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RESEARCH ARTICLE

NANOWASTE: TINY WASTE THAT MATTERS A LOT

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ABSTRACT

Engineered nanoparticles (ENMs) are extensively used in varied consumer products. Along the period of usage for nano-enabled products, these nano particles can be released into the environment and get accumulated in the waste streams. Accidental or deliberate release of nanoparticles in the environment results into a new type of waste which is termed as nanowaste. In the framework of this review paper, details about nanowaste, challenges encountered during nanowaste management, Analysis of nanowaste along with methods for its efficient and methodical disposal are summarized. Suitable analytical methods for characterization of nanowaste form the backbone of the nanowaste management system. Various methods for nanowaste sampling, separation and quantification are used according to the nature of the nanomaterial. These methods are not sufficient as per the current requirement due to their inability to distinguish between ENMs and Naturally occurring Nanomaterials. Development and Standardization of analytical protocol is the need of the hour. Appropriate handling of nanowaste during its analysis and treatment becomes obligatory due to the hazards by nanoparticles on human life and the environment. Method of nanowaste disposal should be carefully chosen while keeping in mind the nature of the material, Post treatment requirements and disposal conditions. Currently, four methods are available, namely Incineration, Recycling, Landfilling and Organic treatment. Landfilling is usually known as the method of ultimate disposal, while other methods are usually followed by Landfilling or recycling at the end. With full regard of the current developments in this field, further work must be conducted for apposite characterization of the nanoparticles. Moreover, improvement of the current disposal methods, to minimize hazards and disposal costs is essential along with development of new methods.

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INTRODUCTION

Invention on nanomaterials dates back to late 1980s, but it wasn't until early 2000s when the use of nanoparticles was started in varied applications. Nanostructured materials are designed to use in fields like controlled drug delivery, semiconductors, optronics, catalysis, photovoltaic cells, to name a few. As their use flourished, waste in the form of nanoparticles (NP) was also generated. Harmful effects and the problems caused by these particles, when exposed to environment was unknown earlier. But as the technology has advanced, disposal of nanomaterials (NM) after their usage has become an arduous task. These new unique forms of waste streams, that contain residue nanomaterials, may pose challenges to the current waste management practices and technologies (Musee, 2010). These new forms of waste streams are generically referred herein as nanowastes. Research in the field of Nanowaste Management gained momentum early in the decade of 2010 with the realization of the fact that nanoparticles possessed some serious threats to the living beings. Nanowaste as shown in Figure 1, is generically defined as the waste streams containing NMs or synthetic by-products on nanoscale dimensions or the waste streams resulting from the end of lifespan of formerly nanotechnologically-enabled materials and products, and even the items contaminated by NMs such as pipes, personal protection equipment, etc are considered to be nanowaste (Musee, 2011).

Nanowaste can exist in the below given forms:

- Pure NMs at the point of production.
- Materials and surfaces contaminated with NMs (containers, disposable personal protection equipment, etc.)
- Liquid suspensions containing NMs
- Solid matrixes containing NMs (e.g. in the bulk, surface, or as a coating on the surface).

Classification of Nanowaste: For the purpose of scrupulous analysis and suitable treatment technique, nanowaste has been classified based on the immanent characteristics of the constituent chemicals, the expected exposure dose and the waste stream quantity (Musee, 2010).

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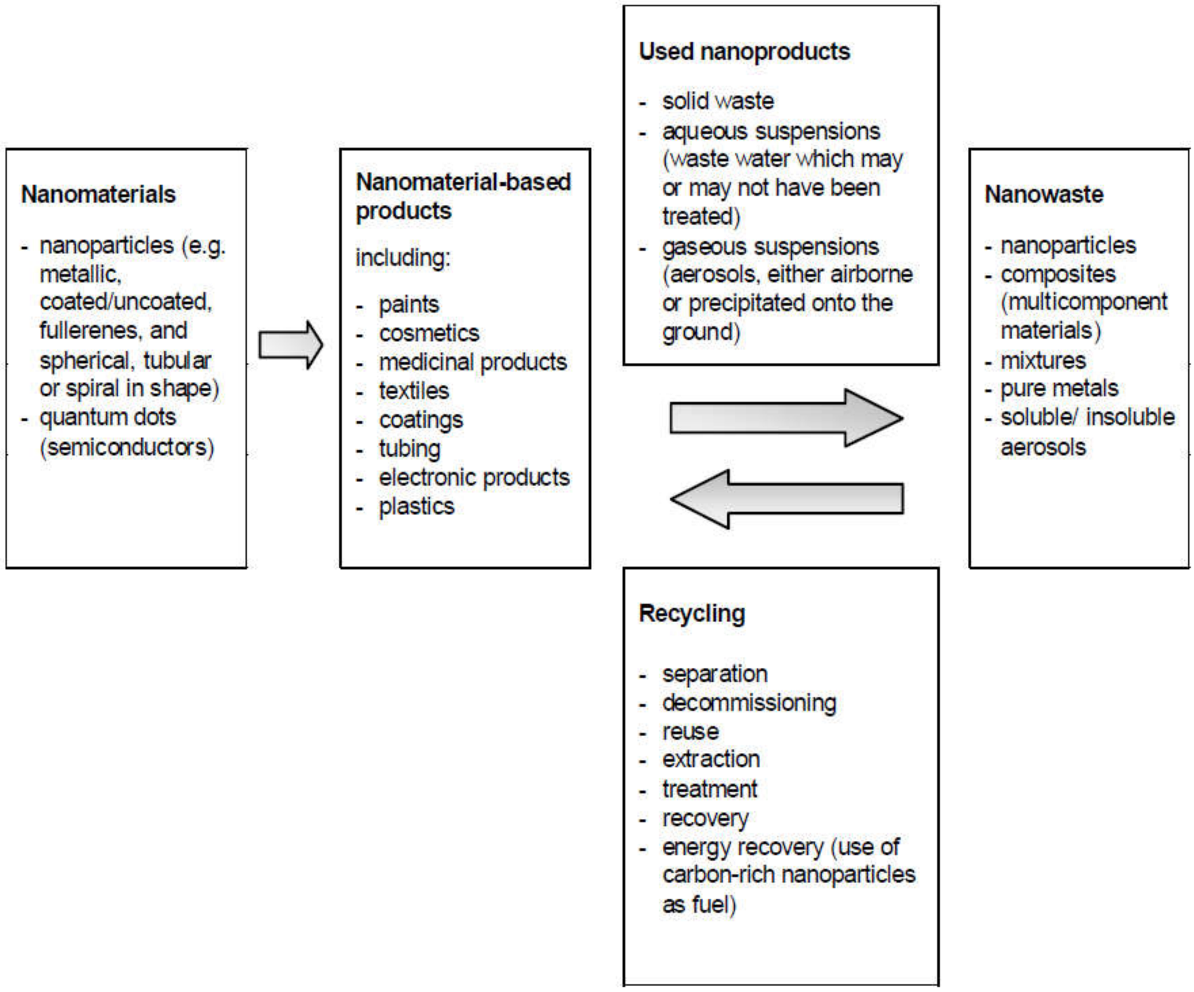

Fig. 1. Lifecycle of Nanomaterials (Musee, 2010)

- **Class I Nanowastes:** The nanowastes which fall under this category have very low or no toxic effects in humans and other ecological systems mainly due to non-toxic constituent NMs. Exposure potency for class I nanowaste has no influence on the hazardousness of the waste stream even if the NMs are bound on the surface or inside the bulk of the material. Examples of such nanowastes are likely to include those generated from display backplane in television screens, solar panels, or memory chips containing silicon nanowires. Although, if the NMs break away or leach out, low to high exposure levels may be observed.

- **Class II Nanowastes:** Class 2 nanowastes are likely to exert harmful or toxic effects on humans and the environment due to the inherent toxicity of its constituent NMs, which ranges from low to high. Based on the results obtained from the matrix developed to derive the nanowastes classes, the overall hazard potential was found to be strongly linked to the exposure potency due to nanostructures bound on the surface or inside the bulk part of the material. If the exposure potency is low or unlikely, such wastes may be handled as non-toxic though they contain highly toxic materials. Examples include nanowastes generated after the lifespan expiry of display backplane and memory chips. These mainly contain single walled carbon nanotube (SWCNT) which are found to be highly toxic for the organisms.

- **Class III Nanowastes:** A nanowaste stream is classified as Class III type if its toxicity can be categorized as toxic to very toxic accompanied by low to medium potential exposure during the disposal phase. Therefore, the resultant waste stream is likely to have medium risk potential to the ecological systems and should be handled as a hazardous waste. Nanowaste arising from food packaging, food additives, wastewater containing personal care products, polishing agents and pesticides can be classified as class III nanowaste.

- **Class IV Nanowastes:** Toxicity hazard of NMs in this category ranges from toxic to very toxic, and the exposure potential ranked as medium to high because the NMs in the nanoproduct are anticipated to be freely bound on the nanoproducts (in liquid- or solid-bound form). These waste streams are regarded as highly hazardous for organisms as well as the environment. Therefore, such waste streams require specialized handling and should be treated adequately either by immobilizing or neutralizing the NMs. Paints and coatings, personal care products, pesticides etc fall under the purview of class IV nanowaste.

- **Class V Nanowastes:** Class V nanowastes are categorized as extremely hazardous dur to their high degree of exposure and their toxicity, which ranges from very toxic to extremely toxic. Such waste streams require specialized handling, effective treatment, and must be disposed of in well-designed and designated disposal sites. Continuous monitoring of the sites is recommended to ensure that the leachates from the disposal site are adequately managed. Among the most suitable technologies for treating such wastes includes immobilization and neutralization processes. The waste stream is not only extremely toxic but also likely to have very high exposure potential when released into the environment because it is in liquid form, which potentially promotes easy interactions with environmental organisms. Pesticides, sunscreen lotions, food and beverages containing fullerenes in colloidal suspensions are very harmful and hence classified as class V nanowastes.

Challenges faced during Nanowaste Management

Behaviour of nanoparticles is quite different from their bulk counterparts as they tend to be more chemically active and toxic than the ordinary, regular sized particles. This problem is further enhanced due to the dynamic transformation of the particles during their entire lifetime. Such transformation influences the fate and behaviour of these materials in different environments owing to nanostructures intrinsic properties (e.g. surface chemistry, aggregation, agglomeration, adsorption or absorption properties, etc.), and the environmental factors (pH, presence or absence of oxidants, complexed ions, zeta potential, effects of macromolecules, presence of other chemicals, etc). Due to this, predicting the behaviour of particles under different conditions is a herculean task and this unpredictability poses serious troubles in material handling.
Varied companies ranging from small scale start-ups to large global entities which are involved in nanomaterial production, produce wide variety of nanoparticles which have different sets of properties. This renders nanowastes management quite challenging task – and unless universal principles and technologies of managing these wastes are developed urgently – a case-by-case approach recommended presently may prove uneconomically viable, laborious, and even impractical considering the number and types of NMs as well as the nanoproducts. Moreover, the rate at which the development in nanoproducts takes place is exorbitant when compared to the rate at which the NMs detection systems have advanced, especially in the soil systems and free water bodies. This makes it improbable to detect, monitor, and develop remediation protocols to mitigate possible nanopollution in the soil environments. Nanowaste is notoriously difficult to contain and monitor; due to its small size, it can spread in water systems or become airborne, causing harm to human health and the environment. Trace analysis and quantification of nanoparticulate species is also very challenging because of the variety of ENM types that are used in products and low concentrations of nanowaste expected in complex environmental media. Adding to the trouble, there is paucity of toxicity data and its relationship to the physicochemical properties of NMs, incoherency of few reported toxicity data and lack of universally agreed units of expressing the NMs toxicity (Musee, 2011).

Analysis of Nanowaste

At the moment, risk assessment of nanowastes is yet to be quantified mainly because of the lack of generic principles that governs risk assessment and characterization of NMs. This means, unless the challenges regarding the response of NMs to organisms and the actual degree of exposure in environment are addressed – at this point – it is difficult to characterize the risks of nanowastes. This is exacerbated by lack of ecotoxicological data to the ecosystems as well as exposure potency of these materials (Baer *et al.,* 2010). For example, very few data exist on NMs regarding their hazard effects, probability of occurrence in diverse environmental systems, and the exposure potency (bioaccumulation and bio persistence).

Therefore, it is scarce or highly unlikely to find data for a specific NM on:

- Its ecotoxicity to organisms both under laboratory and environmental conditions
- Its expected degree of exposure to aquatic and terrestrial organisms in diverse environments (e.g. water and soil)
- Its exposure potency (e.g. bioaccumulation and bio persistence)
- The precise dose response relationship between the levels of exposure to the aquatic and terrestrial organisms, and the consequent adverse effects observed.

Material parameters that are to be analysed for proper characterization of the nanowaste are as follows (Part *et al.,* 2015):

- Particle number concentration and mass concentration are important regarding toxicity and for differentiating between concentrations of species stemming from dissolved ions, nanoparticles or colloids (e.g., metal cations, metallic nanoparticles or larger metal-containing aggregates, with apparent sizes of <1 nm, 1–100 nm, and >100 nm, respectively).
- The elemental composition of nanomaterials leads to different toxicological effects. The fate and behaviour of ENMs also depend on the chemical elements that are used for particle's core, shell or coating (e.g. coated nanocomposites).
- Particle size and particle size distribution are critical parameters for characterization of ENMs as they influence uptake and toxicity mechanisms. For example, some ENMs have the potential to cross biological barriers, such as the blood–brain barrier.
- The shape of nanomaterials can also influence toxicity, transport and behavior of nanomaterials in the environment, which are also influenced by the aggregation state. Similarly, ENM structure and crystallinity can influence the stability and toxicity of nanomaterials (e.g. rutile or anatase TiO_2-ENMs).
- Surface properties such as surface area, charge, functionality and speciation influence bioavailability, toxicity and aggregation kinetics of nanomaterials (e.g. citrate-functionalized Ag or CdTe/ZnS core–shell nanoparticles).

Treatment of Nanowaste

As nanotechnology-based products enter into widespread use, many will end up in disposal waste streams. Disposal is the phase in the product life cycle at which most nanomaterials are predicted to enter the environment. However, this topic has gained recent attention with several life cycle analyses and reviews specifically addressing nanomaterial release through different disposal pathways. There are four methods for treatment of nanowaste to convert them into harmless, simpler molecules. The methods are incineration, landfilling, recycling and organic treatment (Allan *et al.,* 2009).

Incineration

Incineration is a key waste treatment technique with great potential for modifying nanomaterials and either controlling them effectively or releasing them to the environment. Nanomaterials may enter waste streams that will be incinerated through several different pathways: disposal of consumer products as municipal solid waste (MSW), wastes generated from nanotechnology research and development, hazardous wastes, medical and infectious wastes, and sewage sludge from wastewater treatment plants (WWTP) handling nanomaterial laden water. Incineration can be used to remove highly toxic organic wastes, reduce the volume of wastes, and potentially recover some of the energy stored in wastes. Incineration facilities burn hazardous wastes at high temperatures (850–1200 ºC) in oxidative environments to ensure complete combustion of the waste before release to the atmosphere. There are various types of incinerators, including water-wall, modular, multiple hearth, catalytic combustion, waste-gas flare, direct-flame, liquid injection, fluidized bed, rotary kiln, and grate incinerators (moving and fixed). The choice of incinerator type depends on the type, volume, and hazard of the waste to be destroyed. Moving grate incinerators are the most common type in the US and account for 90% of the MSW incinerators in Europe. Moving grate incinerators can handle large volumes of waste with heterogeneous composition and calorific value. Rotary kiln incinerators are commonly used because they can combust various types of wastes, including solid, liquids, and sludge, with minimal processing. Fluidized bed incinerators are also common due to their high combustion efficiency and low emissions compared to other incinerator types. Since the majority of incinerators used for MSW are grate types, the details of the processes in them that may alter, destroy, and form new nanomaterials are presented in greater detail (Al-Kattan *et al.,* 2013).

Behaviour of nanoparticles during incineration

When the nanowaste is exposed to such high temperatures during incineration operation, five opportunities exist for the (re-)formation or destruction of ENMs:

- ENMs are destroyed due to combustion (for example CNT to CO2)
- ENMs are not destroyed or incinerated but captured by the flue gas treatment system (for example metal oxides). These ENMs can be detected afterwards in the fly ash or other residues.
- Certain types of ENMs may not be destroyed during combustion. However, they react with other substances and form new particles (e.g. $CaCO_3$ to CaO and CO_2 or ZnO + HCL give $ZnCl_2$ + H_2O).
- Bigger particles decompose and turn into new, smaller particles or even ENMs. Roes et al. 2012 describes how ENMs can be destroyed and, converted into other ENMs or left unchanged during incineration.
- Agglomeration of ENMs to bigger particles may occur, therefore, those particles lose their "nano" status.

Table 1 summarizes the combustion behaviour for several common nanomaterials. The two nanomaterials with highest production levels (TiO2 and SiO2) are not combustible, likely persist through the combustion zone, and may impact pollutantformation. This suggests that nanomaterials produced in the largest quantities may impact the incinerator system and require greater scrutiny to limit releases to the environment. [H] After the combustion of the nanowaste, flue gas disposal shall be done based on the constituents of the same. Hence, air sampling of the flue gases followed by the outflow streams filtration system is mandatory to intercept the release of harmful nanoparticles in the gaseous stream.

Recycling of Nanowaste

Recycling of waste is one of the strategies of waste minimization or waste prevention and of Sustainable Materials Management (SMM). Individual countries and international bodies have established recycling goals and are taking measures to accomplish them.

Challenges in recycling the nanowaste

The Organization for Economic Co-operation and Development (OECD) states that the principal challenges with safe and environmentally benign recycling procedures for waste containing nanomaterials are:

- Controlling the health, safety and environmental risks arising from recycling processes of waste containing nanomaterials.
- Controlling the technical and environmental quality of secondary materials that may be contaminated with nanomaterial from the original waste stream.

Develop technologies that may be used for the recovery of the nanomaterial from the products, given suitable quantities, concentrations and economic value of the nanomaterials.[I] As observed in Table 2, very less amounts of Nano-ZnO and Nano-TiO_2 are recycled, while more than half of the total fullerene produced is effectively recycled (DTU Environment, 2015).

Risks related to nanomaterials during recycle

During the recycling of Waste Containing Nanomaterials, engineered nanoparticles, like any other particle in the recycling process, might remain individually isolated or form new bigger agglomerates. Below given are the possible exposure pathways that must be taken into account when managing waste containing nanomaterials, especially nano-objects that are free or that are releasable (e.g. due to the recycling process):

- Nanoparticles may penetrate biological barriers.
- They may show intensified effects in the case of substances with toxic properties.
- They may have increased bioavailability.
- They may have different chemical and physical properties from the "parent" material.
- Some types of CNT (carbon nanotubes) and nanowires may have similar effects in the lungs to asbestos fibers.
- Attention must be paid to the risk of dust explosions (as in all applications of inflammable powders or powdery substances).

Table 1. Combustion behaviour of common nanomaterials

Sr. No.	Nanomaterial	Production Level	Combustible	Fire Retardant	Persist through combustion zone	Increases Pollutant Emissions
1	SiO2	High	No	Yes	Yes	Unknown
2	TiO2	High	No	Yes	Yes	Yes
3	CNT	Medium	Yes	Yes	Yes	Unknown
4	CeO2	Low	No	Yes	Yes	Yes
5	Ag	Low	Unknown	Unknown	Unknown	Yes
6	Fullerene	Low	Yes	Unknown	Unknown	Yes

Table 2. Percentages of different nanomaterials recycled in European Union

Nanomaterial	Percentage Recycled, %
Nano-TiO_2	18
Nano-ZnO	11
Nano-Ag	36
CNT	20
Fullerenes	51

Repurposing of Nanowaste

A new variant on the concept of nanoparticle recycling which has been recently garnering interest is the idea of repurposing traditional waste materials into nanomaterials. For example, plastic bags have been used to create carbon dots, which are small carbon nanoparticles (less than 10 nm in size) with interesting optical properties and potential applications as imaging agents. These carbon dots were made by simply cutting the bags into small pieces and heating the in a solution of hydrogen peroxide, a household bleach you can find in your first aid cabinet. Another waste repurposing method is to heat discarded compact discs (CDs) in a furnace along with sand to create silicon carbide nanoparticles nanomaterial with outstanding thermal, chemical, and mechanical properties. The use of CDs is noteworthy because, with increasing urbanization in the world, electronic waste like CDs is estimated. to be accumulating at three times the rate of normal household trash. Nanoparticle-contaminated water will undergo several purification processes, including mechanical filtering and settling treatments (designed to remove large particles [larger than a grain of sand]), followed by digestion with microbes, and ultimately chemical disinfection. Unfortunately, none of these treatment stages are specifically designed to eliminate nanoparticles from the waste stream. As a result, following waste water treatment, nanoparticles may remain in the purified water that is released back into the environment, or nanoparticles may remain trapped in the microbe-bearing sludge left over from the purification process. Often, the bio-sludge left over from wastewater purification is repurposed as fertilizer for farm land.

Landfilling of Nanowaste

Waste disposal on land (dumping) and landfilling remains the most prominent waste management techniques used worldwide. The standards and practices for this type of waste disposal vary greatly ranging from uncontrolled sites to highly specialized and controlled engineered landfills. The potential release of contaminants through landfill gas and leachate is largely dependent on landfill design, site conditions and the sophistication of the control measures in place, including landfill gas recovery and leachate collection and treatment systems. Modern engineered landfills use synthetic barriers, with few relying on natural barriers, to line the bottom of a landfill and incorporate collection systems for both leachate and landfill gas.

The purpose of these collection systems is to capture and treat leachate and landfill gas; thereby preventing the migration of leachate into ground/surface water and the release of untreated landfill gases to the atmosphere. Landfill leachate is generated when rain passes through the waste mass and by the liquid generated due to the breakdown of waste within the landfill. The composition of leachate is highly vacillating depending on the type of waste, the amount of precipitation, the construction and operation of the landfill, the age of the landfill and other factors such as pH, temperature and microbial populations. The disposal of nanowastes from industrial sources into regulated hazardous waste landfills and potentially municipal landfills should not be overlooked. In one process of the manufacturing of fullerenes (carbon based ENMs), only about 10% of this material is usable and the rest are disposed of in landfills. It is observed that the amounts of waste generated from the manufacturing processes are significantly larger than the amount of the final ENM product. However, this may not be indicative of other ENM manufacturing processes and is likely a worst-case scenario considering the economic implications of discarding a high proportion of the product (Bolyard *et al.,* 2013). These considerations along with the following factors, require careful implementation of landfill disposal of ENMs or products containing ENMs:

- The manufacturing of ENMs tends to generate variegated "nano by-products" and other nanowaste streams which possess distinct toxicological characteristics requiring specialized disposal.
- A given nanomaterial may pose a range of risk profiles when subjected to disposal.
- ENMs also bond with pollutants further enhancing their toxicity and may instigate faster translocation of these pollutants through air, soil and water
- The increase in concentrations of ENMs in the environment may cause long-term chronic effects through different food chains

Degradation of nanomaterials in waste

The harsh environmental conditions within landfills, such as low pH and strongly reducing conditions (due to the anaerobic environment), will likely aid the release of ENMs bound in polymers materials bound in plastics/plastic resins/polymers/ metal products, such as those found in construction waste, may also be released into the leachate as a result of mechanical stress and abrasion during compaction and/or contact with leachate of an aggressive nature. Carbon nanotubes are one of the most widely used nanomaterial. If CNT composites are landfilled, they could slowly breakdown depending on their degradability and potentially release ENMs to the leachate (or via dust from weathered composites). However, degradation of the polymer matrix, under conditions in engineered landfills and release of CNTs is likely to be extremely slow. In contrast, the situation in an un-controlled landfill may lead to greater post-consumer and environmental releases of discarded CNT (Brent *et al.,* 2006) composites.

Landfill liner penetration by nanomaterials

Landfill liners are synthetic membranes used in an engineered landfill to separate landfill contents from the environment. Compacted clay has also been used alone to provide a physical barrier and is still currently used today in conjunction with synthetic liners, to provide a secondary barrier. The potential of ENMs to penetrate or migrate through landfill liners is currently being studied; at this time, there is a lack of conclusive findings. Recent studies in this discipline suggest that properly designed and constructed landfills will be able to significantly limit nanoparticle transport to the environment for extended periods of time (approximately 100 years). Barrier properties of geomembranes were evaluated with suspensions of nanoparticles used in paints and the diffusion test showed that the nanoparticles did not cross the membrane, which corresponds to an effective efficiency of geomembranes over 12 years in real conditions. However, ENMs placed near the bottom of MSW landfills are of concern, as they may transport or diffuse through liners, especially if they are near the bottom of the landfill. Since leachate, which is a mobile aqueous phase, could be released to the surrounding environment, which might possess risk to human and environmental health. Synthetic membrane liners will likely contain ENMs and is currently being researched. More extensive work needs to be carried out to determine the potential risk of ENMs seeping through clay liners in older landfills or in situations where uncontrolled landfills depend on natural attenuation for treatment.

Nanowaste landfill leachate treatment

Leachate treatment can incorporate one or a series of different systems such as aeration, sedimentation, settling lagoons, filtration, Ultra Violet (UV) treatment, and biological and/or chemical treatments. The purposes of these treatments are to settle out solids, adjust pH, increase oxygenation and break down or treat contaminants. The effectiveness of leachate treatment systems to adequately manage risks from ENMs will be influenced by the unique properties of ENMs and their behaviour in landfill environments (OECD, 2016).

Factors to be considered while employing leachate treatment include:

- ENMs interaction with leachate and its potential to increase (or decrease) mobility and/or toxicity.
- The integrity and nature of liners and their ability to contain ENMs.
- The impact of ENMs on the effectiveness of the treatment technology itself.

Organic Treatment

Organic treatment is basically used to separate those nanomaterials which are difficult to remove from a complex mixture. Separation of nanomaterials is difficult due to similar chemical nature, extremely small size, lack of appropriate nanomaterial detection systems and unknown quantity and type on nanomaterials present in waste streams. Adsorption of nanomaterials to activated sludge is a major removal mechanism for particles which are gruesome to separate, including manufactured nanoparticles, in conventional activated sludge wastewater treatment plants. The main objectives for conducting organic treatment are (Lens *et al.,* 2013):

- To observe the adsorption of fluorescent NPs to wastewater biomass.
- To quantify and compare biosorption of different types of NPs exposed to wastewater biomass.
- To quantify the effects of natural organic matter (NOM), extracellular polymeric substances (EPS), surfactants, and salt on NP biosorption.
- To explore how different surface functionalities for fullerenes affect biosorption.

Summary

Nanomaterials have found varied application in the fields of packaging, clothing, disinfectants, Food industry, electronic devices, catalysts etc and their use is increasing at a burgeoning rate. Due to their uncontrolled production and usage, it is a herculean task to manage them in the environment. Waste generated during their production, usage or disposal is termed as Nanowaste, which is one of the major environmental concerns in waste management. Efficient waste management systems require a felicitous waste detection and segregation system. Streams containing nanoproducts and the type of nanoparticle needs to be detected which shall decide the plan of action for its treatment. Due to their small size and distinctive properties, detection becomes an onerous process. Further developments in the nanowaste sampling, separation and characterization technology are needed to select the disposal pathway or design a tailor-made method. Selection of the nanowaste disposal method highly depends on the type of nanomaterial and its behavior during the treatment. Incineration cannot be used for a wide variety of materials due to their unpredictable behavior at higher temperatures. Generation of Air pollution is a major concern while disposing waste by incineration. Recycling of waste is the safest method for managing nanowaste but proves to be quite expensive in most of the cases. Not all nanoparticles can be recycled and controlling the properties of those that can be recycled is quite demanding. In case of nanoparticles that are difficult to recycle, repurposing to a more useful material should be preferred. Landfilling of waste (Including Nanowaste) remains to be the most prominent type of waste disposal technique due to easy and economic landfilling operations. Leachate transportation, liner penetration by leachate, contamination of water table, effect on microbial processes and degradation of waste are the main concerns in landfilling of nanowaste. Organic treatment shall be used for separation of nanoparticles which are difficult to segregate from the mixture of nanoparticles. Usually, organic treatment is followed by Incineration, Landfilling or Recycling for the ultimate disposal. For efficient management of Nanowaste, it is necessary to curb the wastage of expensive and toxic nanoparticles. Any of the methods described above, can be used for treatment, while keeping in mind the nature of the material, cost, environmental complications and ease of use.

Further developments in the accurate analysis of waste streams and economic disposal processes can prove to be a boon for the environment.

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