IJPSR (2015), Vol. 6, Issue 3 (Review Article)

INTERNATIONAL JOURNAL OF PHARMACEUTICAL SCIENCES AND RESEARCH

Received on 24 June, 2014; received in revised form, 12 October, 2014; accepted, 16 November, 2014; published 01 March, 2015

A REVIEW ON NANOTIZED HERBAL DRUGS

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Keywords:

Nano Carriers,
Herbal Remedies,
Nano Technology,
Nanopharmaceutical,
Drug Delivery Systems.

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ABSTRACT: A budding interest in Nanopharmaceuticals had generated a number of advancements throughout recent years with a focus on engineering novel applications. Nanophytomedicines are prepared from active phytoconstituents or standardized extracts. The world market for nanomedicine is estimated to reach $130.9 billion by the fiscal year 2016. Liposome nanoparticle (NP) with entrapped doxorubicin was reported by researchers to be 300 times more effective because of better pharmacokinetic ability in treatment of Kaposi sarcoma. Nanotized herbal drug containing active principles of seawort, cassia twig and liquorice root was found to be effective in pulmonary, liver, bone and skin cancer. Nanotechnology also offers the ability to detect diseases at much earlier stages, such as finding hidden or overt metastatic colonies often seen in patients diagnosed with breast, lung, colon, prostate, and ovarian cancer. However, the safety and long-term effects of nanoformulations must not be overlooked. This review will provide a brief discussion of the major nanopharmaceutical formulations as well as the impact of nanotechnology on herbal drugs in future.

INTRODUCTION: Herbal medicines have been widely used worldwide since ancient times and have been recognized by physicians and patients for their better therapeutic values as they have fewer adverse effects as compared to modern medicines. Medicinal plants are now getting more attention than ever because they have potential of providing large benefits to society or indeed to all mankind, especially in the line of medicine. The herbal treatment helps to increase the therapeutic value by reducing the toxicity and side effects of drugs at the same time it also increases the bioavailability [1].

QUICK RESPONSE CODE

DOI: 10.13040/IJPSR.0975-8232.6(3).961-70

Article can be accessed online on: www.ijpsr.com

DOI link: http://dx.doi.org/10.13040/IJPSR.0975-8232.6(3).961-70

In this approach nanotechnology plays a great role and the use of nanotechnology in herbal medicine and more specifically in drug delivery is set to spread rapidly. Nano herbal drug delivery systems have a potential future for enhancing the activity and overcoming the problems associated by medicinal plants [2]. So, the herbal nanocarriers help to treat the dangerous diseases like cancer, Diabetes etc.

For a long time, herbal medicines were not considered for development of novel formulations due to lack of scientific justification and processing difficulties but modern phytopharmaceutical research can solve the scientific needs (such as determination of pharmacokinetics, mechanism of action, site of action, accurate dose required etc.) of herbal medicines to be incorporated in novel drug delivery system, such as nanoparticles, microemulsions, matrix systems, solid dispersions, liposomes, solid lipid nanoparticles and so on. The herbal drugs can be utilized in a better form with enhanced efficacy by incorporating them into modern dosage forms. This can be achieved by designing novel drug delivery systems for herbal constituents [3].

Phytosome is a patented technology developed by a leading manufacturer of drugs and nutraceuticals, to incorporate standardized plant extracts or water soluble phytoconsituents into phospholipids to produce lipid compatible molecular complexes [4]. Asoka Life science Limited launched the world's first poly-herbal mouth dissolving tablet, fast mouth dissolving drug. A wide range of medicinal plants are used to get different rasayanas which possess different medicinal properties against various microbes. A variety of medicinal plants and plants extracts are also used for extracting immunomodulator compounds [5]. Some natural sources (plants) which stimulate immune system are *Boerhaavia diffussa*, *Rhododendron spiciferum, Caesalpinia bonducella, Capparis zeylanica, Luffa cylindrical, Withania somnifera* and *Asparagus racemosus* etc.

Therefore advent of nanotechnology is considered to be the biggest engineering innovation since the industrial revolution. Proponents of this new technology promised to re-engineer the man-made world, molecule by molecule, sparking a wave of novel revolutionary commercial products from machines to medicine. The origin of nanotechnology is often attributed to a concept advanced by Richard P. Feynman. The word nano derived from the greek noun "nano" meaning "dwarf". A nanometer is one billionth of meter or length of 10 hydrogen atom placed side by side or 1/80000 of thickness of human hair.

The particle size of nanoparticles (NP) in medicine ranges from 5 nm - 250 nm. These are produced by various chemical or physical processes and having specific properties. The world market for products using nanotechnology is estimated to reach US$1trillion by 2015. Chinese and Indian government are also investigating and committing to nanotechnology [6]. In 2006 Indian and Australian governments contributed 20 million dollars to start Australia-India Science Research Funding Programme. According to the report published by BCC Research, the market value of the worldwide nanomedicine industry was $63.8 billion and $72.8 billion in 2010 and 2011, respectively. The market is estimated to grow at a CAGR of 12.5% to reach $130.9 billion by the fiscal year 2016. The market for anti-cancer products was valued at $28 billion in the fiscal year 2011 when compared to the 2010 value of $25.2 billion. It is anticipated to reach $46.7 billion by the fiscal year 2016 for a CAGR of 10.8% during the period 2011-2016.

The nanocarriers are made of safe materials, including synthetic biodegradable polymers, lipids, and polysaccharides [7]. The activity of herbal medicines depends on overall function of a variety of active components, as all the constituents provide synergistic action and thus enhance the therapeutic value. Each active constituent plays an important role and they are all related to each other. Some Herbal Drug Nanoparticles presented in **Table 1.**

TABLE 1: HERBAL DRUG NANOPARTICLES

Formulations	Active ingredients	Biological activity	Method of preparation	References
Curcuminoids solid lipid nanoparticles	Curcuminoids	Anticancer and antioxidant	Micro-emulsion technique.	8
Glycyrrhizic acid loaded nanoparticles	Glycyrrhizin acid	Anti-inflammatory antihypertensive	Rotary-evaporated film ultrasonication method.	9
Nanoparticles of cuscuta chinensis	Flavonoids and lignans	Hepatoprotective and antioxidant effects	Nanosuspension method.	10
Artemisinin nanocapsules	Artemisinin	Anticancer	Self assembly procedure.	11
Berberine-loaded nanoparticles	Berberine	Anticancer	Ionic gelation method.	12

In Phyto-formulation research, developing nano dosage forms (Polymeric Nanoparticles Nanospheres and Nanocapsules, Liposomes, Proliposomes, Solid Lipid Nanoparticles, Nanoemulsion, etc.) has large number of advantages [13] for herbal drugs, including enhancement of solubility and bioavailability, protection from toxicity, enhancement of pharmacological activity, enhancement of stability, improving tissue macrophages distribution, sustained delivery, protection from physical and chemical degradation, etc. Thus, the nano-sized drug delivery systems of herbal drugs have a potential future for enhancing the activity and overcoming problems associated with plant medicines**.** Hence, integration of the nanocarriers as a NDDS in the traditional medicine system is essential to conflict more chronic diseases like asthma, diabetes, cancer, and others [14].

Although nanotechnology contributions are advantageous for several medicinal areas, it is essential to highlight some of the disadvantages. Clinical researchers have mentioned some negative factors, such as high cost, difficulty of scaling up processes, and the easy inhalability of nanoparticles, which can result in dangerous lung diseases, and often lead to other diseases that can lead to changes in homeostasis, or even death. It has been widely proposed to combine herbal medicine with nanotechnology because nano-structured systems could potentiate action of plant extracts, promote sustained release of active constituents, reduce the required dose, decrease side effects, and improve activity [15].

Classification of Nanoparticles:

- **Labile Nanoparticles:** Liposomes, micelles, polymers, nanoemulsions etc.
- **Insoluble Nanoparticles:** TiO_2, SiO_2, fullerens, quantum dots, carbon lattices, nanotubes etc.
- **One dimensional Nanomaterial:** Nanowire and nanotube.
- **Two dimensional Nanomaterials:** Self assembled monolayer film.

Need For Nano-Sized Delivery System for Herbal Remedies:

Nano-sized herbal delivery system was selected to overcome the drawbacks of the traditional herbal drug delivery systems because of the following reasons [16]:

- Nanoparticle can be used to target the herbal medicine to individual organ which improves the selectivity, drug delivery, effectiveness and safety and thereby reduces dose and increases patient compliance.
- Nanoparticles can be utilized to increase the herbal drug solubility and help to localize the drug in a specific site thus resulting in better efficacy [17].
- They appear to be able to deliver high concentrations of drugs to disease sites because of their unique size and high loading capacities.
- Delivering the drug in small particle size enhances the entire surface area of the drugs therefore allocating quicker dissolution in the blood.
- Shows EPR (enhanced permeation and retention) effect, i.e., enhanced permeation through the barriers because of the small size and retention due to poor lymphatic drainage such as in tumor [18].
- Exhibits passive targeting to the disease site of action without the addition of any particular ligand moiety.
- Decreases the side effects.

Approaches of Nanotechnology:

Nano carriers:

Nanocarriers entrapping herbal drug will carry optimum amount of drug to their site of action bypassing all the barriers such as acidic pH of stomach, liver metabolism and increase the prolonged circulation of the drug into the blood due to their small size [19].

A nanocarrier is nanomaterial mainly used as a transport module for another substance, such as a drug. Commonly used nanocarriers include micelles, polymers, carbon-based materials, liposomes and other substances. Nanocarriers are currently used in drug delivery and their unique characteristics demonstrate potential use in chemotherapy. Nanocarriers include polymer conjugates, polymeric nanoparticles, lipid-based carriers, dendrimers, carbon nanotubes, and gold nanoparticles. Lipid-based carriers include both liposomes and micelles. Examples of gold nanoparticles are gold nanoshells and nanocages. Different types of nanomaterial being used in nanocarriers allows for hydrophobic and hydrophilic drugs to be delivered throughout the body [20]. Since the human body contains mostly water, the ability to deliver hydrophobic drugs effectively in humans is a major therapeutic benefit of nanocarriers.

Micelles are able to contain either hydrophilic or hydrophobic drugs depending on the orientation of the phospholipids molecules. Moom et al developed multi drug delivery system by utilizing titania nanotube arrays which was used for delivery of both hydrophobic and hydrophilic drugs [21]. One potential problem with nanocarriers is unwanted toxicity from the type of nanomaterial being used. Inorganic nanomaterial can also be toxic to the human body if it accumulates in certain cell organelles. Protein based nanocarriers are one of the promising drug and gene delivery system, since they occur naturally, and generally demonstrate less cytotoxicity than synthetic molecules [22].

Nanocarriers are used in drug delivery, especially in chemotherapy. Since nanocarriers can be used to specifically target the small pores, lower pH, and higher temperatures of tumors, they have the potential to lower the toxicity of many chemotherapy drugs [23]. Also, since almost 75% of anticancer drugs are hydrophobic, and therefore demonstrate difficulty in delivery inside human cells, the use of micelles to stabilize, and effectively mask the hydrophobic nature of hydrophobic drugs provides new possibilities for hydrophobic anticancer drugs [24].

Polymeric Nanoparticles:

Nanoparticles refer to colloidal systems with particle size ranging from 10 to 1000 nm. Nanoparticles have several advantages including solubility enhancement, bioavailability enhancement, efficacy enhancement, dose reduction and improved absorption of herbal medicines compared to traditional herbal dosage forms [25]. Liu et al. developed triptolide-loaded poly (DL-lactic acid) nanoparticles to overcome the problems of poor solubility and toxicity of triptolide [26]. Sahu et al. synthesized micellar nanocarriers using a new biodegradable and self-assembling polymer, methoxy poly (ethylene glycol) - palmitate, for curcumin delivery to cancer cells. The system comprised of methoxy poly (ethylene glycol) as hydrophilic part, palmitic acid as hydrophobic part and curcumin was present in the core of polymer micelle [27]. Zhang and Kosaraju [28] studied a biopolymeric delivery system for controlled release of catechin.

The antioxidant activity of catechin was decreased dramatically when it was introduced in an alkaline environment. Bhatia et al. developed chitosan nanoparticles for the extract of Ziziphus mauritiana and checked the effect on its immunomodulatory activity [29]. Further information about the use of polymeric nanoparticles in herbal medicine delivery is presented in **Table 2**.

TABLE 2: POLYMERIC NANOPARTICLE HERBAL FORMULATIONS

Herbal medicine	Chemical classification	Pharmacological activity	Polymeric nanoparticle formulation	Particle size	Encapsulation efficiency	Benefit of formulation	References
Triptolide	Diterpenoid triepoxide obtained from traditional Chinese medicine *Tripterygium wilfondil Hook F*	Used in the treatment of auto immune diseases, especially rheumatoid arthritis, psoriasis, leukemia anti neoplastic activity.	Poly (DL-lactic acid) nanoparticles	149.7 nm	85.7%	Enhanced solubility of triptolide and reduced toxicity	26

Curcumin	Natural polyphenol isolated from the root of *Curcuma longa*	Antitumor, antioxidant, antiamylodin, antiplatelet aggregation and anti-inflammatory	Methoxy poly(ethylene glycol)-palmitate nanocarrier	41.43 nm	100%	Enhanced solubility and bio availability of curcumin	27
Catechins (Active constituents – (+)-catechin, (-)-epicatechin.	Polyphenolic plant metabolites abundant in teas derived from the tea plant *Camellia sinensis*	Chemopreventive, anticarcinogenic, antiviral, antioxidative, anti-obesity, anti-inflammatory	Chitosan nanoparticles	1.97-6.83 µm	27.9-40.12%	Increased stability of catechins	28
Plant extract of *Ziziphus mauritiana*	Plant extract of *Ziziphus mauritiana*	Immunomodulatory activity	Chitosan nano particles	-	-	Enhanced immunomodulatory activity of extract	29

Ceramic Nanoparticles:
Ceramic nanoparticles are inorganic systems with porous characteristics that have recently emerged as drug vehicles. The biocompatible ceramic nanoparticles such as silica [30], titania and alumina can be used in cancer therapy. However, one of the main concerns is that these particles are non biodegradable, as they can accumulate in the body, thus causing undesirable effects.

Metallic Nanoparticles:
Metallic particles such as iron oxide nanoparticles (15–60 nm) generally comprise super paramagnetic agents that can be coated with dextran, phospholipids or other compounds to inhibit aggregation and enhance stability. These particles are used as passive or active targeting agents. A Gold shell nanoparticle is a novel category of spherical nanoparticles consisting of a dielectric core covered by a thin metallic shell, which is typically gold. These particles possess highly favorable optical and chemical properties for biomedical imaging and therapeutic applications. Leonard et al. developed biocompatible ginseng capped gold nanoparticles which impoved its stability and antioxidant activity [31].

Topical ointments and creams containing silver nanoparticles are used to prevent infections of burns and open wounds. Satyavani et al. synthesized silver nanpoarticles [32] by using leaf extract of citrullus colocynthis which had applications in the field of medicines, cancer treatment, drug delivery, commercial appliances, biomedical sensors etc.

Solid Lipid Nanoparticles:
Solid lipid Nanoparticles (SLNs) are colloidal carrier systems, developed in the early 1990s, that combine the advantages of other colloidal systems (such as emulsions, liposomes, and polymeric nanoparticles) for drug delivery, while avoiding, or minimizing, some of their drawbacks. SLNs have higher physicochemical stability, offer better protection against degradation of labile drugs and can also be produced on a large scale. SLNs are colloidal particles containing highly purified triglycerides, composed mainly of lipids that are solid at room temperature and further stabilized by surfactants.

The matrix of the lipid particle is solid; it can protect drug molecules against chemical degradation. However, when the system is produced, crystallization occurs, resulting in lower encapsulation efficiency and drug release. Adding a liquid lipid (oil) to an oil/water emulsion containing a solid lipid, or mixture of solid lipids, promotes the formation of SLNs. Due to their small size (50–1,000 nm) and biocompatibility, SLNs may be used in the pharmaceutical field for various routes of administration, such as oral, parenteral, and percutaneous. The SLNs are prepared by different methods such as homogenization and the warm micro-emulsion high-speed stirring ultrasonication and solvent-diffusion method. Dong et al. formulated Doxorubicin and Paclitaxel-loaded lipid-Based Nanoparticles to overcome Multidrug Resistance by inhibiting P-Glycoprotein and Depleting ATP [33]. Shi et al. also prepared and characterized solid lipid nanoparticles loaded with frankincense and myrrh oil [34]. Few other solid lipid nanoparticles are listed in **Table 3.**

TABLE 3: SOLID LIPID NANOPARTICLE HERBAL FORMULATIONS

Herbal medicine	Chemical classification	Pharmacological activity	Particle size	Encapsulation efficiency	Benefit of formulation	References
Curcuminoi-ds	Natural polyphenol isolated from the root of *Curcuma longa*	Antitumor, antioxidant, antiamylodin, antiplatelet aggregation and anti-inflammatory.	447 nm	70%	Enhanced stability of curcuminoids	8
Cryptotanshinone	Cryptotanshinone is the major active ingredient from the roots of *Salvia miltiorrhiza Bunge*	Anti-inflammatory, cytotoxic, anti-bacterial, anti-parasitic, anti-angiogenic.	121.4-137.5 nm.	94.2-96.3%	Enhancement of bioavailability of cryptotanshinone	35

Liposomes:

Liposomes are nanoparticles comprising lipid bilayer membranes surrounding an aqueous interior. The amphilic molecules used for the preparation of these compounds have similarities with biological membranes and have been used for improving the efficacy and safety of different drugs. Usually, liposomes are classified into three categories on the basis of their size and lamellarity (number of bilayers): small unilamellar vesicles or oligolamellar, large unilamellar vesicles and multilamellar vesicles. The active compound can be located either in the aqueous spaces, if it is water-soluble, or in the lipid membrane, if it is lipid-soluble.

Recently, a new generation of liposomes called 'stealth liposomes' have been developed. Stealth liposomes have the ability to evade the interception by the immune systems, and therefore, have longer half-life. Rane et al. developed Paclitaxel entrapped Liposomes, the developed formulation was pH-Sensitive and remained in the circulation for longer duration[36]. Singh et al. designed Elastic Liposomes containing colchicine for its sustained drug delivery and enhanced anti gout activity [37]. Similarly Wen et al. formulated Liposomes entrapping essential oil from Atractylodes macrocephala Koidz by modified RESS technique [38].

Proliposomes:

Although liposomes possess several advantages for drug delivery, they still have some disadvantages of physicochemical instability (aggregation, sedimentation, fusion, phospholipid hydrolysis, oxidation and sterilization in large scale production). In order to overcome these problems a novel method for liposome production has been reported, namely Proliposomes. Proliposomes are dry, free flowing particles that immediately form liposomal suspension in contact with water. Solid properties of liposomes help resolve the stability problems of liposomes. Researchers conducted a study on oral bioavailability of dehydrosilymarin encapsulated into proliposome. Dehydrosilymarin proliposomes had > 90% encapsulation efficiency and improved bioavailability as compared to its pure form [39].

Carbon Nanomaterials:

Carbon nanomaterials include Fullerenes and Nanotubes. Fullerenes are novel carbon allotrope with a polygonal structure made up exclusively of 60 carbon atoms. These nanoparticles are characterized by having numerous points of attachment whose surfaces also can be functionalized for tissue binding. Nanotubes have been one of the most extensively used types of nanoparticles because of their high electrical conductivity and excellent strength. Carbon nanotubes can be structurally visualized as a single sheet of graphite rolled to form a seamless cylinder. There are two classes of carbon nanotubes: single-walled (SWCNT) and multi-walled (MWCNT). MWCNT are larger and consist of many single-walled tubes stacked one inside the other. Functionalized carbon nanotubes are emerging as novel components in nanoformulations for the delivery of therapeutic molecules [40].

Quantum Dots:
Quantum dots are nanoparticles made of semiconductor materials with fluorescent properties. It is crucial for biological applications, quantum dots must be covered with other materials allowing dispersion and preventing leaking of the toxic heavy metals. Quantum dots glow very brightly when illuminated by ultraviolet light. They can be coated with a material that makes the dots attach specifically to the molecule they want to track. Quantum dots bind themselves to proteins unique to cancer cells, literally bringing tumors to light [41].

Microemulsion and Nanoemulsion:
Microemulsion is a system of oil, water and amphiphile that is optically isotropic and thermodynamically stable liquid solution. Among the various drug delivery systems, microemulsion is considered an ideal alternative for oral delivery of poor water-soluble compounds. They have several advantages including ease of preparation, low viscosity, thermodynamic stability, enhanced dissolution of lipophilic drugs and bioavailability improvement. Furthermore they can be administered through different routes such as transdermal, parenteral, pulmonary and ocular. Yin et al. developed Docetaxel microemulsion to improve its oral bioavailability [42]. Similarly, Wang et al. made an attempt to enhance anti-inflammatory activity of curcumin by formulating it into nanoemulsion [43]. Megha et al. formulated nano-encapsulated poly-herbal ointment for anti inflammation [44]. Zhaoa et al. developed Self-nanoemulsifying drug delivery system (SNEDDS) for oral delivery of Zedoary essential oil [45].

Toxicity of Nanoparticles:
The growing use of nanotechnology in high-tech industries is likely to become another way for humans to be exposed to intentionally generate engineered nanoparticles. Nanotechnology is also being applied in medical sciences trying to achieve a personalized medicine. However, the same properties (small size, chemical composition, structure, large surface area and shape), which make nanoparticles so attractive in medicine, may contribute to the toxicological profile of nanoparticles in biological systems. In fact, the smaller particles are, the more the surface area they have per unit mass; and this property makes nanoparticles very reactive in the cellular environment.

Therefore, any intrinsic toxicity of the particle surface will be enhanced. The respiratory system, blood, central nervous system (CNS), gastrointestinal (GI) tract and skin have been shown to be targeted by nanoparticles. A typical urban atmosphere contains approximately 107 particles/cm^3 of air that is less than 300nm in diameter. Carbon in elemental form is a major component of these particles and the size of these particles is a determinant of their ability to cause systemic cardiovascular effects. Indeed, fine and ultrafine particulate matter (from 0.1 to 2.5 mm in mass median aerodynamic diameter) that can more easily access the vasculature via inhalation are linked to cardiovascular dysfunctions, particularly in subjects with preexisting vascular diseases.

Future Prospects of Nano Herbal Medicines:
Nanotized drug delivery systems for herbal drugs can potentially enhance the biological activity and Overcome the problems associated with herbal drugs. However, significant challenges remain for implementation of clinically viable therapies in this field. Trials of novel methods to control the interactions of nanomaterials with biological systems represent some of the current challenges to translating these technologies to therapies.

New challenges in the development of nanotechnology-based drug delivery systems include: the feasibility of scale-up processes that bring innovative therapeutic techniques to the market quickly, and the possibility of obtaining multifunctional systems to fulfill several biological and therapeutic requirements [46]. Some additional new challenges include probing the targeting efficiency of nanoparticles, and satisfying international standards for their toxicology and biocompatibility.

The development of engineered nanoparticles with substantial biomedical significance has posed new opportunities and challenges for pharmacology and therapeutics [47]. Nanomaterials and nanoparticles are likely to be cornerstones of innovative nanomedical devices to be used for drug discovery and delivery, discovery of biomarkers and molecular diagnostics. As nanoparticles may also exert toxicological effects, nanotoxicology has emerged as a new branch of toxicology for studying undesirable effects of nanoparticles. Therefore, development of novel nanoparticles for pharmacology, therapeutics and diagnostics must proceed in tandem with assessment of any toxicological and environmental side effects of these particles. As the bioenvironment is already polluted with nanoparticles of particulate matter, caution should be taken to prevent any environmental hazard by intentionally generated nanomaterials.

The diversity of engineered nanoparticles and of several possible side effects represents one of the major challenges for nanopharmacology and therapeutics. Modern medical instruments (such as pacemakers, computerized artificial limbs, implanted joints, endoscopic lasers, and cardiovascular grafts) alter the human body (on a macroscale) that was hard for people to imagine a hundred years ago.

In the future, Nano Pharmaceuticals may alter the human body (on a nanoscale) in ways that we cannot imagine now but it is essential to consider benefits and side effects of the use of Nano Pharmaceuticals.

CONCLUSION: The globalization of trade and market has brought about an integration of different kinds of herbal medicines worldwide. At present, herbal medications or related products in the global market are derived from Chinese herbs, Indian herbs, Arabic herbs, and Western herbs.The collaborative research among the traditional "Herbal remedies" and newer approaches of modern drug delivery system, i.e., "Nanotechnology" has established the attractive therapies to the pharmaceutical that will enhance health of people. It is anticipated that the effectual and valuable relevance of the natural products and herbal remedies being applied with the nanocarrier will enhance the significance of existing drug delivery systems.

CONFLICT OF INTEREST: The authors declare that they have no conflict of interest.

ACKNOWLEDGEMENT: The authors are grateful to authorities of Dayanand Dinanath College Institute of Pharmacy, Kanpur (UP), India for supporting our study by providing necessary facilities such as library, internet surfing and technical support.

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How to cite this article:
Sachan AK and Gupta A: A Review on Nanotized Herbal Drugs. Int J Pharm Sci Res 2015; 6(3): 961-70.doi: 10.13040/IJPSR.0975-8232.6 (3).961-70.