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REVIEW

Efficient Au nanostructures for NIR-responsive controlled drug delivery systems

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Abstract

Different photo-thermal agents such as gold nanostructures with various shapes and sizes including nano-rods, nano-cages, nano-shells exhibit tunable optical properties, surface chemistry, resonance frequency and non-toxicity. Furthermore, gold nanoparticles (Au NPs) also utilized for diagnosis purposes and drug carriers for thermo-therapy of biological cells targets and potential bio-medical applications against various cancer treatments. To control complications of conventional cancer therapeutics, Stimuli-assisted drug delivery systems (DDS) together with interior and exterior Stimuli-assisted prompts have received particular interest by the scientific community around the globe. Among both of them, external stimuli-responsive parameters provide more controlled DDSs that circumvent individual discrepancy. Many researchers reported that light-responsive DDSs manifest interesting features because of proficiency and preferable spatiotemporal control. Among them, near-infrared light-responsive DDSs are potential candidates that follow various mechanisms such as photo-thermal effect, two photon absorption, and up converting nano-particles. This review presents an over view of different morphologies of Au nanostructures boosting the efficiency of DDSs by utilizing the phenomenon of surface plasmon resonance by overcoming the major challenges.

Keywords Au plasmonics · Stimuli-responsive drug delivery systems · Spatiotemporal control · Photo-thermal · Cancer treatments

Introduction

Cancer is significantly attracted crucial health problems and is the second dominant causal agent of death around the globe (Dai et al. 2016; Siegel et al. 2016, 2017). Akhtar et al. have reported that nearly 7 million new patients were predicted by 2030 (Akhtar et al. 2014; Sutcliffe 2012). The conventional therapeutics including chemotherapy and radiotherapy were not contemplated sufficient due to inadequate explicitness and toxicity (Chen et al. 2016; Cho et al. 2015; Liu et al. 2013; Schmaljohann 2006). In recent few years, nano-medicines and drug delivery systems have attracted researchers due to its wide utilization in diagnostic tools and controlled drug delivery in various drugs (Patra et al. 2018). During ancient times, various drug delivery systems used plant-based natural products for the treatment of various chronic diseases such as cancer, diabetes, cardio-vascular, inflammation, and microbial diseases due to significant advantages including extraordinary chemical diversity, chemical and biogenic effectiveness along-with macromolecular specificity and non-toxicity and side-effects, low-price, and unique therapeutic efficiency. However, these drug delivery systems present some drawbacks such as in-vivo instability, insufficient bio-availability, and inefficient solubility, less-absorption in the body, problems in target-specific deliveries, and tonic-effectiveness, and major drawbacks of drugs. To overcome these challenges, new DDSs has been introduced for targeting drugs to specific sites (Jahangirian et al. 2017; Martinho et al. 2011). Consequently, in the recent few years, nanotechnology has been emerged significantly due to its potential applications utilized in diagnosis, drug formation, controlled drug delivery with great success and cure of diseases. Advancement in

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nanotechnology offers new nano-materials that have promising applications in bio-medical domain (Kong et al. 2017). Various nano-materials such as liposomes, poly-meric micelles, graphene, CNTs, quantum dots, ferro-ferric oxide nanostructures and metallic nanostructures were utilized in drug delivery, imaging (Iqbal et al. 2020a, b), and stable therapeutic treatments (Ajnai et al. 2014). As compared to other nanostructures, different plasmonic nanostructures manifest unique photo-thermal properties (Iqbal and Afsheen 2016b) that has been utilized to improve DDSs. Plasmonic nanostructures present photo-thermal effect that exhibits phonons which transforms light energy into vibrations in crystal structures. These captivated photons converted into phonons through frequent electron–phonon mitigation, due to phonon–phonon mitigation, causes enhancement in the temperature of the system and its environment through conduction (Kuppe et al. 2020), so generating sectional heat. These nanostructures tend to transform NIR light to heat adroitly as compared to photo-thermal dyes due to SPR effects (Vempati et al.2015). Giljohann et al. have investigated that among other plasmonic nanostructures, gold nanostructures manifest promising bio-medical applications due to its unique features (Iqbal et al. 2020a, b) such as it is chemically and biologically stable, less cytotoxic in organic surroundings, and its availability as surface functionalization with distinctive biogenic ligands (Giljohann et al. 2010). On the other hand, silver nanostructures exhibit unique features including substantial plasmonic resonance, optical properties that's why it is utilized as photo-thermal light-to-heat conversion transducers (Dos Santos et al. 2014; Wei et al. 2015). Furthermore, Akter et al. have reported that silver and copper nanostructures exhibit high toxicity for in-vivo applications and manifest low stability as compared to gold (Akter et al. 2018; Lee et al. 2018a, b; Liao et al. 2019; Zhang et al. 2018a, b). Kim et al. have studied that these plasmonic nanostructures have been utilized in clinical practices but still need more research and innovations to upgrade bio-compatibility and stability in substantial surroundings. Hence, it is concluded that Au nanostructures are highly suitable for bio-medical applications especially for photo-thermal practices (Kim et al. 2019). Iqbal and co-workers have reported that those bio-medical treatments which include poly-meric nanoparticles, liposomes, micelles, and so on are used in DDSs due to their improved permeable and retentive nature, higher half-life, easy availability, and good targeting capability using aptamers (Iqbal et al. 2016; Rasheed et al. 2017; Schmaljohann 2006). However, pre-mature and limited drug release at target sites manifests crucial impediments for this type of drug delivery (Ali and Ahmed 2018; Yang et al. 2016). To overcome pre-mature and limited drug release at target sites, Stimuli-responsive Drug Delivery Systems (SRDDSs) are contemplated as very helpful (Ali and Ahmed 2018; Chen et al. 2016; Cho et al. 2015; Liu et al. 2013; Liu et al. 2017a, b, c; Schmaljohann 2006; Yang et al. 2016). In these systems, a particular stimulus controls the drug release. SRDDSs are utilized easily when needed so it also called intelligent or smart DDSs (Hosseini et al.2016). Ding et al. have reported that the mechanism of Stimuli-responsive DDSs, that accurately controls release functions which are based upon relevant stimuli-signals and their release processes. These stimuli signals are mainly classified in to two types including internal and external stimuli. Intramural stimuli including pH, Temperature, Hypoxia, Enzymatic process, and Glutathione concentrations. The significant environmental variations between normal and tumor tissues (Bae et al. 2003; Bhatnagar and Venuganti 2015; Hosseini et al. 2016) becomes the cause viciousness in pH, redox condition, nature of bio-molecules, because tumor tissues exhibit higher temperature, acidic pH, higher concentrations of GSH and over-expression of particular enzymes as compared to normal tissues. These internal stimuli signals act as ideal activators due to these instinctive gradients to control drug release system and upgrade its precision against tumor tissues (Ding et al. 2016). However, external stimuli include Light, Magnetic field, Electric field, and Ultrasound-based DDSs that exhibit controlled drug release and avoid separate variability in contrast to internal stimulus parameters (Liu et al. 2013, 2016; Wang and Kohane 2017; Yang et al. 2016). Out of above mentioned extrinsic-stimuli, Light is contemplated as an interesting extrinsic-stimulus because of proficiency and preferable spatio-temporal commands (Sahu et al. 2018). Furthermore, due to extra stimuli along-with internal stimuli at the infected area, it provides précised control over the Drug delivery systems. So SRDDSs are significantly utilized to target selective tumor tissues with less toxicity (Ali and Ahmed 2018; Bhatnagar and Venuganti 2015; Liu et al. 2017a, b, c). Hang et al. have investigated that drug delivery systems that can work under specific stimulus also have some drawbacks such as inter-individual discrepancy, uncontrolled drug liberation, and composition problems of these systems (Hang et al. 2017; Liu et al. 2016). Several light-responsive DDSs have been developed in recent years. Most of them, work under UV light which demonstrates some limitations such as less penetration power andhigh toxicity to tissues. Owing to major limitations, UV-based DDSs were not able to be utilized for clinical applications (Yang et al. 2016). However, near infrared (NIR) light ($\lambda = 650–900$ nm) manifests interesting features such as better penetration into tissues because of finite attenuation and toxicity (Li et al. 2017; Rahoui et al. 2017; Yang et al. 2016). Thus, NIR-assisted DDSs are dormant couriers for targeted drug release that is helpful for clinical applications (Mura et al. 2013). These DDSs release drugs when exposed to NIR light which is due to photo-thermal effect. Due to photo-thermal effects temperature increases. High

temperature causes cell death which alliances drug operation (Noh et al. 2015; Wang et al. 2016a, b). In recent decades, different plasmonic nanostructures (Afsheen et al. 2020) have the ability to convert NIR light to heat efficiently in contrast to photo-thermal dyes due to SPR effects (Iqbal et al. 2019a, b). The binding of anti-bodies and other biological macromolecules also makes it an efficient candidate for treatment of various diseases (Villalba-Rodriguez et al. 2017; Yang et al. 2017). Among all of them, Gold (Au) nanostructures have engrossed great attention because of its distinctive features including quantum size effects, SPR, higher catalytic-activities and self-assemblies (Afsheen et al. 2019; Barrow et al. 2012; Daniel and Astruc 2004; Elghanian et al. 1997; Ijaz et al. 2020a, b; Mirkin et al.1996; Turner et al. 2008). Ciganda et al. have investigated that synthesis of Au nanostructures takes a shorter time as compared to other plasmonic nanostructures (Ciganda et al. 2016; Leng, Pati and Vikesland 2015). Mostly Sajanlal and co-workers have studied that Gold nanostructures including Au- nano-wires, Au- nano-plates, Au- nano-prisms, etc. exhibits various shapes and sizes due to addition of various stabilizing agents (An and Somorjai 2012; Huang et al. 2011; Sajanlal et al. 2011; Sun and Xia 2002; Tao et al. 2008) that significantly affects the Au nanostructures optically, mechanically, electrically, and chemically also (Lee et al. 2018a, b). In recent few years, it may be considered a new manifesto to demonstrate interesting applications in various areas especially in medicine (Dhar et al. 2009; Dreaden et al. 2009, 2012; Dykman and Khlebtsov 2012; Giljohann et al. 2009; Kim et al. 2010; Wang et al. 2005) as presented in Fig. 1.

Au nanostructures have been extensively reported as a potential candidates for various bio-technological applications such as therapeutic diagnosis, drug delivery (Cho

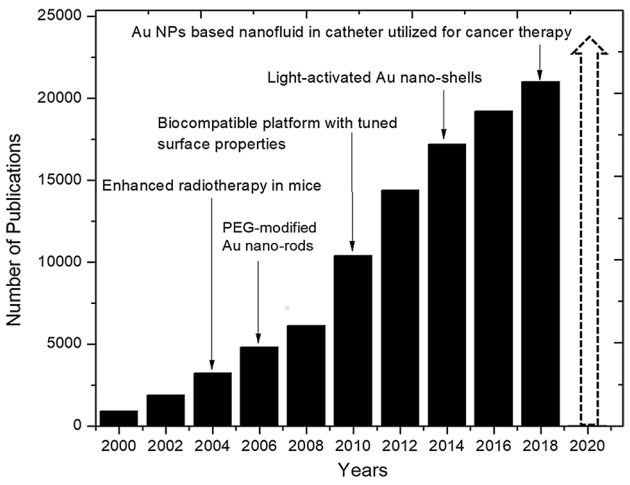


Fig. 1 Yearwise contribution and stepwise development of Au nanostructures in cancer treatment extracted from scholar.google.com

2010; Ajnai et al. 2014), tumor treatment, and cure of diseases. In 1857, colloidal gold was produced by utilizing phosphors to reduce gold chloride (Faraday 1857) and it showed red color which manifests the presence of smaller sized Colloidal Au molecules in it. Turkevich and co-workers (Turkevich, Stevenson and Hillier 1951) worked on reducing agents of colloidal gold such as $Na_3C_6H_5O_7$. In recent few years, interrelation between light and Au nanostructures have peaked interests (Bohren and Huffman 2008; Kerker 2013; Kreibig and Vollmer 2013; Link and El-Sayed 1999a, b, 2000, 2003; Papavassiliou 1979) due to various distinctive features. These metallic nanostructures showed efficient absorption in the visible-region because of strong resonance between metal free electrons and frequency of visible light which is known as Surface Plasmon Resonance (SPR) (Afsheen et al. 2019; Mohsin et al. 2016a; Bohren and Huffman 2008; Kerker 2013; Kreibig and Vollmer 2013; Link and El-Sayed 1999a, b, 2000, 2003; Mie 1908; Papavassiliou 1979). These resonant frequencies strongly depend upon the type, morphology, size and dielectric constant of the medium which surrounds them. The dielectric constant of the surrounding tools has the ability to tune optical properties of Au nanostructures. Furthermore, an increase in size gives effective absorption of surface plasmons and clusters of Au nanostructures that manifests SP absorption towards red-shifts (Xiaohua 2008). Au nanostructures can be easily prepared by chemically reduced chloroauric acid ($HAuCl_4$). It manifests various shapes such as spherical, rod-shaped and cage-like etc. and core sizes approximately ranging from 1 to 150 nm. Au nanostructures exhibit negative charges, so they can offer a functionalized platform to all types of bio-molecules like drugs, genes, and targeting ligands (Fratoddi et al. 2015). Au nanostructures can be simply altered with controlled dispersion. The surface of Au nanostructures is safe, stable and can be modified easily by adding different substances or binding particular receptors conjoined with different kinds of therapeutics (Cobley et al. 2011; Libutti et al. 2010) to form a particular monolayer to extend its firmness and enhance the distribution in organic-medium, and for further confederations of attacking drugs (Love et al. 2005) on cancerous cells by either active or passive targeting systems (Daniel and Astruc 2004; Gao et al. 2004). Moreover, the shape is a key factor for cellular uptake of Au nanostructures. The shape and size remarkably affected Au-NPs optically and electrically (Verissimo et al. 2016). Tong and co-workers reported that optical properties examined by size of Au nanostructures when they absorb UV light (Tong et al. 2009). Furthermore, Au nanostructures exhibit various distinguishing features (Iqbal et al. 2019a, b) such as biocompatibility (Hainfeld 2006), chemical inertness (Yang et al. 2017), extremely small size, macroscopic quantum

tunneling effect, large-scale production, high optical absorption coefficient, the existence of SPR bands (Kumar et al. 2013) and tunable surface properties as illustrated in Fig. 2.

The conjugates of Au nanostructures also exhibit significant properties like enhanced binding affinity, systematic delivery and selective targeting to specific tissues (Dreaden et al. 2011). Recent work reported that there is an extensive range of research and nanotechnology-based treatments under development for cancer (Cobley et al. 2011; Libutti et al. 2010). The transport of Au nanostructures-based conjugated drugs present elevated perforation rate to target tumor tissues which lead to reduce anti-tumor drugs and make it lesser malignant to normal cells with negligible side-effects.

Different morphologies of Gold (Au) nanostructures

Au nanostructures with various morphologies (Champion et al. 2007) and sizes (Jiang et al. 2008) manifest interesting features to enhance the mechanism NIR-light-responsive drug delivery systems due to photo-thermal effect. Chithrani et al. have reported that smaller sized Au NRs provides a higher up-take area to the cells as compared to larger sized Au NRs (Chithrani et al. 2006).

Gold nano-rods

Gold nano-rods (Au-NRs) emerging significantly in recent decades because of their promising Photo-thermal and NIR-responsive applications especially in chemical monitoring, biogenic-tomography, drug delivery and photo-therapeutics (Murphy et al. 2011). In recent years, Au NRs are widely utilized for in-vitro and in-vivo photo-thermal treatment of cancer due to their efficient absorption of NIR light. Wang et al have reported that Au NRs which are used to ablate cancerous tissues are garnering significant attention now a days (Wang et al. 2012, 2013). In 2006, El-Sayed and his co-workers first time utilized Au-NRs in vitro studies (Dickerson et al. 2008). Different strategies have been developed to target these Au NRs because of various surface modification techniques (Wu et al. 2013). Cho et al. (2014) have studied conjugated Au NRs specifically for anti-epidermal growth factor receptor (EGFR). Zhang and co-workers investigated the applications of photo-thermal properties of modified Au NRs due to (EGFR) monoclonal anti-body. EGFRmAb Au-NRs have been developed by EGFR monoclonal anti-bodies (mAb) on Au NRs but modified Au NRs have same optical properties after coating. To examine apoptosis ability of these modified Au NRs for in-vitro and in-vivo photo-thermal therapy, experiment has been performed on Hep-2 and in-vivo experiment demonstrated that modified Au NRs strongly ablate Hep-2 cells under laser light. These surface modifications basically increase the up-take of Au NRs into Hep-2 cells and increase the apoptosis rate as compared Au NRs without modifications. These modified Au nanostructures were also utilized in EGFR-expressing bladder cancerous tissues (Ahmad et al. 2016a, b). Different biological sites were used to target drugs such as mitochondria, lysosome, Endo-plasmic reticulum and nucleus but mitochondria rarely utilized for drug targeting in photo-thermal therapy. Ju et al. have found that various hydrophobic drugs loaded with nano-carriers such as SiO_2 encapsulated Au-NRs coated with a cytochrome c-specific binding aptamer to target mitochondria of cancerous tissues using chemo-thermal therapy. These nano-carriers loaded hydrophobic drugs were used to reduce poisonous effects of drugs and improve the efficient therapeutic applications. After laser irradiation, the drug is released at mitochondrial target sites and causes apoptosis. This DDS used both chemo and photo-thermal therapy to target mitochondrial avenue to kill cancer cells and also helpful to fabricate multi-functional mitochondria as Targeted drug delivery agents for tumor therapeutics (Ju et al. 2014). Cheng and co-workers investigated that electro-spun fibrous membrane coated Au-NRs used to ablate cancerous cells by photo-thermal therapy. Electro-spun membrane is significantly utilized as Au-NRs carrier due to its surgical recovery nature and efficient biodegradability. To achieve selective killing of cancerous cells and post-surgical recovery, these membranes were utilized. Recent work also proposed Au-NRs coated multi-walled CNTs with RGD peptides which were utilized for photo-acoustic imaging of gastric cancer cells. These modified Au-NRs absorb NIR radiations to generate heat and exhibit high potential to target in-vivo gastric cancerous tissues with less damage (Cheng et al. 2014). Bai et al. (2014) have reported

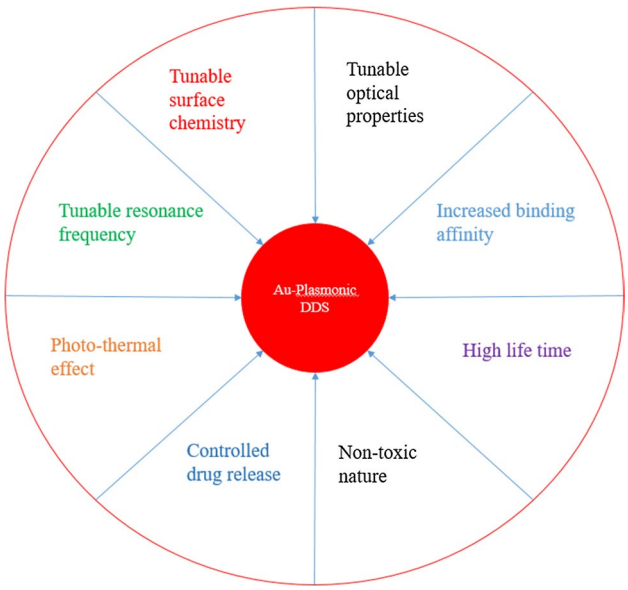


Fig. 2 Distinctive features of Gold nanostructures

another approach to analyze photo-thermal therapy in mice relevant to human tumor xeno-grafts. Coated Au-NRs also utilized to monitor tumor xeno-grafts. In 1992, C. R. Martin synthesized Au-NRs for the very first time (Pérez-Juste et al. 2005) but the stability of Au-NRs is due to coating of cetyl tri-methyl ammonium bromide (CTAB) which was first introduced by Murfey in 2001 (Pérez-Juste et al. 2005). Recently, CTAB has been emerging due to Au-NRs strong dependence and significant bio-medical applications. Many researchers reported that CTAB bounded nanostructures show negligible cytotoxic effects (Connor et al. 2005). To overcome these limitations surface coating is one of the best choices. To reduce the vulnerability of CTAB molecules from Au nanostructures surface to the local environmental cells. Furthermore, complete substitution of CTAB molecules from Au nanostructures surface destabilizes the shapes of Au-NRs which results in the agglomeration of particles with one another. Various surface modifications of Au NRs were developed to enhance their efficiency. Mahmoud et al. have reported that the multi-dentate PEG functionalized Au-NRs for in-vivo NIR-based photo-thermolysis of invasive tissues (Mahmoud et al. 2019). Recently, some researchers successfully developed Au-NRs laminated with carboxylated bovine serum albumin (BSA) to replace CTAB (Kumar et al. 2015). To achieve enhanced photo-thermal therapy, hydroquinone utilized as reducing agent during Au-NRs preparation. Using surface modification techniques stable and low toxicity Au-NRs were developed with length ranges from 30 to 90 nm. These BSA modified Au-NRs exhibit tunable size, broad range of Longitudinal Surface Plasmon Resonance (LSPR), anti-cancer efficiency by in-vivo laser-based photo-thermal ablation and many other tremendous applications in the bio-medical fields. Moreover, mesoporous SiO_2-coated Au-NRs have been used to extract CTAB molecules (Li et al. 2014). Results showed that this method has the ability to completely remove these molecules from SiO_2-coated Au NRs openings without destroying the Au-NRs that were immersed in SiO_2 which enhanced the drug loading ability and proved as useful to kill gastric-cancer cells due to photo-thermal treatment. Liao and co-workers found that the thiol-modified poly-ethylene glycol coatings (Liao and Hafner 2005), while Wei and his co-workers have investigated the poly (N-isopropyl-acryl-amide polymer (Wei et al. 2008). Recently, Vigderman et al. have also reported the complete replacement of CTAB with 16-Mercapto-hexadecyl tri-methyl ammonium bromide (MTAB), that is unable to maintain stability but manifest many promising bio-logical applications (Vigderman et al. 2012). Chen and co-workers have designed Enhanced Green Fluorescent Protein (EGFP) Plasmid DNA along with Au-NRs and utilized to evaluate HeLa Cells (Chen et al. 2006). This study also concluded that suitable surface coatings on Au-NRs enhance important parameters that are utilized in DDSs. Alkilany and his co-workers developed Plasma protein alongwith distinctive exterior coatings, and also described the binding of plasma proteins with the exterior of Au-NRs in contrast to outer-charges and ligands. Results show that these absorbed proteins exhibit negligible impacts on the solidity of appropriately covered NRs (Alkilany et al. 2009). Akiyama et al. have found the poly-ethylene grafting of Au-NRs in mice and have also investigated that high poly-ethylene grafting manifests various advantages reticulo-endothelial (RES) clearance system. Recently many researchers have reported that Au NRs exhibit two bands come from SPR spectrum. Longer wavelength band lies in NIR-region and shorter-wavelength band falls in visible-region due to longitudinal and transverse oscillations of electrons, respectively (Gans 1915; Link and El-Sayed 2005; Link and El-Sayed 1999b, 2000, 2003; Link et al. 1999; Murphy et al. 2005; Nikoobakht and El-Sayed 2003). Moreover, Au NRs exhibit significant sensitivity towards aspect ratio, because the greater aspect ratio increases the SPR absorption wavelength. Despite worthy advantages and higher efficiency as compared to other nanostructures, toxicity of Au-NRs is alarming (Ahmad et al. 2016a, b; Choi et al. 2013; Liao and Zhang 2011).

Gold nano-cages

Gold nano-cages (Au-NCs) have been gained tremendous interest now a days due to significant photo-thermal properties that are utilized in various therapeutic applications (Chen et al. 2007; Au et al. 2008). Large scale production and tunable LSPR peak positions of Au-NCs may be achieved by changing titrated quantity of chloroauric acid during synthesis process (Skrabalak et al. 2007). Chen et al. investigated that the Au-NCs exhibit five times higher NIR-absorption cross-section in contrast to other previously utilized organic dyes including indo-cyanine green (ICG) with up to 40 nm size that may be utilized in in-vivo delivery (Chen et al. 2005). Moreover, significant hollow structures manifest efficient photo-thermal properties with NIR-light which has been utilized for controlled drug-release and drug encapsulation processes (Yavuz et al. 2009). In vitro studies also revealed that Au-NCs conjugated with cancer cells exhibit unique anti-bodies that cause photo-thermal demolition of cancerous tissue when irradiated with low-intensity laser light as compared to other types of gold nanostructures that required high-intensity laser beam. Matsumura et al. reported that in-vivo photo-thermal efficiency of Au-NCs for bi-lateral tumor model. The present work reported a passive targeting DDS that utilized modified Au-NCs. These surface modifications developed when mono-layer of PEG applied on exterior of Au-NCs that permits them to retain themselves for a longer time in bloodstream that can be agglomerated in the tumors because of Enhanced Permeability and

Retention (EPR) effects, but punctured tumor vasculatures consist of wider inter-endothelial junctions and a mal-functioning lymphatic system (Matsumura and Maeda 1986). During in-vivo photo-thermal therapeutics, an increase in temperature provides effective information about tumors. To study non-invasive consequences of photo-thermal therapy on tumor cell metabolic process, fluoro-deoxyglucose positron emission tomography (^{18}F-FDG-PET) has been utilized. When tumor cells present decreased metabolic activities, effective therapy can be observed for those tumor cells which are irradiated with laser light and combined with Au-NCs. Additionally, a bio-distribution study revealed that modified Au-NCs through PEG, enhanced the uptake capacity of Au-NCs by tumors and in peripheral parts of tumor cells, Au-NCs concentration is higher as compared to core that gas been significantly utilized during cancer treatment (Chen et al. 2010). Xia et al. have investigated the suitability of Au-NCs in-vitro tumor treatment applications (Chen et al. 2007). When these Au-NCs are combined with anti-HER2 anti-bodies to kill HER2 type breast cancerous cells. This study also found that the damaged area of tumor cells strongly follows the power density of laser and apoptosis of tumor tissues also significantly dependent upon the irradiation of laser light and their exposure time. After steady-state, exposure to laser light can be dangerous for normal body tissues (Au et al. 2008).

Li et al. have found that the in-vivo active targeting and bio-distribution of Au-NCs have been by utilizing anti-EGFR conjugated Au-NCs and 111 (Melancon et al. 2008). Moreover, the targeted Au-NCs exhibit three times higher tumor empathy as compared to non-targeted Au-NCs (Huang and El-Sayed 2011). The geometry of nano-cage can significantly change the resonance wavelength as presented in Fig. 3 which is highly required for the tuning of operation wavelength for DDS systems. It is, therefore, important

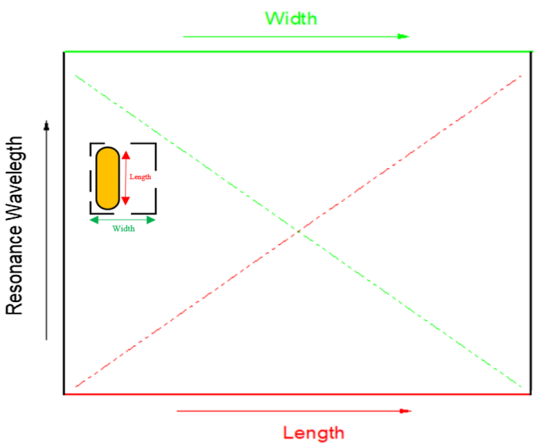


Fig. 3 Effect of geometry of Au nano-cage on resonance wavelength

feature making Au nano-cages applicable in controlled DDS systems with the desired region of light.

Gold nano-shells

The gold nano-shells (Au-NSs)-based NIR-responsive photo-thermal tumor treatment has been investigated by Halas and co-workers in 2003 (Hirsch et al. 2006). These Au-NSs were synthesized and stimulated by PEG so, Au-NSs functionalized with PEG has been used to destroy breast cancerous tissues under NIR-light with intensity of 35 w/cm^2 for 7 min. This study also found that breast cancer xeno-grafted tumors have been killed after Au-NSs intra-tumoral inoculation after few minutes of NIR-laser exposure (O'Neal et al. 2004). These Au-NSs manifest approximately 4 h of blood half-life, retain themselves in liver and spleen of mice and its elimination from the body has been started after 6 months (Lal et al. 2008). Oldenburg et al. have investigated that SiO_2 core Au-NSs with 100 to 200 diameter (Oldenburg et al. 1998). These nano-shells have significant NIR-responsive absorption, scattering (Prodan et al. 2003), and by changing the ratio Au-NSs and SiO_2 core, optical resonance can be changed. As this ratio is smaller, the greater will be the chance for SPR absorption wavelength towards red-shifts (Loo et al. 2004). Various Core–shell nanostructures have been fabricated by depositing Au-NSs on different inner cores including Organic polymers, Magnetic NPs and SiO_2 (Ahmad et al. 2016; Chen et al. 2014; Fu et al. 2013; Goodman et al. 2014; Phillips et al. 2014). It has been reported that Au coated iron-oxide core has been developed due to attractive photo-thermal properties and multi-modal imaging (Chen et al. 2017; Hu et al. 2013; Kirui et al. 2013). These studies also found that the less toxic exterior of iron-oxide's core due to presents Au-NSs in it that have been attracted for promising therapeutic applications while enhancing the efficiency of these core–shell nanostructures by managing their physico-chemical attributes. Chen et al. synthesized Au-NSs along with iron-oxide nanostructures or SiO_2-core doped with indo-cyanine dye. These Au-NSs showed effective photo-thermal therapy and provide contrast images using Fluorescence and Magnetic Resonance Imaging techniques (Chen et al. 2014a, b). Similar studies also found that Au-NSs has been prepared by utilizing DOX-loaded mesoporous SiO_2-core. These Au-NSs exhibited higher cellular-uptake and improved photo-thermal therapeutics of Ehrlich Carcinoma in in-vivo analysis due to chemo-photo-thermal therapy under on NIR-light. This in-vivo evaluation studies committed the localized passive targeting at tumor sites that transform NIR-light into heat and photo-thermal heat significantly triggers the drug release efficiency to kill tumor cells. Moreover, those tumor cells that do not exhibit strong acidic extra-cellular matrix required smooth drug emancipation so DOX-loaded Au-NSs has been considered

as eminent photo-thermal transformation conversion factor that instigates sustained drug liberation (Elbialy et al. 2014). Chen et al. investigated that photo-thermal properties can also be used to destroy tumor-based lymphatics. The statistical results of this study also manifested that Au-NSs-assisted photo-thermal heating has been significantly utilized to kill tumor cells with greater efficiency when irradiated laser in contrast to other cancer therapeutics (Chen et al. 2014a, b). Topete and co-workers have also fabricated branched Au-NSs with Dox-loaded organic core by seeded-growth surfactant less technique. To target tumor cells, folic-acid has been covalently bonded with albumin or indo-cyanine green to provide HASICG-FA that specifically targets tumor sites and enhanced fluorescent imaging ability with efficient multi-modal capacity to kill cancer tissues under exposure of laser light. These studies revealed that the surface of Au-NSs exhibits photo-thermal effects to kills tissues but dye mediated photodynamic effects also been demonstrated due to presence of dye and release of Dox at the same time under laser light (Topete et al. 2014).

Gold nano-spheres

Spherical Gold nanoparticles (Au-NSs) are garnering great interest now a days due to its extensive use in promising applications of DDSs. The solution of chloroauric acid with sodium citrate have been utilized to synthesize Au-NSs. This study also demonstrated that by changing the stoichiometric-ratio of both precursors, the size of these nanostructures can be controlled (Frens 1973). The radius of the gold nano-spheres can be used to adjust the resonance wavelength as presented in Fig. 4 which is also a challenge for the DDS systems. Recent studies have been reported so indicating the improvements in DDS systems due to insertion of plasmonic Au-NSs. In 1951, Dr. Turkevich successfully published synthesis technique of Au-NSs for very first time (Foss et al. 1992). Lin et al. have investigated the contrast agents of Au-NPs by utilizing Au-NSs under visible pulsed laser light (Pitsillides et al. 2003). Under laser light, anti-CD8 immuno Au-NPs with efficiently bound to T- lymphocytes and destroy them completely. After visible light photo-thermal therapy, Zharov and his co-workers have found use of pulsed laser photo-thermal treatment of cancer cells (Zharov et al. 2005). These studies have been investigated that nanosecond pulses of up to 15 nm sized Au-NPs with energy ranges from 2 to 3 J/cm^2 cause cell death. El-Sayed and co-workers investigated that the Au-NPs have been also utilized for cancer cells therapeutics under visible photo-thermal therapy using argon-ion laser (El-Sayed et al. 2006). These visible photo-thermal therapeutics has been received significant importance in in-vitro studies, but it has restricted in-vivo feasible applications due to low penetration in cancer tissues. For in-vivo clinical treatments of tumors within tissues, NIR-light is required due to its deeper penetrating power because of minimum absorption of Hemoglobin and H_2O molecules in cells. Thus it is reported that the NIR-responsive plasmonic nanostructures have been significantly emerging nowadays in tumor therapeutics because of distinctive photo-thermal characteristics (Huang and El-Sayed 2011). Similar studies also revealed that size of nanostructures has significant importance during efficient designing of Targeted DDSs so to control particle size, the damaging effects may be reduced. Recently, many researchers investigated that smaller sized Au-NPs interfused with therapeutic peptides such as PMI and selected peptides like CRGDK on the exterior side of gold nano-particles. When Au-NPs become targeted with various concentrations of CRGDK moieties, the internalization of Au-NPs has been improved efficiently. Moreover, these smaller sized Au-NPs manifest strong penetrating power within nucleus, which significantly enhanced the efficiency to target breast cancer cells (Kumar et al. 2012). Huang et al. have reported that approximately10 nm sized Au-NPs has been penetrated into the nucleus of the cell, while bigger sized particles have been accumulated in the cytoplasm (Huang et al. 2012).

Additionally, ultra-small NPs efficiently supplied anti-cancer Drugs including Dox within the nuclear core of cancerous tissues, that why these are the potential candidates that have been utilized in efficient DDSs for resistant tumors therapeutics (Zhang et al. 2011). Arosio et al. have found that those Au nanostructures operationalized with cyclic-RGD peptides exhibit unique properties to diagnose breast cancerous cells (Arosio et al. 2011).

Other morphologies of Au NPs

In 2006 Yamamoto and co-workers along with Hafner's synthesized gold nano-stars (Au-NSs) for the very first time (Nehl et al. 2006), attained significant acceptance due to SPR usage in photo-thermotherapies. To accomplish

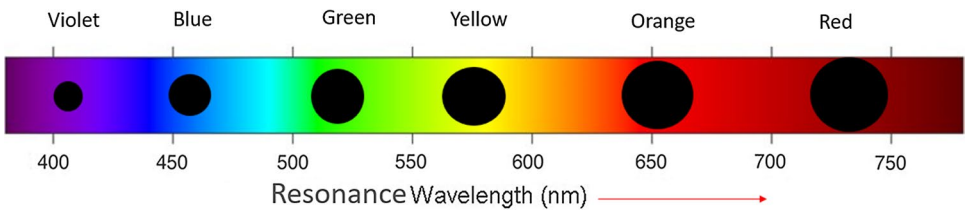


Fig. 4 Effect of radius of Au nano-spheres on resonance wavelength

healthier performance and low harmful effects, many other morphologies of gold nanostructures are garnering great attention now a days. Gold nano-stars also utilized for photo-thermal therapy due to unique plasmonic features which significantly convert NIR radiations to heat energy. Due to smaller core and numerous slim branches of Au nano-stars, it shows higher absorption cross-sections with better tunability in NIR region and less scattering effects. Yuan et al. have reported significant tomography and photo-thermal ablation in mice experiments (Ndokoye et al. 2016). Nie et al. have reported that Au nano-stars have widely utilized for in vivo volumetrical photo-acoustic molecular angiography and restorative examination (Nie et al. 2014). In this case, RGD-peptides conjugated with Au-NSs to kill significant integrin (v3) on cancerous neo-vasculature and extremely susceptible angiography and photo-thermal therapy can work simultaneously. Photo-acoustic angiography with RGD-GNS has been demonstrated as a significant platform for tumor identification, photo-thermal therapy, and monitoring therapeutics. RGD-peptides conjugated with Au-NSs has been utilized for tumor bearing mice. Tumor angiogenesis manifest magnified contrast images. When laser light falls on tumor sites, tumor growth has been observed along with PA-imaging again due to effective inhibition of tumors growth and photo-thermal ablation. Furthermore, it is concluded that diagnosis efficiency of photo-acoustic technique offered deep imaging with homogeneous resolution in contrast to other optical tomographic approaches for premature identification of cancerous angiogenesis along with immediate nano-therapeutic assessment (Weber et al. 2016). Jo et al. have also prepared dual aptamer Au-NSs by seed-mediated method and PEG with mercapto-polyethylene glycol mono-methyl ether to target prostate cancer cells (Jo et al. 2014). Song et al. have found that nano-flowers (Au-NFs) have been received attention due to unique properties such as sharp tips that enhanced the local magnetic-fields (Song et al. 2013). Han and his co-workers also developed synthesis techniques to prepare hollow Au-NFs (Chen et al. 2018). These NFs were negligibly toxic and highly effective photo-thermal effect. Meanwhile, surface plasma resonance of Au-NFs has been successfully regulated to NIR-light by varying the particle-size of nano probe. This study also revealed that larger sized gold nano-flowers (Au-NFs) exhibit low cytotoxic when irradiated with visible light but when it was exposed by laser light, it cytotoxicity frequently improved due to photo-thermal ablation. Similar studies have been investigated that the gold coated Fe_3O_4 nano-roses (Au-NRs) for photo-thermal therapy with unique properties utilized for targeted delivery, chemo-therapy, MRI and optical-imaging (Madkour 2018).

Photo-thermal properties of NIR-responsive DDSs

Plasmonic-based drug delivery systems represent entirely unique mechanism known as photo-thermal effect (PTE). In photo-thermal therapy, NIR light converts into heat radiations. Due to photo-thermal effects of plasmonic nanostructures (Baffou and Quidant 2013), these DDS systems are garnering great attention now a days. When NIR-responsive materials irradiated, they continuously convert light into heat and targeted at tumor sites and release a drug in a controlled way (Ahmad et al. 2016a, b) as demonstrated in Fig. 5.

Thermal energy is used to destroy cancerous tissues due to their strong responsivity towards heat as compared to normal tissues (Luk et al. 1980). Many thermo-responsive materials are utilized to prepare these systems. These NIR-responsive DDSs absorb near-infrared light and produce heat, this heat increases temperature of the system and as a result drug release enhanced either by varying phase or interruption of drug carrier's structure. Higher Temperature (hyper-thermia) increases cytotoxic effects and chemo-photo-thermal therapy as well (Hasan et al. 2009). Those drug-carriers which have powerful absorption in NIR region

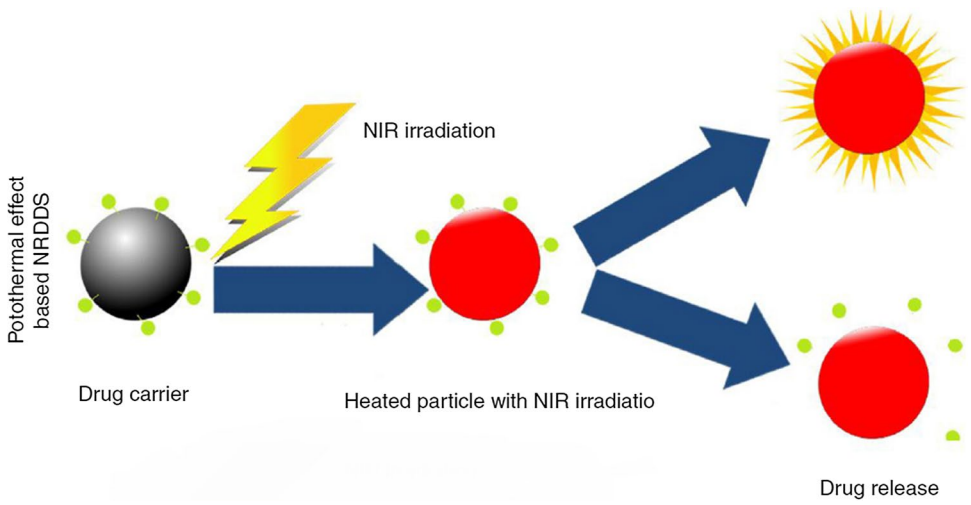


Fig. 5 Photo-thermal mechanism of NIR-responsive DDSs (Raza et al. 2019)

Table 1 Different types of Au-NPs along with EM images and DDSs type

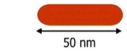
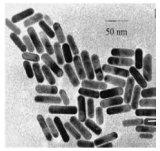
Morphology of Au nanostructures	Approach used	EM images	DDS modality type	Key findings	References
Nano-rods	Chemo-photo-thermal therapy	 	NIR-responsive	In-vivo analysis of S180 exhibiting Swiss-albino mice and In-vitro evaluation of Si-Ha, ME-180, Ha-Cat, and 3T3	(Prasad et al. 2018)
			CD44-NIR-responsive	In-vivo analysis of tumor infected mice and In-vitro study of MCF-7 Cells	(Xu et al. 2017a, b)
			pH NIR-responsive	In-vitro analysis of MCF-7 and HeLa Cells, In-vivo study of S-180 tumor infected mice	(Xu et al. 2017a, b)
	Photo-thermal therapy		Targeted, NIR-responsive	In-vitro studies on cancer cells elimination	(Alkilany et al. 2012; Black et al. 2013; Huang et al. 2006)
			Non-targeted, NIR-responsive	In-Vivo studies on cancer theraputics	(Dembereldorj et al. 2014; Sugiura et al. 2015)
Nano-rods laden Macro-phages	Photo-thermal therapy		Targeted, NIR-responsive	In-vivo studies on cancer treatment	(Betzer et al. 2015; Li et al. 2016)
	Chemo-photo-thermal therapy		NIR-responsive	In-vitro analysis of MCF-7 and In-vivo evaluation utilizing Dalton- Lymphoma ascites exhibiting Swiss-albino mice	(Arunkumar et al. 2015)
			NIR-responsive	In-vitro analysis on MCF-7 using fluorescence imaging	(Wang et al. 2016a, b)
			Thermo/NIR-responsive	In-vitro studies for U87 cells	(Tang et al. 2012)
	Photo-thermal therapy		NIR-responsive	In-vitro analysis of T6-17 cells	(Charati et al. 2010; Hribar et al. 2011)
			Targeted, Irradiated with laser light	In-vitro elimination of HepG2 cells	(Jin et al. 2012a, b)
			Targeted	In-vitro studies on drug delivery	(Amreddy et al. 2015a)
			Targeted, NIR-responsive	Enable to control drug release	(Charati et al. 2010; Hribar et al. 2011)
			Irradiation with laser light, specific	In-vitro studies on cancerous cells elimination	(Jin et al. 2012a, b)
			Targeted	In-vitro study on DDSs	(Amreddy et al. 2015b)

Table 1 (continued)

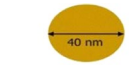
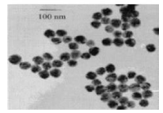
Morphology of Au nanostructures	Approach used	EM images	DDS modality type	Key findings	References
Nano-shell	Chemo-photo-thermal therapy		NIR-responsive	In-vitro evaluation of 143B and HeLa cells	(Liu et al. 2017a, b, c)
	Photo-thermal therapy		NIR-responsive	In-vitro studies to control drug release	(Bikram et al. 2007; Conde et al. 2012)
	Chemo-photo-thermal therapy		pH NIR responsive	In-vitro evaluation of HeLa cells	(Wang et al. 2017)
	Photo-thermal therapy		NIR-responsive	In-vitro study on cancer treatment	(Bikram et al. 2007; Sershen, Westcott, Halas and West 2000)
SiO_2 coated nano-shell			Non-targeted, NIR-responsive	In-vivo analysis of PEG-Coated Au nano-shells in DDSs	(Gobin et al. 2007; Hirsch et al. 2003; IANCU 2013; Lal et al. 2008; Loo et al. 2005; O'Neal et al. 2004; Stern et al. 2006, 2008)
			Targeted, NIR-responsive	In-vitro analysis of PEG-coated cell elimination	(Carpin et al. 2011; Loo et al. 2005)
			NIR-responsive	In-vitro Study on solid Tumors treatment	(Chhetri et al. 2014; Choi et al. 2007)
Nano-sphere	Photo-thermal therapy	 	Targeted, visible light responsive	In-vitro study on cell elimination	(El-Sayed et al. 2006; Pitsillides et al. 2003; Qu et al. 2012)
			Targeted, visible and NIR-responsive	In-vitro study on cell elimination	(Lapotko et al. 2006; Zharov et al. 2005)
			Targeted, NIR-responsive	In-vitro evaluation of Cancer Cells elimination	(Huang et al. 2007)
			Targeted	In-vivo and in-vitro study on drug delivery	(Coelho et al. 2016; Zhang et al. 2016)
Nano-cage	Photo-thermal therapy		Targeted, NIR-responsive	In-vitro study on cell elimination	(Chen et al. 2007)
			NIR-responsive	In-vitro evaluation of MCF7/ADR Cells	(Zhang et al. 2018a, b)
PEG coated nano-cage			Targeted, NIR-responsive	In-vivo study on cancer treatment	(Chen et al. 2010)
Nano-ring			Targeted, NIR-responsive	In-vitro study on cell elimination	(Chu et al. 2015)
Nano-star			Chemical delivery	In-vitro studies on Cancer Cells elimination	(Yuan et al. 2012)

Table 1 (continued)

Morphology of Au nanostructures	Approach used	EM images	DDS modality type	Key findings	References
Nano-flower	Photo-thermal therapy		Non-targeted, NIR-responsive	In-vivo and in-vitro study of cancer Therapeutics	(Han et al. 2014; Huang et al. 2015)
Nano-cluster			NIR-responsive	In-vivo study to control rate of drug release	(Kwon et al. 2015)

are used for effective photo-thermal therapy, administrated intravenous infections due to strong tumor homing ability, bio-degradability, and welfare (Huang et al. 2006). Table 1 presents previously reported various morphologies of Au-NPs, their EM images and DDSs in which their unique photo-thermal properties have been utilized.

Above mentioned morphologies of Au nanostructures exhibit various photo-thermal properties that have been utilized to kill cancer cells. Prasad and co-workers investigated that Au-NR-based NIR-responsive DDSs have been extensively used for in-vivo analysis of S-180 in Swiss-albino mice and in-vitro evaluation of Si-Ha, ME-180, Ha-Cat, and 3T3 using chemo-photo-thermal therapy technique (Prasad et al. 2018). Similarly, Huang et al. used photo-thermal therapy of Au-NRs for targeted NIR-assisted DDSs for in-vitro studies on cancerous cell elimination (Alkilany et al. 2012; Black et al. 2013; Huang et al. 2006). Chu et al. have found that plasmonic gold nano-rings utilized for targeted NIR-assisted DDSs for in-vitro evaluation of tumor cell elimination (Chu et al. 2015). Zhang et al. have reported that the gold nano-cage-based NIR-responsive DDSs for in-vitro analysis of MCF7/ADR cells (Zhang et al. 2018a, b). Han and co-workers have developed non-targeted NIR-assisted DDSs using gold nano-flowers for in-vivo and in-vitro of tumor therapeutics (Han et al. 2014; Huang et al. 2015). Zharov et al. investigated that gold nano-sphere-based targeted DDSs used for in-vitro analysis of cancer cells elimination when irradiated with visible and NIR-light (Lapotko et al. 2006; Zharov et al. 2005). The silica coated plasmonic Au nano-shells have also been utilized to develop non-targeted NIR-responsive DDSs for In-vivo evaluation of tumor cells (Gobin et al. 2007; Hirsch et al. 2003; Iancu and nanomedicine 2013; Lal et al. 2008; Loo et al. 2005; O'Neal et al. 2004; Stern et al. 2006, 2008). Kwon et al. found Au nano-clusters that have been significantly enabled to develop NIR-responsive DDSs for in-vivo analysis to control drug release in cancer treatments (Kwon et al. 2015). Yuan and co-workers investigated Au nano-star-based chemical drug delivery to study in-vitro evaluation of tumor cells (Yuan et al. 2012).

Conclusion

The plasmonic nanostructure-based drug delivery systems exhibited promising applications in various medical industries, in-vivo studies, clinical evaluations and specifically to target cancer cells. Stimuli-based NIR-responsive DDSs exhibit significant importance in cancer treatments as compared to conventional therapies but have many road blocks in way. As compared to internal stimulus-based DDSs, external-stimuli activated NIR-responsive DDSs such as NIR-light-responsive DDSs significantly utilized by

plasmonic Au nanostructures and manifest efficient photo-thermal properties to target deeper cancerous tissues and kill them. To achieve distinctive photo-thermal attributes, various shapes and sizes have been successfully fabricated by Au- NPs by utilizing various techniques. Furthermore, resonance wavelength of Au-nanostructures entirely depends upon the size and shape of the nanostructures. This review summarized that the plasmonic-based NIR-responsive DDSs not only deliver drugs but also used to diagnose tumor sites and monitor behavior of targeted nanostructures. In recent decades, scientific community focused on the surface modification techniques of plasmonic nanostructures to enhance the duration of these nanostructures. The growing medical demands required the fabrication of multifunctional nanostructures.

Compliance with ethical standards

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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