Global population increase and food demand

Continuous increase in the global population results in a permanent increase in the demand for energy, water, and food. It is estimated that the global food production will increase by 70% to meet the demand of a 9-billion population by the year 2050.\(^1\)\(^2\) This continuous increase in the global population exerts a driving force on agriculture in order to meet the food demand, and the production of large amounts of food in environmentally safe and sustainable manner has been of great importance in the past few decades.\(^3\)

Current agricultural productivity needs to be enhanced, and fertilisers play an important role in providing higher yields of crops and more secure food with a qualified nutritional fact.\(^4\) Therefore, the scope of enhancement of fertilisers in many aspects such as physical strength, nutrient efficiency, and kinetics release into the soil have been gaining attention.

Projections imply the necessity of adjusting the diet structure and reduction of food waste as much as possible in order to reduce the food demand in the future, and achieve a balance between supply and demand. There are several suggestions for an increase in global food production, such as increasing irrigated areas and amount of fertilisation, as well as efficient usage of water and fertilisers.\(^5\)

It is estimated that the global crop demand will increase by 100–110 % between 2005 and 2050, and utilisation of fertilisers has been increasing directly proportional from 15 to 194 Mt in the last 50 years to meet this urgent requirement.\(^6\) Although today agriculture already has environmental impacts, the future of expansion in agricultural applications is yet unclear. This uncertainty allows some environmental concerns in terms of the effect of extensively used mineral fertilisers on marine, freshwater, and terrestrial ecosystems.

Consumption of limited resources and permanently increasing human population puts emphasis on the need to develop innovative technologies for high-yield agricultural outputs without causing significant decrease in the resources.\(^7\) It is crucial to understand the future of agriculturally derived impacts on the environment, and predict alternative pathways to achieve higher yields within the concept of more environmentally friendly and sustainable approaches.\(^8\) Thus, interdisciplinary research implementing both engineering and agronomic principles should focus on innovative solutions for increasing productivity and nutritional quality in an environmentally friendly manner.\(^9\)

Mineral fertilisers and the environment

Plant nutrition is the most important parameter in order to achieve high quality production and maintain soil fertility in agriculture. The nutrient content of soil depends on various kinds of parameters, and mineral fertilisers meet the nutrient requirements of plants in the case of soil lacking...
nutrients. Mineral fertilisers provide plants with primary (N, P, K) and secondary (S, Mg, Ca) nutrients, and according to their chemical formulation they can also provide micronutrients (Cu, Mn, Fe, Zn, B).10

Mineral fertilisers have been an important agricultural input since the green revolution, and they have been used extensively in order to achieve higher yields and production on a large scale. These products are applied through the soil by surface broadcasting, sub-surface placement or dissolving in water.11 These conventional methods might lead to releases into the atmosphere or groundwater, resulting in both losses and pollution of ecosystems. Excessive application of nitrogenous fertilisers results in nitrogen release through volatilisation in the form of NH₃ or nitrous oxides (NOₓ), and leaching into groundwater in the form of NO₃. Similarly, excessive phosphorus application results in the formation of phosphorus complexes with Ca, Mg, Al, Fe, and Zn, which are not available for plant uptake. These metal complexes are accumulated in the soil or washed by rainwater into waterways and cause pollution problems.

Although utilisation of mineral fertilisers is favourable for healthy plant growth, application of excessive doses, more than optimum or standard amounts, over extended time might result in several impacts on the environment, such as air-water pollution, water eutrophication, soil degradation, soil pollution due to heavy metal and radionuclide exposure, etc.12 Moreover, nitrous oxide release upon fertilisation increases greenhouse gas emissions and contributes to global warming. However, the environmental and ecological impacts of mineral fertilisers have been realised after a period of time. Among these impacts, nutrient loss and gaseous emissions related to agricultural applications have been the leading reason for environmental pollution and climate change.13

Current projections show that, although the crop demand by 2050 will be met by adapting to high-yielding technologies, only 0.2 billion ha of land will be cleared. Moreover, greenhouse gas emissions and global N utilisation will decrease to the level of 1 Gt/year and 225 Mt/year, respectively.14 Additionally, intensive demand for mineral fertilisers results in some shortcuts in the resources and dramatic increases in agricultural costs. This cost increase reduces the farmers’ profit margins.

Mineral fertilisers meet nutrient demand of plants for their healthy growth, and the current solution approach to satisfy the increasing food demand relies on extensive use of fertilisers. However, conventional manufacturing processes might be inadequate to meet the demand for mineral fertilisers. The inverse proportion between high demands and low supplies results in an increase in the price of fertilisers. Conventional fertilisers are not only costly, but also result in some environmental impacts under excessive utilisation circumstances. Moreover, the increasing demand for these products is leading to continuous consumption of limited resources, which could possibly cause their depletion within the next 80 years.15 This phenomena has triggered the necessity of relevant research to develop environmentally friendly fertilisers with improved nutrient use efficiency and enhanced soil fertility.

3 Nanotechnology in agriculture

Nanotechnology approaches offer tailor-made production of fertilisers by introducing novel tools such as nanofertilisers, nanopesticides, nano-enabled agrochemical carriers, and nanosensors into conventional agricultural applications (Fig. 1).16–18 Application of these engineered nanomaterials is conducted as foliar spraying, seed coating, and soil amendment.19

Conventional application of mineral fertilisers offers a relatively low nutrient use efficiency, 30–35 %, 18–20 %, and 35–40 % for nitrogen, phosphorus, and potassium, respectively.20 Application of these products results in a high release rate of the nutrients just after application. This high nutrient concentration might be much more excessive than the actual absorption rate of plants, resulting in low nutrient uptake efficiency. Moreover, heavy fertiliser concentrations in the early stages might result in further deficiencies, and negatively affect the growth of plants due to nutrient toxicity.20 Thus, the development of novel nutrition sources for plants with enhanced fertiliser use efficiency has gained increasing interest, and industry has confirmed that nanotechnology will provide competitive advantages rather than conventional applications.21,22 Nanotechnology approaches in agriculture primarily aim to increase the yield and reduce nutrient losses by slow and sustained release of nutrients.23,24 Furthermore, slow release of nutrients might also help undesired nutrient-microbe interactions on soil flora, providing surface protection and improving the plants’ resistance to diseases.25

Nanotechnology is defined as the study, synthesis, and application of materials on nano-scale within the range of 1 to 100 nm, implying the capability to build tailor-made nanostructures for desired functions by atomic and molecular level controls.26,27 Particles at this scale are called nanoparticles, having different physical and chemical properties than their origin, providing novel functionalities and high reactivity due to surface area/volume ratio. This interdisciplinary field of science has provided a broad spectrum of emerging studies in the agriculture sector in the first decade of 21st century, especially focusing on enhancing food value, reducing agricultural inputs, improving the nutrient contents, and providing longer shelf-life of foods.28,29 Nanoparticles are synthesised from a wide range of materials, i.e., metal oxides, ceramics, magnetic materials, semiconductors, synthetic or natural polymers, emulsions, etc.30,31 Intense stud-
Advantages of nanofertiliser utilisation in agriculture

Nanofertilisers are defined as nanoparticles with specified formulations that help plants grow successfully so that they can benefit from nutrients for a longer time. Nanofertilisers have higher nutrient uptake efficiency than mineral fertilisers, and they do not have to be applied in larger amounts. The dissolution rate of nanoparticles in soil solutions would be higher than that of mineral fertilisers in bulk form, since the former fertilising products have much smaller particle size and higher specific surface area than those of the latter. Owing to their small particle size, they can enter into the pores of plant cell walls, but further interactions of the nanoparticles with other cell organelles, and the mechanism of how the nutrients are transferred from the nanoparticle to the plant still lack research. However, the suggested primary uptake mechanism states that, after application onto soil, the nanofertiliser products are firstly dissolved in water and soil solutions, followed by absorption of nutrients by the roots of the plants. Application of nanofertilisers not only reduces fertiliser wastage, but also helps to overcome bioavailability and uptake of nutrients in an environmentally friendly manner. Nanofertilisers are not only applied as particles or emulsions in nanoscale dimensions, they also provide nutrients in the form of nanotubes or nanoporous materials, which are immersed into a nanoparticle, and covered by a protective polymer film from the outside.

Nanofertilisers can be obtained from both organic and inorganic based nanomaterials. Inorganic nanomaterials are generally metal oxides, i.e., ZnO, TiO₂, MgO, AgO, etc., whereas organic nanomaterials are lipids, polymers, and carbon nanotubes. However, the classification of nanofertilisers is conducted in terms of their nutrient content as macronutrient and micronutrient nanofertilisers, respectively. Macronutrient nanofertilisers chemically consist of one or more macronutrients, i.e., N, P, K, Mg, and Ca, providing these essential nutrients to plants. Unlike traditional mineral fertilisers, the nutrient uptake efficiency of nanofertilisers is higher and there are no nutrient losses into surface and groundwaters. Application of these innovative fertilisers would enable safe and sustainable agricultural applications without harming the environment. Thus, emerging research has been ongoing with a high priority in the fertiliser industry. Micronutrient nanofertilisers provide Fe, Mn, Zn, Cu, and Mo. These elements are required only in trace levels. Conventional application of these elements is to introduce them into the formulations of N, P, and K fertilisers as soluble salts in low dosages. However, uptake of these micronutrients might not be efficient. Micronutrient nanofertilisers enhance the uptake of these nutrients.

Nanofertilisers can be designed according to the following principles. Firstly, they can be designed as nanotubes in which the mineral nutrients are provided in encapsulated forms. They can also be prepared as nanoporous structured materials coated with a thin polymer layer. Lastly, they can be applied as particles or emulsions at nanoscale, providing larger surface area compared to their total sizes. Fig. 2 gives a brief illustration of the advantages of utilisation of nanofertilizers in agricultural applications.

**Fig. 2 – Advantages of nanofertiliser utilisation in agriculture**

Nanofertilisers can benefit from nutrients for a longer time, increased nutrient uptake efficiency and crop yield, reduced environmental impacts, small particle size, high surface area, and high reactivity.

3.1 Nanofertilisers

3.2 Hydroxyapatite as phosphorus nanofertiliser

Phosphorus is one of the essential nutrients for plant development, and phosphorus uptake in plants is obtained through phosphate salts derived from phosphoric acid such as monoammonium phosphate (MAP), diammonium phosphate (DAP) or triple superphosphate (TSP). Thus, phosphoric acid is the main raw material in the manufacturing of phosphate-based mineral fertilisers. Phosphoric acid is mainly produced via thermal and wet process methods. Thermal process provides high-purity phosphoric acid, and is mainly consumed in food and pharmaceutical applications. Wet process is basically the dissolution of phosphate rock at 70–80 °C with sulphuric acid (H₂SO₄). Although high-purity phosphoric acid can be produced, thermal process phosphoric acid is generally not preferred in industrial scale productions due to high energy cost and corrosion problems. 85 % of globally produced
phosphoric acid is consumed for phosphate fertiliser production, and the fertiliser industry generally utilises the wet process method for phosphoric acid production.

Continuous consumption of phosphate rock remains an increasing concern, since there are finite resources of phosphate rock, and most of these reserves have been consumed in an unsustainable manner. Moreover, most of the farmlands on a global scale lack phosphorus, thus phosphorus is the least accessible essential nutrient.\textsuperscript{47,48} Due to these challenges, for phosphorus resources in fertiliser production, sustainable and more efficient approaches are urgently needed in order to provide phosphorus to the plants.\textsuperscript{49} Moreover, due to their large particle size, phosphate mobility in the soil is very limited upon application of conventional phosphate fertilisers, which decreases the phosphate concentrations in the root zone of the plants. Nano-hydroxyapatite would offer efficient P nutrient uptake while reducing the contamination risks and overcoming the problems related with utilisation of conventional phosphate fertilisers.

Natural hydroxyapatite (Ca\textsubscript{10}(PO\textsubscript{4})\textsubscript{6}O\textsubscript{2}) crystals are present in the form of mineral phase in the structure of shells, teeth, and bones in mammalian bodies, whereas synthetic hydroxyapatite usually exists in white-powdery form. Synthetic nanoscale hydroxyapatite crystals have excellent chemical analogy with those of a biologically calcified tissue, making it a biocompatible and bioactive material.\textsuperscript{50} Thus, in the past few decades they have been extensively used in biomedical applications for hard tissue reconstruction, such as osseous defect repair, bone grafting, in dental implants as bioactive ceramic coatings.\textsuperscript{51–53} Nanoscale hydroxyapatite is generally synthesised via wet chemical precipitation of Ca(OH)\textsubscript{2} derived from eggshells, followed by calcination at relatively low temperature.\textsuperscript{54} However, hydroxyapatite with appropriate characteristics at nanoscale is required for different applications, thus reinforcements with other advanced nanomaterials are performed in order to prepare these nanocomposites. Natural or synthetic nano-fiber-based electrospun polymers, graphene oxide or organic modifiers, such as ethylenediaminetetraacetic acid, polyethylene glycol, and cetyltrimethylammonium bromide, are used in the reinforcement of nanohydroxyapatite and synthesis of nanocomposite materials.\textsuperscript{55–57} Nano-hydroxyapatite (nHA) has gained significant attention in agriculture as a potential phosphorus fertiliser, and there exists a wide range of studies regarding increasing the functionality of nHA-based nanostructured fertilisers.

Phosphorus uptake upon nHA fertiliser application occurs via the dissolution of nanoparticles in soil media and the release of nutrients, followed by absorption by the plant roots. Due to their small particle size and high specific surface area, dissolution rate and extent of nanoparticles in soil media is expected to be higher than that in conventional (bulk) fertilisers. Dissolution mechanism of nHA is described in Eq. (1).

\[
\text{Ca}_{10}(\text{PO}_4)_6\text{OH} \rightleftharpoons 5\text{Ca}^{2+} + 3\text{PO}_4^{3–} + \text{OH}^–
\]  

Dissolution rate of nHA is one of the most important parameters in terms of phosphorus availability. Soil pH, salinity index or mineral concentration in soil would affect the chemical stability and dissolution properties of nHA in soil medium. Due to its chemical structure, nHA nanoparticle is a rich source of both calcium and phosphate nutrients. It can also be used in combination with some other essential elements favourable to plant growth such as zinc, copper, and iron, by producing growth hormones and chloroplast, enhancing photosynthesis efficiency, taking part in some enzyme processes, and acting as antibacterial agent.\textsuperscript{58–60} There are vast studies on the utilisation of urea-incorporated nHA, reporting that the hybrid fertiliser can perform slow release of both nitrogen and phosphorus nutrients, providing the decomposition of bare urea in soil for an extended period.\textsuperscript{61–63}

### 3.3 Theoretical studies on phosphorus uptake efficiency by nanohydroxyapatite utilisation

Liu and Lal\textsuperscript{62,64} were the first to introduce nanoscale P research in agronomy. In this study, they prepared carboxy-methyl cellulose (CMC) stabilised nHA particles via a one-step wet chemical method, and proposed a greenhouse study in order to observe the effects of utilising synthetic nHA on soybean (\textit{Glycine max}). Results showed an increase in the growth rate by 32.6 % and seed yield by 20.4 % compared to conventional P fertiliser. In addition, aboveground and underground biomass production increased by 18.2 % and 41.2 %, respectively.

Bala et al.\textsuperscript{64} evaluated the efficacy of sol-gel synthesised hydroxyapatite nanorods on seed germination and growth of chickpea (\textit{Cicer arietinum}) plant. Results showed a 2-fold increase in both germination rate and plant growth by HA nanorod compared with the control.

Li and Huang\textsuperscript{65} studied the utilisation of nHA in pakchoi (\textit{Brassica Chinensis} L) in terms of biomass, cadmium uptake, chlorophyll content, vitamin C, MDA, and activity of antioxidant enzymes in cadmium-contaminated soil. Cadmium amount in the soil was 10 mg kg\textsuperscript{-1} and the application of 5, 10, 20, and 30 nHA kg\textsuperscript{-1} increased the plant biomass by 7.97 %, 13.21 %, 19.53 %, and 20.23 %, respectively. Additionally, cadmium content in the root parts decreased by 27.12 %, 44.2 %, 50.91 %, and 62.36 % compared with the control group, respectively.

Sharonova \textit{et al}.\textsuperscript{66} also conducted a greenhouse study with a nanostructured water-phosphorite suspension derived from natural raw phosphorite. They noted an increase in the fruit yield of 14.5 % to 24.1 %. The morphometric indices of the plants, fresh yield, and crop production quality also increased from 8.3 % to 3.5-fold, 2.4 % to 2.2-fold, and 0.3 % to 2.6-fold, respectively.

Montalvo \textit{et al}.\textsuperscript{67} evaluated the transport of nano- and bulk-sized HA in saturated soil column experiments, and studied the availability of nHA and bulk-sized HA as phosphorus fertiliser in comparison with TSP in andisols and oxisols with the model wheat plant (\textit{Triticum aestivum}). Soil column experiments showed no movement in bulk-sized HA, and 5 % and < 1 % leaching of nHA in andisol and oxisol, respectively. P uptake of wheat was the highest in TSP followed by nHA, and bulk-sized HA. Although nHA showed better performance than bulk-sized HA, probably because of its higher dissolution property in soil media, TSP showed better efficiency.
Xiong et al. synthesized nHA particles with different surface charges and observed their effect as phosphorus fertiliser on sunflower (Helianthus annuus) using phosphorus-deficient soils in a greenhouse study. Results showed a 2-fold increase in the biomass of sunflower after utilisation of negative-charged nHA, compared with the plants treated with the same amount of P in the form of TSP.

Taşkın et al. studied the wet chemical synthesis, characterisation, and effect of nHA in terms of growth and P uptake by lettuce (Lactuca sativa L.) on both low and high calcareous soil. Lettuce plants were also treated with H3PO4 as a soluble P source for comparison. They noted that dry weights of lettuce plants showed a significant increase in both P sources (nHA and H3PO4). However, nHA seemed to be more effective on the growth and resulted in higher P concentration in lettuce plants.

Xiong et al. examined the efficacy and controlled release characteristics of nHA compared with TSP in two kinds of P-deficient soils having different pH ranges, vertisol and ultisol. Ultisol is relatively more acidic (pH < 5), and TSP solubility in this pH range is rapid, resulting in a rapid increase in P availability, whereas nHA showed a controlled release. Vertisol pH was around 8, and nHA showed no increase in P availability due to low solubility at this pH range. This study confirmed the inverse proportion of soil pH and P uptake when utilising nHA as phosphorus fertiliser due to low solubility of nHA in alkali pH range.

Yoon et al. studied self-assembly synthesis of a multifunctional P fertiliser by natural or synthetic humic substances and hydroxyapatite nanoparticles. Efficacy of as-synthesised fertiliser was tested on corn (Zea Mays) and compared with superphosphate and bare nHA. Results showed enhanced improvement in plant growth, productivity, and resistance to NaCl-triggered abiotic stresses with humic substance-assembled nHA fertiliser.

Tang and Fei synthesised nHA particles through biomass added alkali-enhanced hydrothermal process from calcium hydrogen phosphate (CaHPO4) and calcium pyrophosphate (Ca3P2O7). Plant cultivation tests showed an increased phosphorus use efficiency by 45.87 % and 46.21 % upon utilisation of CaHPO4 and Ca3P2O7 based nHA fertiliser, respectively. Phosphorus use efficiency with conventional phosphorus fertiliser results were reported to be 23.44 %.

Li et al. studied the potential of nHA as P fertiliser on soybean (Glycine max) with various precipitation intensities via foliar spray and soil amendment application. Synthesised nHA nanofertilisers showed enhanced results in plant nutrient content in high precipitation intensity ranges. The results of 100 % precipitation intensity showed 32.6 % more P and 33.2 % more Ca in shoots; 40.6 % more P, and 45.4 % more Ca in roots.

Li et al. evaluated P uptake of fungicidal coated, Bacillus coated, and both fungicidal and Bacillus coated soybean seed treatments in oxisol soil with 32P labeled nHA fertiliser. They concluded that the growth of soybean was pH dependent, and P uptake was maximised with fungicidal and Bacillus coated seeds when the soil was unlimed. Continuous consumption of phosphate rock is facing a challenge in terms of phosphorus availability in plant nutrition, and alternative phosphorus resources are needed. nHA offers a sustainable and more efficient approach for phosphorus uptake. Greenhouse studies related to phosphorus uptake upon nHA utilisation provide promising results; however, these studies should be elaborated with field studies.

### 4 Conclusions

Engineered nanofertilisers are specially designed in terms of their size, morphology, composition, and surface characterisation for the uptake of desired nutrients for plants within the scope of controlled release and translocation of the nutrients in plants. Replacing conventional mineral fertilisers with nanofertilisers might provide sustainable solutions in agriculture and soil health, and utilisation of nanotechnology in agriculture for enhanced crop growth and production capacity has gained significant attention. Innovative nanotechnology approaches provide the desired nutrient composition with improved nutrient use efficiency, resulting in higher yields for upgrading global farming applications while decreasing the environmental impacts. Phosphorus is one of the essential nutrients for plant development and phosphate uptake in plants remains in 5–30 % range with the application of conventional phosphorus fertilisers. Phosphoric acid is the main raw material of conventional phosphate fertilisers and it is commonly produced via wet process in which phosphate rock is treated with sulphuric acid. However, phosphate rock reserves are globally limited, and continuous consumption of phosphate rock remains an increasing concern. Nanohydroxyapatite is a promising alternative phosphorus fertiliser, offering reduction in the consumption rate of phosphate rock and efficient phosphorus uptake while reducing the contamination risks and overcoming the problems related with the utilisation of conventional phosphate fertilisers. This study focuses on studies regarding increasing the functionality of nHA-based nanostructured fertilisers in order to maintain phosphorus uptake in plants in a more sustainable manner. However, promising results of preliminary greenhouse studies should be supported with long-term field studies, and discuss the uptake, translocation, and interactions of nHA with other components in real-time field conditions.

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### List of abbreviations

- MAP = monoammonium phosphate
- DAP = diammonium phosphate
- TSP = triple superphosphate
- nHA = nano-hydroxyapatite
- EDTA = ethylenediaminetetraacetic acid
- PEG = polyethylene glycol
- CTAB = cetyltrimethylammonium bromide
- CMC = carboxy-methyl cellulose
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Fosfor, sadržan u fosfora gnojiva, jedna je od esencijalnih tvari za zdrav rast biljaka. Fosfatne stijene strateški su izvori fosfora u poljoprivredi i njihova dostupnost u bliskoj budućnosti postati vrlo ograničena. Tijekom posljednjih decenija iz thentickih vrtova dostupnosti fosfornih sirovina se smanjuje zbog prekomjernog potrošnje fosfornih stijena. Stoviše, prekomjerna prilagodba fosfornih stijena fosfora u poljoprivredi postao je problem u svijetu, a prekomjerna primjena fosfornih gnojiva glavni je uzrok ekoloških problema u poljoprivredi. Prilikom primjene fosfornih gnojiva u poljoprivredi, bio je utvrđen da su fosfornih stijena fosfora u poljoprivredi neodoljivi, a prekomjerna primjena fosfornih stijena fosfora u poljoprivredi postao je problem u svijetu, a prekomjerna primjena fosfornih gnojiva glavni je uzrok ekoloških problema u poljoprivredi.