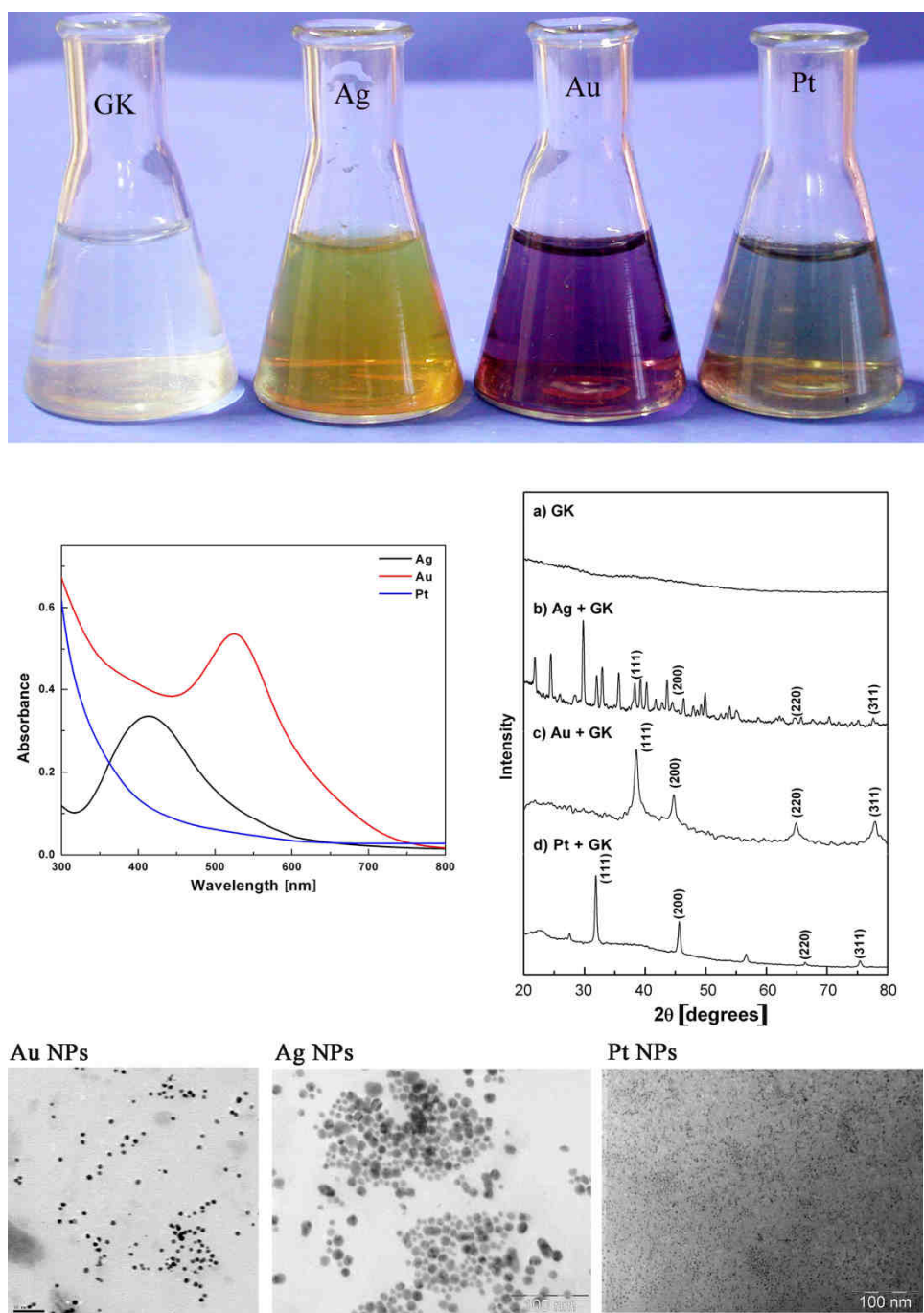


Fig. 3. Striking colours of NPs (Ag, Au, and Pt) synthesised using KG as reducing and stabilizing agent (panel 1); UV-VIS and XRD spectral analysis of NPs (panel 2); and TEM pictures of NPs (panel 3)

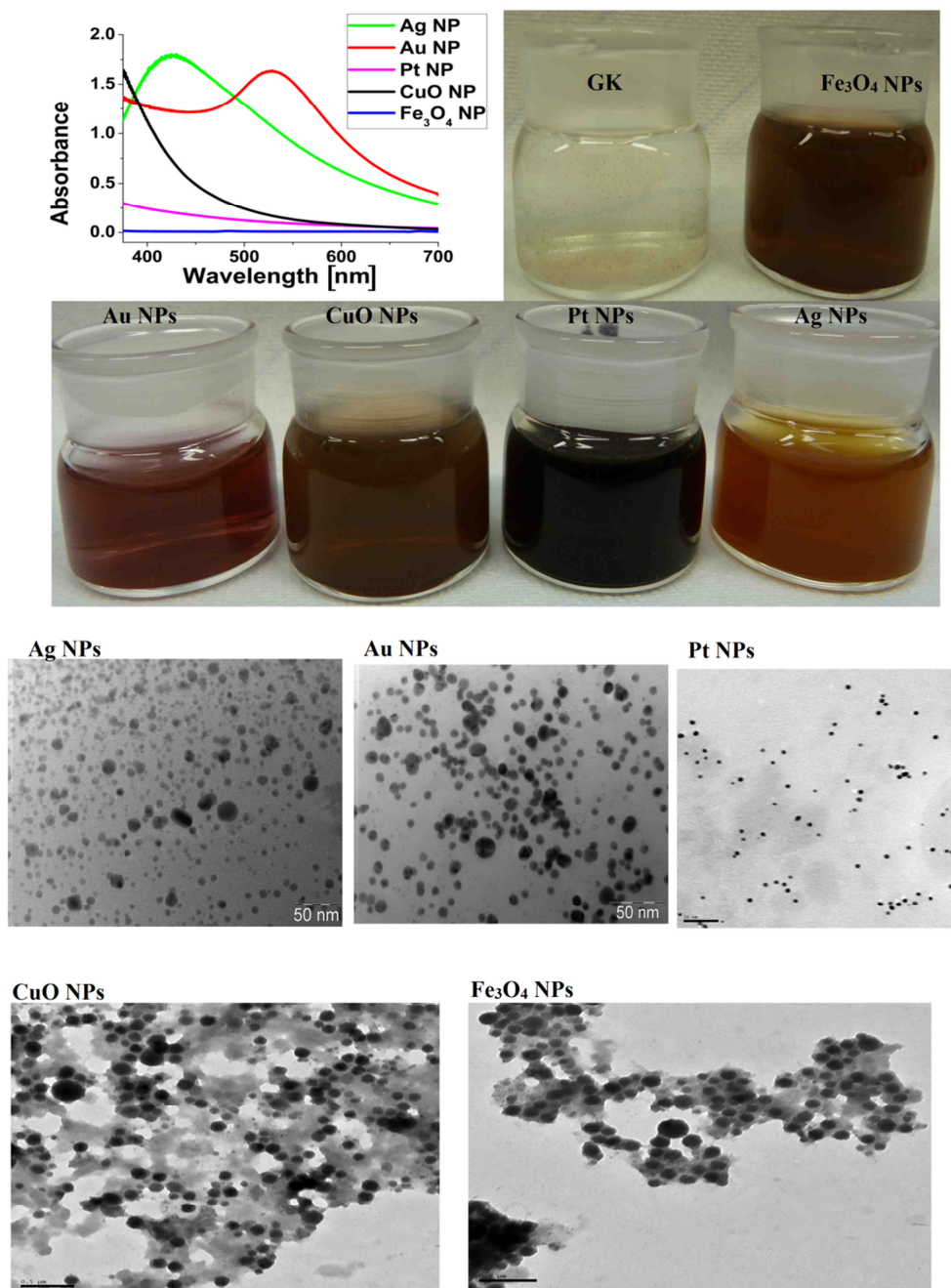


Fig. 4. NPs synthesized using GK; UV-VIS spectral analysis of NPs; showing the formation various synthesized NPs (Ag, Au, Pt, Fe₃O₄, and CuO); and TEM analysis of NPs

“Green” electrospinning based on tree gums

Fibres produced by electrospinning have been known for over ninety years, when the Czech-American physicist John Zeleny studied techniques known today as electrospinning and electrospraying. When a conductive liquid in a capillary is exposed to an electric field in a range of ~10 kV, the liquid shape forms a cone, which emits a jet of liquid. Fibres are formed by the solution drying during flight to the opposite electrode (collector). Industrial production based on roller electrospinning was developed by Oldrich Jirsak, professor of TUL, in the Czech Republic in 2004. Nowadays, sub-micron fibres and nanofibres can be formed from solutions and melts, from various chemicals and material mixtures, from traditional or “green” materials, in a DC or AC field, and with a core-shell structure of two different materials, etc. Similar to the broad spectrum of fibre types, the potential for their application is even broader and includes mechanical engineering, optics, electronics, medicine and drug production, biotechnologies, environmental protection etc.

The development of electrospun fibres and membranes based on natural renewable materials for energy and environmental applications is still underway. “Green” electrospinning technology relies on the development of non-toxic, inexpensive and environmentally friendly polymers and solvents (such as water or ionic solvents) for the fabrication of electrospun fibres and membranes. Electrospun natural biodegradable polymers have shown great applicability in many fields such as the development of filtration membranes, catalytic nanofibres, tissue engineering, drug delivery and sensors [140, 141]. The application of electrospun nanofibres in the area of biomedicine, drug delivery, tissue engineering, wound dressing, water purification, and energy has been reported [142-144]. Electrospun nanofibres from natural tree gum based bio-polymers are an innovative group of economic and environmentally friendly membranes for the potential applications in energy, environmental and biomedical fields.

Natural biopolymers such as Chiton, chitosan, gum tragacanth, and guar gum blended with synthetic biocompatible polymers (PVA or PEO) have been reported to be suitable for producing electrospun membranes [145-150]. Very recently, natural tree polymers such as GA, GK, and KG have undergone electrospinning and the system and process parameters have been standardised to produce smooth and uniform nanofibres [151-155]. Furthermore, many combinations of tree gums have been blended with PVA or PEO and the solubility of the polymer has been determined to subsequently improve the spinnability, solubility, biocompatibility, biodegradability, and mechanical properties of the electrospun membranes. In addition, chemical modifications of natural polymers, with DDSA or Ag NPs to develop the antibacterial membranes have also been reported [152].

Electrospun fibres and membranes of GA, GK, and KG

As reported by Padil et al [151, 152, 154, 155], aqueous PVA (10-12 wt. %) was mixed with GA, GK, and KG solutions (varying from 2 to 5 wt. %) in different weight proportions of PVA or PEO with GA, GK, and KG (100/0, 50/50, 60/40, 70/30, 80/20, 90/10 and 0/100), to determine the good spinnability and uniform size of nanofibres after electrospinning. The electrospinning is carried out on a Nanospider electrospinning machine (Elmarco, NS IWS500U, Liberec, Czech Republic) with interchangeable electrode systems, working with both water or non-water soluble polymers. The details of the electrospinning conditions are as follows: spinning electrode width of 500 mm, effective nanofibre layer width of 200-500 mm; spinning distance of 130-280 mm, substrate speed of

0.015–1.95 m/min, voltage of 0–55 kV and process air flow of 20–150 m³/h. The ratios of the best combinations of the blend mixtures of gum were as follows: PVA was found to have a 30:70 to 10:90 ratio of gum/PVA in the final electrospinning mixture. The SEM and TEM images of GA, GK, and KG with their electrospun fibres and plasma treated fibres are presented in Figures 5, 6, and 7, respectively.

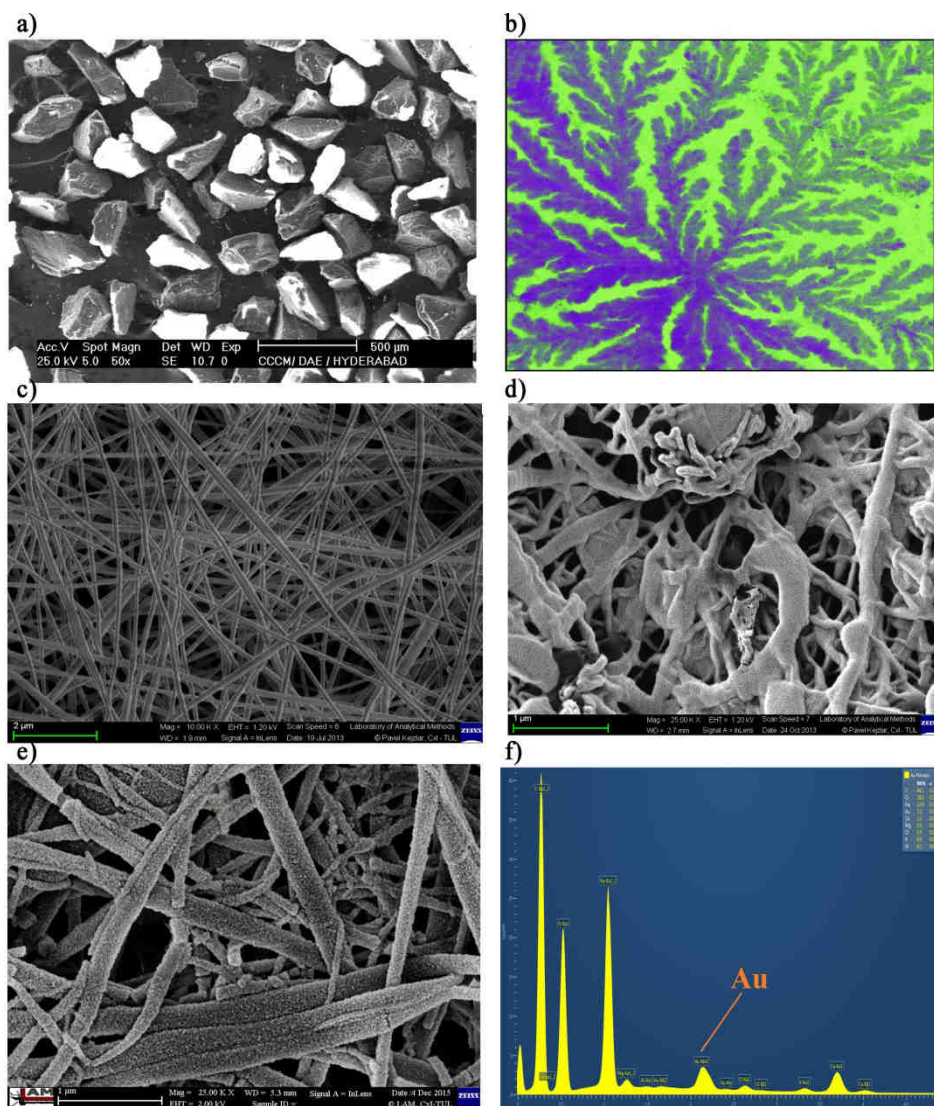


Fig. 5. SEM photographs of KG showing irregular particle sizes (a); TEM analysis of KG showing network molecular structure (b); electrospun fibres of KG (c); plasma treated membranes of KG (d); Au NP adsorbed on fibres of KG (e); EDX analysis of Au adsorbed structures of KG (f)

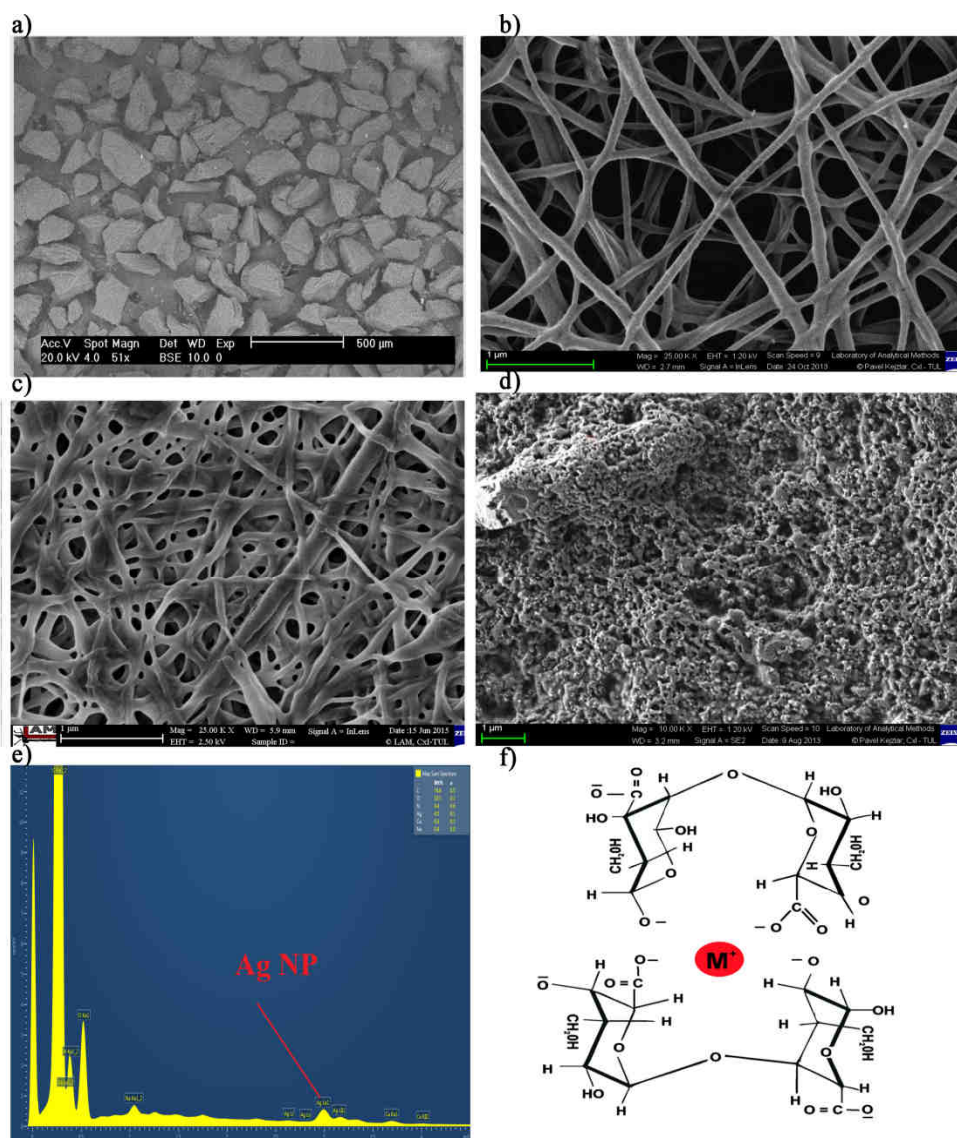


Fig. 6. SEM picture of GK showing the irregular particle sizes (a); electrospun fibres of GK (b); plasma treated membranes of GK (c); Ag adsorbed membranes of GK (d); EDX analysis of Ag NPs adsorbed membrane (e); schematic interpretation of metal ion interaction with GK structure (f)

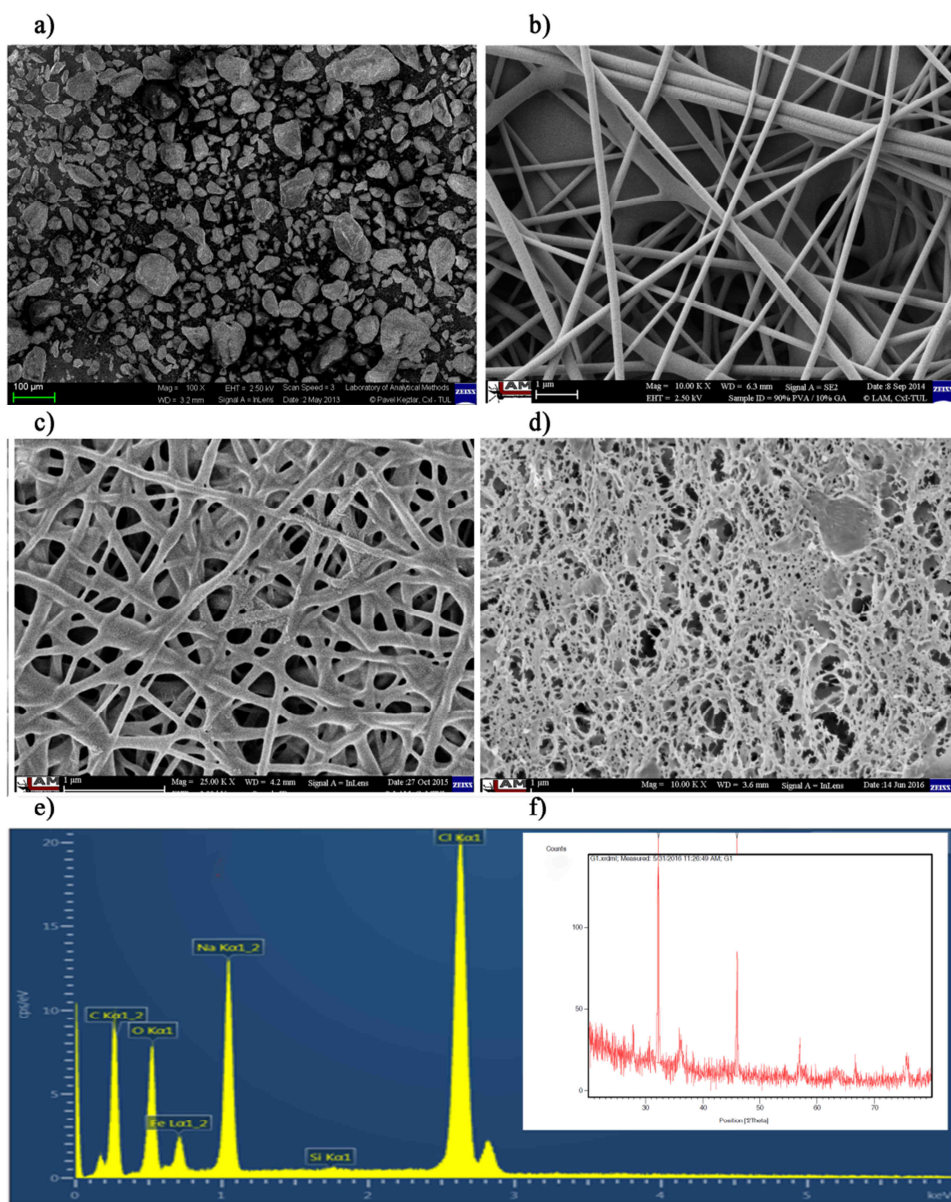


Fig. 7. SEM pictures of GA indicating the different range of particles sizes (a); electrospun membranes of GA (b); plasma treated membranes of GA (c); Adsorption of Fe_3O_4 onto GA electrospun membranes (d); EDX analysis showing Fe traces on the GA- Fe_3O_4 membranes (e); XRD patterns of Fe_3O_4 observed in GA- Fe_3O_4 membranes (f)

Membrane treatments and characteristics

Plasma treatment

Plasma-surface modification is an efficient and cost-effective surface treatment technique used in biomedical research such as sputtering and etching, implantation, deposition, polymerization, spraying, and laser plasma deposition [156]. The unique advantage of plasma modification is that the surface properties and biocompatibility can be significantly altered, while the bulk qualities of the materials remain unchanged [157]. Various gas plasma treatment methods (oxygen, argon, and methane) have been developed to modify the surface properties of polymers, such as the balance of hydrophilicity/hydrophobicity and surface free energy [158]. Typically, plasma treatment modifies the surface by grafting hydroxyl (–OH), carbonyl (–CO), and carboxylate (–COOH) groups [159–162]. Through the processes of plasma modification, it is possible to influence the change of the wettability and water contact angle either to strongly hydrophilic or more hydrophobic, depending upon the nature of the plasma used for the modification and the plasma treatment time [162–165]. As it is stated in the literature, the treatment of membranes using various gas plasma treatment methods such as oxygen, argon, nitrogen has been developed to modify the hydrophilic surface properties of polymers [165, 166]. However, methane or sulphur hexafluoride plasma treatment on various polymeric membranes is used to enhance the hydrophobicity of the polymer surfaces, resulting in higher contact angles [167, 168].

The GA, GK, and KG fibres were treated with methane plasma to improve their hydrophobicity, stability, water contact angle and surface area [151, 152, 154]. The methane plasma treated membrane was prepared in a 13.56 MHz radio frequency (RF) plasma reactor (BalTec Maschinenbau AG, Pfaffikon, Switzerland). The plasma chamber was thoroughly purged with a continuous flow of the gas used during the treatment to reduce trace amounts of air and moisture. During the treatment, the gas flow was adjusted in order to keep a constant pressure of 20 Pa inside the chamber. The plasma conditions and process parameters were as follows: voltage of 300 V; power 20 W; time of 5 minutes; plasma gas purity of 99.997%; electrode area of 48 cm²; inter-electrode distance of 50 mm, and chamber volume of 1,000 cm³.

Cross-linking and stability of electrospun fibres

Heat and plasma treatments were conducted on the fibres and membranes for their application in environmental remediation and antibacterial fields. The GA, GK, and KG membranes are heated to 130–150°C and their stability was tested against an acid, alkaline and aqueous solution, and it was determined that all of the heat-treated membranes were stable up to 90–95% under all of these tested conditions [151–154]. Subsequently, the water contact angle (θ), membrane thickness, porosity, apparent density, BET surface area and degree of stability were found to have increased after methane plasma modification [151–154]. Figure 5 presents the morphological changes that occurred due to the plasma treatments on the tree gums.

Applications of electrospun nanofibres for environmental bioremediation

The simplicity of the fabrication scheme, the diversity of materials suitable for use with electrospinning, as well as the unique and interesting features associated with electrospun nanofibres, all make these techniques and their resultant structures attractive for various applications such as filtration, drug delivery, sensor devices, tissue engineering scaffolds, wound dressing etc.

Nanotechnology for water remediation will play an increasingly crucial role in water security and consequently the food security of the world. Nanoscale filtration techniques, the adsorption of pollutants on nanoparticles and the breakdown of contaminants by nanoparticle catalysts are the major applications of nanotechnology in the clean-up of contaminated water [8-10]. The advantages of electrospun nanofibre membranes are that they can offer both adsorption and filtration characteristics. Therefore, nanofibre membranes offer an attractive solution of heavy metals removal. In this context, green electrospun fibres and membranes are emerging and innovative materials for environmental bioremediation application.

Natural gums for the removal of toxic metal ions

Heavy metals (Pb, Ni, Cu, Zn, Cr, As, Hg, As, and U) are a serious biological problem in aquatic systems. Adsorption and filtration are the commonly used methods for removal of these contaminants from the water. In a recent study, gums have been successfully used for the removal of various toxic heavy metals and radioactive and industrial effluents [108-112, 139, 169, 170-175]. The experimental results showed that sorption, ion-exchange, functional group interactions, modified surface properties, and high surface areas were the possible mechanisms for the adsorption of toxic metals onto gum structures [108-112, 139, 169, 170-175].

Electrospinning gum nanofibre for the removal of nanoparticles

There are potential environmental and health impacts of engineered nanomaterials due to the increased presence of nanomaterials in commercial products. Currently, most sectors of nanotechnology are developing with no guidelines and in an environment ideally suited for entrepreneurship. The lack of disposal of such products will lead to potential contamination of water and ecosystems [176]. Nanomaterials are emerging contaminants in water and show significant toxicity to living systems. Further studies are still in progress on the fate, transport, and transformation of nanoparticles and nanocomposite bioavailability and exposure of humans and other living species [5, 7, 177]. Electrospinning polymers, with various functionalities and combinations of different types of natural and synthetic polymeric materials and their potential applications for the removal of toxic heavy metals from water, have been comprehensively reported [18].

The electrospun membranes of GA, GK, and KG were effectively used for the removal of metal and metal oxide nanoparticles from water [58, 151, 155]. The nanofibres were treated by methane plasma to improve their physicochemical properties and resulted in high adsorption capacities towards nanoparticles (Ag, Au, Pt, CuO, and Fe₃O₄) in an assessment with untreated membranes [58, 151, 155]. The adsorption capacities of the membrane for the removal of nanoparticles (NPs) from water diverge in the order Pt>Au>Ag>CuO>Fe₃O₄ [58, 151]. Figures 5 (e and f), 6 (d and e) and 7 (d and e) show the

nanoparticles (Au, Ag, and Fe_3O_4) adsorbed onto electrospun fibres. Various adsorption mechanisms such as sorption, functional group interactions, complexation reaction between metal/metal oxide nanoparticles with various functional groups present in nanofibre and modified surface properties such as the balance of hydrophilicity/hydrophobicity, surface free energy, and high surface area of the plasma treated membrane are the possible mechanisms of NPs adsorption onto nanofibres [58, 151].

Natural gum functionalized nanoparticles and nanofibres for anti-bacterial applications

Many antimicrobial agents, such as metallic/metal oxides (Ag, Au, Cu, and CuO), organic agents (quaternary ammonium and DDSA) functionalized with natural polymers (chitosan, tree gums (GA, GK, KG, GT, and GG) and cellulose), have been used in textiles and membranes for bio-medical and antibacterial applications [26, 30, 57, 118, 124, 126-130, 132, 133, 135, 137, 138, 146, 152, 153, 172, 177].

Vinod et al studied the electrospun membranes of GK and KG and both of their nanoparticles (Ag and CuO) and DDSA functionalized membranes were tested for their anti-bacterial efficiencies [26, 128, 152, 177]. The electrospun membrane was cross-linked by heating to 130°C and later by methane plasma treatment in order to improve its antibacterial efficiency [152, 128]. The anti-microbial activities of the fibres and membranes of natural gums were investigated against Gram-negative *Escherichia coli* and *Pseudomonas aeruginosa*, and Gram-positive *Staphylococcus aureus*. It was found that plasma modification enhances the antibacterial properties of the membrane in comparison to unmodified samples [152]. Figure 8 shows the plasma treated fibres of GA, GK, and their antibacterial properties. This study suggests that plasma modified electrospun membranes may be used as antimicrobial membranes in food, water and environmental applications. Furthermore, natural gum functionalized nanoparticles and DDSA derivatives of these gums provide enhanced antibacterial properties and are an attractive approach for the fabrication of such nanomembranes via electrospinning for applications in the food, pharmaceutical and other industrial fields.

Conclusion and prospects

The development of an ecologically viable electrospinning process using tree based carbohydrate polymers to produce nanofibres in “green” pathways has been explored. The current development in “green” electrospinning technologies provides important clues to the potential roles of these electrospun membranes in water purification and antibacterial applications. The use of tree based polymers in various applications is relatively rare compared to synthetic polymers due to the incompatibility of the choice of the polymer for particular applications and in some cases due to poor chemical and mechanical properties. Nevertheless, further developments are required to find new functionalized hybrid polymer systems based on natural and synthetic polymers, which are suitable for electrospinning with improved functionalities for various applications. Based on the current studies, it is in no doubt that electrospun nanofibres based on natural hydrocolloids are expected to play a significantly vital role in the future in many important applications, such as water purification, renewable energy, scaffolds in tissue engineering, food, biotechnology, and environmental protection.

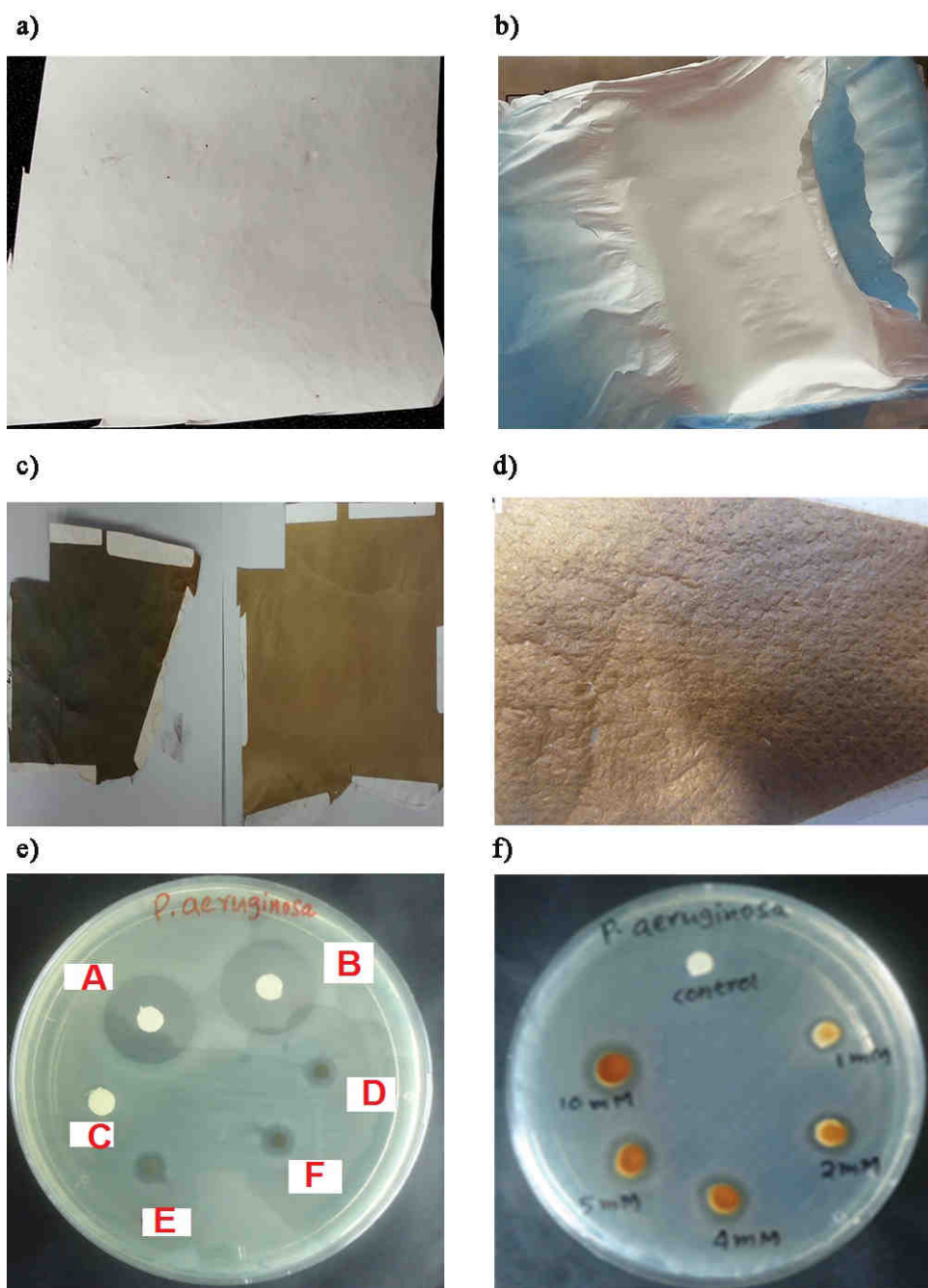


Fig. 8. Electrospun fibres of GA (a, c, and e) and GK (b, d, and f) with their corresponding methane plasma treated membrane showing potential antibacterial efficiencies. A, B (both contain 10 mM of Ag/GA), C is the control, and D, E and F contain 1 mM of Ag/GA fibre; f) Ag/GK plasma treated membrane containing Ag from 1 to 10 mM

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