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Review Article

## Nanoclays And Clay-Based Nanocomposites – A Review of Emerging Applications in Drug Delivery

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### ABSTRACT

Nanomaterials have become centre point of attraction in research because of their potential application in the field of drug delivery system. Nanoclays are the natural nanomaterials composed of phyllosilicates with nanopores. Nanoclays can be categorized into four main groups as the kaolinite group (zeolite or halloysite), the montmorillonite or smectite group, the illite group, and the chlorite group. Nanoclays can also be synthesized under proper hydrothermal conditions. Nanoclay with intercalation of quaternary ammonium cations is known as organoclays and is used as rheological modifiers. Nanoclay has layered structure which allows it to swell or shrink liquids accumulate or are removed from the layers. Layered structure, chemical stability, mechanical and thermal strength, gas-barrier properties, low cost and eco-friendly character has broadened the area of application of nanoclays in the field of drug delivery system. Chemical modification through intercalation of ions, surface modification and formation of clay-based nanocomposites enhance their utility to greater extent. This review will focus on the various applications of nanoclays and clay-based nanocomposites in drug delivery system.

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### 1. INTRODUCTION

Nanoscience is the science that causes changes in structure of material to get smaller particle size in the range of nano meters with improved physical and chemical characteristics [1]. Therefore, these nanoparticles are more useful than the original substance and have wide range of applications in chemical science, biological science, material science, electrical science, mechanical science as well as in engineering. Nano tubes, nanodots, nanofillers and nano composites have become an

important part of industrial development because of their chemical & mechanical strength, electronic, magnetic, kinetic & optical properties as well as have low cost and eco-friendly behaviour [2]. This is the reason that nano science has made its impactful presence in many aspects of our daily life [3]. In the world of nanoparticles, nanoclay has a special place because of their large availability, low cost, high strength, environment friendly nature and extraordinary stability [4-6]. As

per the location of finding, clay minerals can be classified into two groups - residual clay and transported or sedimentary clay. Surface weathering of rock is responsible for generation of residual clay whereas chemical decomposition of rock and their separate deposition far away from original stock by erosion is responsible for production of transported or sedimentary clay [7]. According to the chemical composition, clay can be categorised into four groups as smectite, chlorite, kaolinite (halloysite) and illite [8]. Nanoclays are nanoparticles of layered mineral silicates with layered structural units [9] and every individual layer unit is made up of octahedral and/or tetrahedral sheets [10]. The structure of clay minerals depends on the way of stacking of tetrahedral and octahedral layers through Van der Waals attraction [7]. The arrangement of alternating sheets of "SiO<sub>2</sub>" and "AlO<sub>6</sub>" units is the cause of different structure of clay. These units are present in 2:1 ratio in smectite and 1:1 ratio in kaolinite [11] and it is shown in Figure 1.

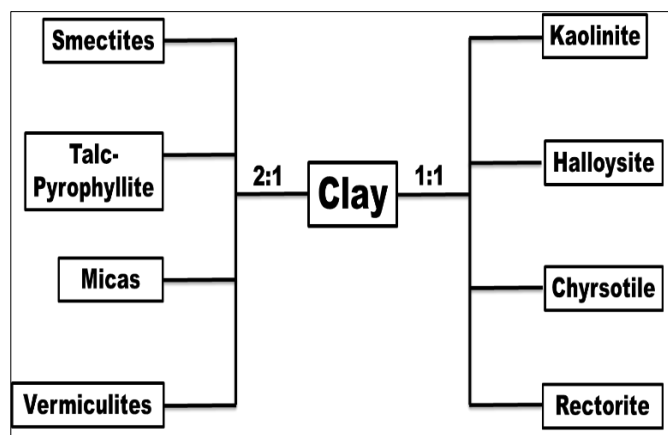


Figure 1: Different types of clay

Clay is the natural source of nanomaterial. Clay is the material made up of particles of size less than 2µm of diameter. Therefore, clay already includes nanoclay particles of size less than 100 nm of diameter. Nanoclay can be extracted from clay material by centrifugation and cross-flow filtration, energetic stirring, centrifugation and freeze-drying ultracentrifugation and ultrasonication [11- 17]. Nanoclays can also be synthesized under proper hydrothermal conditions. Nanoclay with intercalation of quaternary ammonium cations is known as organoclays and is used as rheological modifiers [2]. The nanoclay has crystalline structure with different layers consist of tetrahedral and octahedral sheets. Different arrangements of these sheets are responsible for different structure of nanoclay [8]. Nanoclay is the refined form of clay minerals with numerous enrichments of properties such as high aspect ratio, potentially exfoliation characteristics and better mechanical performance [18]. Chemical modification through intercalation of ions, surface modification and formation of clay-based nanocomposites enhance their utility to greater extent. Therefore, nanoclay has become much popular as reinforcing fillers for composites among the several nanoparticles [18]. This review will focus on

the applications of nanoclays and clay-based nanocomposites in drug delivery system.

## 2. Types of Nanoclay

Nanoclays are group of clay minerals with particle size ranges between 1 – 100 nm. The manipulation of clay minerals at this scale imparts novel characteristics and functionalities, making nanoclays valuable in fields such as materials science, nanotechnology, and polymer composites. The different types of nanoclays are as follows:

### 2.1 Montmorillonite Nanoclays

This is a species of the smectite group of hydrous phyllosilicates and has a 2:1 layered structure [19]. The layers consist of aluminum or magnesium octahedral sheets sandwiched between two tetrahedral sheets of silica. Montmorillonite nanoclays are known for their high surface area and cation exchange capacity. Although a montmorillonite particle is several hundred nm long (and wide), its thickness is in the nanoscale range (<10 nm). Montmorillonite is naturally hydrophilic because of the presence of hydrated inorganic counter ions (Na<sup>+</sup> or Ca<sup>2+</sup>) in the interlayer space. Since applications involve intimate mixing of mineral with an organic or polymer phase, the montmorillonite surface must first be rendered hydrophobic. This may be achieved by replacing the counter ions with organic cations, commonly long chain quaternary ammonium or phosphonium cations [20]. Montmorillonites intercalated with quaternary ammonium cations (QACs) are known as 'organoclays' [21].

### 2.2 Kaolinite Nanoclays

Kaolinite belongs to the kaolin group of clay minerals. It has a 1:1 layered structure, consisting of a layer of aluminum octahedra bonded to a layer of silicon tetrahedral [22]. Kaolinite nanoclays are widely used in the production of ceramics, coatings, and paper due to their unique rheological and optical properties. In addition, they find application in the pharmaceutical and cosmetics industries for their use in drug delivery systems and as additives in skincare products [23].

### 2.3 Halloysite Nanoclays

Halloysite is a tubular form of kaolinite and is characterized by its hollow cylindrical structure [24]. These nanotubes have a lumen and an external surface, providing additional functionality. Halloysite nanoclays are employed in various applications, including drug delivery systems, catalysis, and as reinforcements in polymer composites. The tubular structure of halloysite allows it to encapsulate and release active substances, making it a promising candidate for controlled drug delivery [25].

### 2.4 Bentonite Nanoclays

Bentonite is swelling clay that belongs to the smectite group. It is known for its ability to absorb water and increase in volume. Bentonite nanoclays find applications in diverse industries, including the pharmaceutical, environmental, and construction

sectors. In the pharmaceutical industry, bentonite nanoclays are used as excipients in drug formulations, while in the environmental sector, they are employed in wastewater treatment due to their adsorption capabilities [24].

### 2.5 Illite Nanoclays

Illite is a non-swelling clay belonging to the mica group of minerals. It has a 2:1 layered structure similar to montmorillonite but with a different arrangement of layers [22]. Illite nanoclays are used in the ceramics industry for their contribution to the formation of ceramic bodies. Additionally, they find applications in drilling fluids for oil and gas exploration due to their rheological properties.

### 2.6 Rectorite Nanoclays

Rectorite is a mixed-layer clay mineral containing layers of both illite and smectite [26]. This unique combination gives rectorite nanoclays distinct properties, making them suitable for applications in the cosmetic and pharmaceutical industries. Rectorite nanoclays are used in skincare products, such as masks and lotions, due to their ability to improve the texture and stability of formulations.

### 2.7 Palygorskite and Sepiolite Nanoclays

Palygorskite and sepiolite are fibrous nanoclays that belong to the group of clay minerals known as the magnesium silicates. These nanoclays have a needle-like morphology and are characterized by high aspect ratios [24]. Palygorskite and sepiolite nanoclays find applications in the production of nanocomposites, rheological modifiers, and as additives in the paint and coatings industry.

### 2.8 Layered Double Hydroxide (LDH) Nanoclays

LDH nanoclays are unique in that they are not traditional clay minerals but are layered materials with a positively charged layer and exchangeable anions. They exhibit excellent anion exchange capacity and are used in applications such as flame retardants, drug delivery, and as catalysts in various chemical reactions [27, 28].

## 3. Properties of nanoclay

Nanoclays, also known as nano-sized clay minerals or layered silicate nanoparticles, represent a fascinating realm within the vast field of nanotechnology. These materials have gained significant attention due to their unique properties and versatile applications across various industries. Some of the properties of nanoclay are as follows:

### 3.1 Nanostructure and Morphology

At the heart of nanoclays lies their nanostructure, characterized by extremely small particle sizes, often in the range of a few nanometers. The morphology of nanoclays is predominantly layered, forming stacks of plate-like structures. These layers can be separated easily, providing a large surface area and giving rise to the remarkable properties that set nanoclays apart from their macroscopic counterparts [8].

### 3.2 Surface Area and Reactivity

One of the most notable features of nanoclays is their exceptionally high surface area. The increased surface area is a result of the nanoscale dimensions and the layered structure, allowing for enhanced reactivity [29]. This property is exploited in various applications, such as catalysis and adsorption processes, where the large surface area facilitates efficient interactions with other substances.

### 3.3 Mechanical Reinforcement

Nanoclays exhibit remarkable mechanical properties, making them desirable for reinforcement applications in composite materials. When incorporated into polymers, nanoclays enhance the mechanical strength, stiffness, and thermal stability of the resulting composite. This reinforcement effect is attributed to the ability of nanoclays to form a well-dispersed and interconnected network within the polymer matrix [29].

### 3.4 Barrier Properties

Nanoclays act as effective barriers to the permeation of gases and liquids. The interlayer spaces between the clay layers create tortuous paths that hinder the movement of molecules. This property is exploited in packaging materials to improve the shelf life of food products by preventing the ingress of oxygen and other gases. Additionally, nanoclays find applications in coatings to create barriers against corrosion and environmental degradation [29-31].

### 3.5 Thermal Stability

The thermal stability of nanoclays is another noteworthy aspect. These materials can withstand high temperatures without undergoing significant degradation [32]. This property is particularly advantageous in applications where heat resistance is crucial, such as flame-retardant coatings and high-temperature polymer composites.

### 3.6 Optical Properties

Nanoclays exhibit intriguing optical properties that can be tuned based on their composition and structure. The interaction of light with nanoclays results in interesting phenomena, such as changes in transparency and color. This property is harnessed in the development of optical devices, sensors, and even in cosmetic formulations where light-scattering effects are utilized for a desired appearance [33].

### 3.7 Rheological Properties

The rheological behavior of nanoclay suspensions is distinctive, and it plays a crucial role in various applications. The addition of nanoclays can significantly alter the viscosity and flow properties of a material. This is exploited in industries such as cosmetics, where the rheological properties of nanoclays contribute to the texture and stability of products like creams and lotions [34].

### 3.8 Environmental Friendliness

Nanoclays are often considered environmentally friendly materials [2]. Their natural origin and abundance contribute to their sustainability. Additionally, their use in improving the barrier properties of materials can lead to reduced resource consumption and waste by extending the lifespan of products.

### 4. Clay nanoparticles in Drug Delivery

Clay minerals are used as active ingredients in healthcare sector because of their morphology, biocompatibility, biostability, surface physiochemical, mechanical and thermal properties [25]. Studies showed that the simultaneous use of medicine and clay minerals causes reduction in adsorption capability of medicines which can be used for controlled drug release in pharmaceutical industry. Nanoclays such as montmorillonite, halloysite, and

kaolinite are some of the examples which are used as nanocontainers and nanocarriers in drug delivery system [25]. These nanoclays work through entrapment, intercalation, encapsulation, microencapsulation, adsorption and ion exchange mechanism for modified and sustained release of drugs [35-40]. These drug-nanoclay hybrids are available in the form of tablets as well as in the form of polymeric matrices for use. Benzotriazole, Dexamethasone, Nifedipine, Paclitaxel, Tetracycline, Doxorubicin, Tramadol, hydrochloride, Carboplatin are few examples of drug-nanoclay hybrids used for controlled drug release in healthcare [23, 35-40]. The active participation of nanoclay in drug delivery system is shown in Table 1.

**Table 1:** Role of clay nanoparticles in drug delivery system

| Drug category                              | Type of Drug    | Type of Nanoclay used | Application of Nanoclay   | Benefits   | Reference                 |
|--|-----------------|-----------------------|---|--|---------------------------|
| Antibiotics                                | Oxytetracycline | Montmorillonite       | Nanoclay as carriers for antibiotic drugs                                       | <ul style="list-style-type: none"><li>Enhanced stability of antibiotics</li><li>Controlled release for prolonged therapeutic effect</li><li>Prevention of microbial resistance</li></ul>         | [23, 25, 40, 41, 42, 43,] |
|  | Metronidazole   | Montmorillonite       |   |  |                           |
|  | Amoxicillin     | Halloysite            |   |  |                           |
|  | Cefradine       | Montmorillonite       |   |  |                           |
| Anti-cancer                                | Cardanol        | Halloysite            | Nanoclay-based drug delivery systems for targeted drug delivery to cancer cells | <ul style="list-style-type: none"><li>Enhanced drug solubility and bioavailability</li><li>Controlled release for sustained therapeutic effect</li><li>Reduced systemic toxicity</li></ul>       |                           |
|  | Irinotecan      | Montmorillonite       |   |  |                           |
|  | Doxorubicin     | Kaolinite             |   |  |                           |
|  | Fluorouracil    | Montmorillonite       |   |  |                           |
| Cardiovascular Drugs and Anti-hypertensive | Atenolol        | Montmorillonite       | Nanoclay in formulations for cardiovascular and anti-hypertensive drugs         | <ul style="list-style-type: none"><li>Improved drug stability and shelf life</li><li>Controlled release for sustained therapeutic effect</li><li>Targeted delivery to specific tissues</li></ul> |                           |
|  | Carvedilol      | Montmorillonite       |   |  |                           |
|  | Hydralazine     | Montmorillonite       |   |  |                           |
|  | Venlafaxine     | Montmorillonite       |   |  |                           |
| Anti-inflametry                            | Glutathione     | Montmorillonite       | Nanoclay in formulations for sustained release of anti-inflammatory drugs       | <ul style="list-style-type: none"><li>Prolonged drug action</li><li>Improved patient compliance</li><li>Targeted delivery to inflamed tissues</li></ul>  |                           |
|  | Resveratrol     | Montmorillonite       |   |  |                           |
|  | Amphetamine     | Halloysite            |   |  |                           |
|  | Sildenafil      | Montmorillonite       |   |  |                           |

Role of different types of nanoclay in drug delivery system is discussed below:

#### 4.1 Halloysite in drug delivery

Halloysites are a promising option for drug administration because of their biocompatibility, large surface area, capacity to interact with drug molecules through ion exchange reactions or surface adsorption, solubility in aqueous solution, and curved surface. There are several distinct morphologies of halloysite, including spheroidal, platy, short tubular and elongated tubes, which are the most prevalent. Halloysite nanotubes are significantly larger than various synthetic porous materials, such as carbon nanotubes, and exhibit a 1D porous tubular

structure on the mesoporous (2–50 nm) and even macroporous (>50 nm) scale. Their distinct characteristics enable them to have a wide range of possible uses, including supporting the loading of various functional moieties at the nanoscale. Two different kinds of hydroxyl groups can be found on the outside and interior of halloysite nanotubes out of which inner one can be used for loading of drugs. The natural halloysite nanotubes as well as the halloysite nanotubes modified by chemicals, both can be used for the drug delivery. Thus, halloysite nanotubes can be used as nanocarriers for loading and subsequent release of various drugs like amoxicillin and sodium salicylate, aspirin, ibuprofen, ciprofloxacin, and curcumin [25].



Lvov et al, 2016 found that halloysite clay nanotubes can be loaded with pharmaceuticals at a weight percentage of 10–30 weight percent. This allows for a steady, controlled release of the medication for a few hours (when halloysite is utilised in its purest form) or for days, weeks, or even months if the tube ends are polymer-capped or plugged. Since the nanotubes disperse in water, PBS buffer, or cream preparations, halloysite-drug formulations can be employed.

In oral formulations, halloysite may prolong the period of drug release and is an effective excipient for tablet compression. Moreover, polymeric matrices for dental resins, bone implants, and tissue engineering (such as electrospun microfibers) may be doped with halloysite at a weight percentage of 5 to 10% medication-loaded halloysite can be doped into polymeric composites to boost their strength and surface adhesion while also delivering a long-lasting and sustained release of the medication in contrast to direct the drug admixed to the same material.

#### 4.2 Kaolinite in drug delivery

Due to the special physical, chemical and surface physicochemical properties biocompatibility, biostability and non-immunogenic properties, Kaolinite the key ingredient of ceramic industry, plays a vital role in drug delivery systems. They are used for detoxification of gastrointestinal tract as they are very good adsorbent for heavy metals or toxic antibiotic compounds. It also has antitumor activity i.e. it can reduce the mass of tumour. Therefore, it can be used for the treatment of cancer patients [44]. Due to a high level of biocompatibility and very low cytotoxicity, Kaolinite and its derivatives can be used as biofriendly biomaterials for treatment of prostate cancer, hepatocellular cancer, cervical cancer, oesophageal cancer, pancreatic cancer, and thyroid cancer [23].

The active hydroxyl group present between the layers of kaolinite structure is the active site available for modification to increase the drug delivery efficiency of it. Certain modification of kaolinite structure can increase the stability and dissolution profile of a drug and can also enhance the therapeutic effect on malignant tumors [41, 42].

Due to the non-symmetrical distribution of atoms in kaolinite domain, polar molecules are present in the interlayer of structure. Because of this polarity and formation of hydrogen bond between the kaolinite layers, solid bonds formed between the sheets of kaolinite which make it difficult to insert molecules in its interlayer. This issue is resolved in kaolinite based nanohybrids which can be obtained by surface, intercalation or exfoliation modification of kaolinite [45].

In surface modification kaolinite surface has been coated with layer of organic compounds which further improves the physical and chemical properties of the kaolinite and hence improves its drug loading and releasing capacity [45]. In intercalation modification polar organic molecules are directly introduced between the layers of kaolinite which dislocate the interlayer hydrogen bonds of it and form new hydrogen bonds to decrease the structural disorder and to weaken the bonding force between the layers. As a result kaolinite dissociates into

smaller particles, due to which surface area and dispersibility increases. In exfoliation modification of kaolinite, inlayer hydrogen bond are broken due to an external force such as mechanical grinding [46], liquid-phase exfoliation [47], and chemical preintercalation [48]. These decrease the strength of the layers and break them into nanosize and make them suitable for drug delivery applications.

#### 4.3 Montmorillonite in drug delivery

Excellent adsorption capacity, large surface area due to platy structure, good cation exchange capacity and low cytotoxicity has made montmorillonite able to exchange bioactive molecules with other ions present in biological system. The medication can be delivered into the body and displaced from its substrate in biological fluids by counter ions. Therefore, montmorillonite is the potential option for drug delivery system. Research found it useful for drug delivery to the eye [49], transdermal delivery [50], colon targeted drug delivery [51] and gastro retentive system [52]. Moreover, the polyamide montmorillonite hybrid obtained by intercalation of ions by ion exchange process is also useful for sustained drug release in oral administration and also show effective antimicrobial activity [43, 53]. One of the examples of this is anti-diabetic drug metformin hydrochloride which was obtained by intercalation in silicate layers of smontmorillonite by ion-exchange mechanism.

Montmorillonite is also a key ingredient of biodegradable nanoparticles by including Poly(lactide)- vitamin E TPGS copolymer for oral chemotherapy by using docetaxel as a model anticancer drug. This combination exhibits high drug high emulsification effect, encapsulation efficiency, high cellular adhesion and adsorption [25].

Iliescu et al., 2014 prepared and characterised irinotecan nanocomposite beads based on montmorillonite and sodium alginate as drug carriers. This hybrid is obtained by ionotropic gelation technique. X-ray diffraction, IR spectroscopy, thermal analysis and scanning electron microscopy studies showed that the hybrid is able to control the release of irinotecan by making it sustained, without any robust effect, and by reducing the released quantity and by decreasing the rate by which it released. Therefore, this nanocomposite beads may be a promising drug delivery system in chemotherapy.

Salahuddin et al., 2014 prepared polyamide-montmorillonite nanocomposites by intercalation of polyamide loaded 1,3,4-oxa(thia)diazoles into montmorillonite through ion exchange process and the structure of the composite was analysed by proton nuclear magnetic resonance, Fourier transform infrared-spectroscopy, X-ray diffraction, thermogravimetric analysis, and transmission electron microscope. They found that these nanocomposites had good antimicrobial activity and ability of sustained release of 1,3,4-oxa(thia)diazoles in oral administration.

#### Future scope and challenges in use of nanoclay

While nanoclays offer a myriad of advantages, challenges persist in their large-scale production, dispersion in various matrices, and understanding the long-term effects of their use.

Researchers continue to explore novel synthesis methods and applications, with on-going efforts to optimize their properties for specific industrial needs.

The properties of nanoclays make them a fascinating and promising class of nanomaterials with diverse applications across multiple industries. Their unique combination of mechanical reinforcement, barrier properties, thermal stability, and more opens up new possibilities for innovation in fields ranging from materials science to medicine. Nanoclays are useful for drug delivery and sustainable release of drugs and delivery of active ingredients. As research in nanotechnology advances, the full potential of nanoclays is yet to be realized, promising a future where these miniature wonders play a significant role in shaping the materials of tomorrow.

## CONCLUSION

Nanoclay stands at the forefront of nanotechnology, offering a multitude of advantages in drug delivery system. Its unique combination of enhanced mechanical properties, barrier capabilities, flame retardancy, and biocompatibility positions it as a versatile material with the potential to revolutionize the way of designing and manufacturing products. The nano clay properties like large surface area, good capacity of ion exchange, good porosity and simple to functionalize, make it able for efficient encapsulation, controlled release of drugs, and delivery of therapeutic agents to a particular target. Apart from these advantages of nano clays, low solubility, low stability, large scale synthesis, long term toxicity and transformation from laboratory product to clinical use create hurdle in their pathway of drug delivery system.

Researchers and industry professionals must work collaboratively to develop innovative solutions, refine production processes, and establish robust standards to ensure the responsible and sustainable integration of nanoclay into modern drug delivery system. As research in nanotechnology continues to advance, the full scope of nanoclay's benefits is likely to expand, contributing to a more sustainable, efficient, and technologically advanced future in drug delivery process.

## CONFLICT OF INTEREST: Nil

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