



## Macromolecular Nanotechnology

# Synthesis and characterization of a magnetic bio-nanocomposite based on magnetic nanoparticles modified by acrylated fatty acids derived from castor oil

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## ABSTRACT

The present work addresses the synthesis and characterization of a new bio-nanocomposite based on acrylated fatty acids (AFA) derived from castor oil, ethyl acrylate and AFA surface modified  $\gamma\text{-Fe}_2\text{O}_3$  ( $\gamma\text{-Fe}_2\text{O}_3\text{@AFA}$ ) magnetic nanoparticles (MNPs). It was shown that the *in situ* formed bio-nanocomposite containing around 2.0 wt.% of  $\gamma\text{-Fe}_2\text{O}_3$  MNPs with average particle size of ca. 9.0 nm, as determined by XRD and TEM measurements, displayed good magnetic response and superparamagnetic behavior. The magnetization measurements indicated that the incorporation of  $\gamma\text{-Fe}_2\text{O}_3\text{@AFA}$  MNPs into the poly(ethyl acrylate-co-AFA) matrix did not affect the magnetization saturation, remanence and coercivity of the MNPs, preserving the magnetic properties of the  $\gamma\text{-Fe}_2\text{O}_3$  precursor. Additionally, the use of AFA to chemically modify the surface of MNPs opens a new scenario on the chemical modification of MNPs with agents that undergo free-radical reactions, allowing for the proper dispersion of MNPs into the thermoplastic polymer matrices and minimizes the undesired leaching of the MNPs.

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## 1. Introduction

There is already consensus in society [1,2] and in the scientific community [3–5] that global warming is a reality and that its origin lies in the gas emissions that cause the greenhouse effect, as the  $\text{CO}_2$  from the burning of fossil fuels. On the other hand, the oil reserves are decreasing [6–8], despite controversies in this regard [9], it is illogical to think that the oil reserves (known and not yet known) have infinite duration. For instance, in 1978 (nearly four decades ago) the physicist Albert A. Bartlett said [10]: “It is possible to calculate an absolute upper limit to the amount of crude oil the earth could contain.” Assuming that the maximum volume of petroleum in our planet is the volume of the earth, i.e.,  $6.81 \times 10^{21}$  barrels, and the petroleum consumption rate of 1978 he estimated “this earth full of oil will last only 342 yr!”. In addition, we must consider the political and socio-economic aspects related to producing countries, since nearly 50% of world crude oil reserves are in the Middle East [11] a region with high potential for warring conflicts.

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This scenario has led governments, companies and researchers to find alternatives for the replacement of petroleum as feedstock for obtaining fuels and other materials. In this context, vegetable oils have been intense and widely investigated as an environmentally friendly alternative, as they are a renewable source, can be found both in nature and through planting, are biodegradable, present none or low toxicity to humans and are a low cost resource. In the last decades vegetable oils have been used to obtain biofuels (e.g. biodiesel and bio-oil) [12] and different types of polymers [12,13]. Recent reviews in the literature cover the use of vegetable oils as raw material in the synthesis of paints and coatings [14–16,11,17], thermoplastics [18], polymer composite materials [19] adhesives for biomedical purposes [20,21], among others [22].

Due to the huge possibility of obtaining materials with distinct characteristics, as well as to modify the final properties of conventional polymers derived from non-renewable fossil resources, vegetable oils have assumed a lead role in polymer chemistry. In addition, derivatives of vegetable oils have low toxicity, biodegradability and biocompatibility as well as low production cost, which makes their use extremely promisor [17,21].

Stemmelen et al. [23] described the synthesis of vegetable oil-derived diamines by thiol-ene coupling with cysteamine hydrochloride intended to be used in curing processes with commercial epoxidized linseed oil. Based on the crosslinking kinetic studies, it was showed that the evaluated monomeric system was more reactive than other commercial fatty amides, resulting in slightly more rigid materials.

Gobin and coauthors [24] addressed the synthesis and characterization of bio-based polyester derived from broccoli seed oil and short to long chain dicarboxylic acids and carboxylic diacids. The resulting crosslinked bio-based polymers exhibited rubber-like features and low glass transition temperatures.

Due to the increased demand for adhesives, the use of vegetable oils to produce bio-based pressure-sensitive adhesives can be regarded very attractive, opening excellent opportunities for industrial applications. Very recently, Maaßen et al. [25] addressed the synthesis and the characterization of bio-based pressure-sensitive adhesives focusing on the evaluation of the mechanical properties relevant to processing and application. According to the experimental approach, monomers derived from methyl oleate and methyl erucate polymerized via free radical polymerization conducted to the formation of polymers presenting high molecular weight and adhesive properties.

Vendamme et al. [26] reported the synthesis and characterization of renewable adhesives. As a first stage of the process, hydroxyl-ended polyesters with linear and branched architectures were produced through polycondensation of dimerized fatty acids and fatty diols, followed by curing with maleinized soybean oil. According to the authors, the synthesized polymer derived from soybean oil presented good biocompatibility and could be applied as a pressure-sensitive adhesive.

Pokeržnik et al. [27] studied the copolymerization of n-butyl acrylate and glucose-based vinyl surfmer via semi-batch emulsion process in order to produce pressure sensitive adhesives. According to the authors statements, conventional pressure adhesive polymers based on acrylate monomers may be replaced by bio-based pressure adhesive polymers containing modified glucose in the recipe.

Magnetic nanoparticles have been thoroughly investigated by several research groups due its versatility and wide possibility of surface modification. Among the polymer composite materials there are those formed by dispersion of magnetic nanoparticles (MNPs) in a polymer matrix or MNPs covered by polymers. MNPs are playing an important role within the nanoscience and nanotechnology, since some properties of MNPs are different from those of the corresponding bulk materials [28]. This kind of material find applications in bio-medicine as contrasts for magnetic resonance image (MRI) [29], drug delivery [30], hyperthermia [31,32], intravascular embolization procedures [33], environmental science [34] and catalysis [35]. Recently, a review on the synthesis and applications of MNPs has been published with focus on methods that use some principles of green chemistry [36].

Composite materials present properties of the different materials that compose them, and may also have unique properties characteristic of the composite material. For instance, covering the MNPs with a polymer can promote the stabilization of the MNPs against aggregation, enhancing its application as a contrast agent in MRI or as a drug delivery vector [37]. As pointed by Hanemann and Szabó [38], polymer matrix properties such as electrical and thermal conductivity, polymer phase behavior and thermal stability, mechanical properties like stiffness, Young's modulus, wear, fatigue, flame retardancy, density, and physical properties such as magnetic, optic, or dielectric properties can be modified depending on the particle size, particle shape, specific surface area and chemical nature. Methods of synthesis and applications of this kind of material has already been reviewed [38–41].

Araujo et al. [42] developed magnetopolymeric nanocomposites based on vinyl pivalate and  $\text{Fe}_3\text{O}_4$  superparamagnetic nanoparticles intended for biomedical applications, as for instance, intravascular embolization and hyperthermia treatment. Oleic acid-modified  $\text{Fe}_3\text{O}_4$  were properly dispersed into poly(vinyl pivalate) thermoplastic matrices through *in situ* suspension polymerization, leading to the formation of magnetic polymer with good magnetic properties.

Feuser et al. [43] encapsulated surface modified magnetite nanoparticles with poly(methyl methacrylate) through miniemulsion polymerization process. According to the experimental results, poly(methyl methacrylate)/ $\text{Fe}_3\text{O}_4$  nanoparticles presenting superparamagnetic behavior did not present cytotoxicity in murine fibroblast (L929) and U87MG cells and exhibited high blood compatibility.

Meiorin et al. [44] described the synthesis of a magnetic nanocomposite based on tung oil with styrene cationically copolymerized in the presence of  $\text{Fe}_3\text{O}_4$ . According to this work, although the magnetic nanocomposite display superparamagnetism, the presence of magnetic nanoparticles in the polymeric matrix significantly affected the properties of the final material, such as the morphology, the dynamic-mechanical and mechanical properties.

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