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Magnetic Properties of Encapsulated Magnetite in PLGA Nanospheres

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In this study, the biocompatible magnetic fluid was encapsulated in biodegradable polymer PLGA (poly D,L/lactide-co-glycolide acid) by the nanoprecipitation method. We characterized these spheres in terms of morphology, magnetite content and magnetic properties. The results showed good encapsulation with magnetite content 22wt% and magnetization 3.4 mT. The transmission electron microscopy and scanning electron microscopy images showed that magnetic particles have almost a spherical shape with approximate size 250 nm. Infrared spectroscopy and thermogravimetric analysis measurements were used to confirm incorporation of magnetic particles into the PLGA polymer.

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1. Introduction

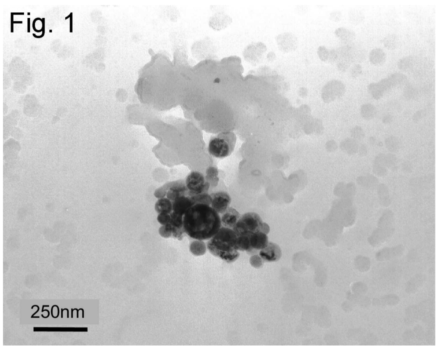
Magnetic nanoparticles, especially iron oxide nanoparticles, are inorganic materials that can be embedded in a polymeric matrix [1]. The constituents of magnetite polymer nanoparticles play different roles: the polymeric matrix acts as a shell, reservoir, and vehicle for the active component, whereas magnetite is the component which makes targeting possible by external magnetic field manipulation. Magnetite polymer nanoparticles can be used for delivery of active components, such as drugs, vaccines, proteins, enzymes and others [2]. The aim of our paper is to prepare poly D,L/lactide-co-glycolide acid (PLGA) nanospheres loaded with biocompatible magnetic fluids (MFs) by modified nanoprecipitation technique and investigate their magnetic properties.

2. Experimental methods and results

The coprecipitation method of ferric and ferrous salts in an alkali aqueous medium was used to prepare magnetite particles [3]. Sodium oleate

($C_{17}H_{33}COONa$) as a first surfactant was used for the modification of prepared magnetic particles to prevent their agglomeration. To improve stability and increase the circulation half time of the particles, the polyethylene glycol (PEG) as a second surfactant was added to the system magnetite–oleate and stirred over 3 h. This magnetic fluid-PEG had a solid loading of 110.8 mg/ml. The solid loading in this case includes the weights of the iron oxide, sodium oleate and PEG. The composition 37 wt% magnetite, 58 wt % sodium oleate and 5 wt% PEG was determined by thermogravimetric analysis (TGA). The saturation magnetization of the prepared biocompatible magnetic fluid-PEG with a concentration of Fe_3O_4 $c = 60$ mg/ml and average diameter of magnetite particles were estimated from SQUID measurements. The obtained values were 3.4 mT and 10 nm, respectively. The modified nanoprecipitation method was used to entrap magnetic fluids into polymer nanospheres [4].

The prepared samples were observed by transmission electron (TEM) and scanning electron (SEM) microscopy to obtain information about the morphology and surface characterization (shape, distribution, aggregation) of the magnetite polymer nanospheres. Figure 1 shows a TEM image of magnetic nanoparticles embedded in the PLGA polymer matrix, where dark-contrast images correspond to the magnetic particles implanted in PLGA. The SEM image (Fig. 2) clearly shows that nanoparticles have spherical shape with size ≈ 250 nm.



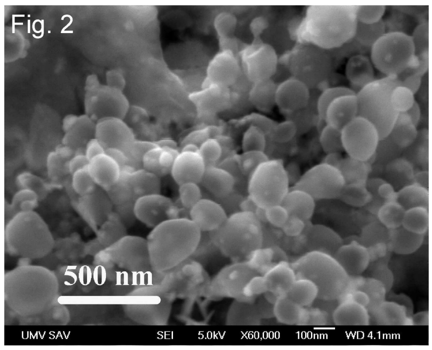
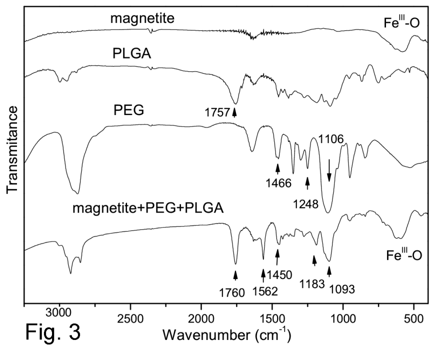


Fig. 1. TEM image of magnetite-PLGA nanospheres.
Fig. 2. SEM image of magnetite-PLGA nanospheres.

With the aim to confirm adsorption of PEG on the surface of magnetic particles as well as encapsulation in PLGA matrix, Fourier transformed infrared (FTIR) spectroscopy was used. The infrared spectra of magnetite, pure PEG, pure PLGA and magnetite-PEG/PLGA were obtained by the KBr pellet method. The IR spectra MF-PEG/PLGA (Fig. 3) shows the presence of all bands of the PEG (C–O–C and C–O–H in the region from 1093cm^{-1} to 1348 cm^{-1}), PLGA (C=O stretch band at 1760 cm^{-1}) and a weak peak at $582cm^{-1}$ that is attributed to the magnetite particles. The amount of magnetite in the PLGA matrix was also calculated from TGA data (Fig. 4) and combined with data collected from the SQUID measurement. The thermogravimetric investigations were carried out on a SETARAM thermobalance model TGDTA92 at the heating rate of 10°C/min in temperature interval from 30° to 600°C in dynamic conditions in the argon atmosphere. The TGA residue for MF-PEG at 600°C was 53.31 wt%. The TGA residue for MF-PEG/PLGA was 36.8 wt%. The 16.5 wt% difference was associated with the PLGA presence. This data correlated well with SQUID results.



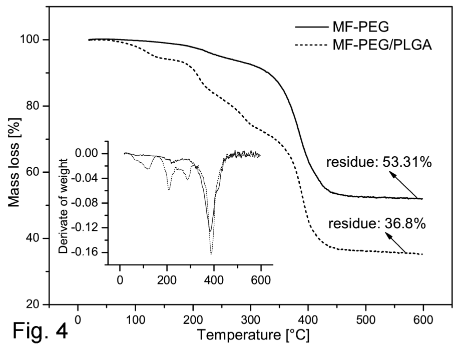


Fig. 3. FTIR spectra of magnetite, pure PLGA, pure PEG and PLGA coated MF-PEG (the spectra are shifted vertically for clarity).

Fig. 4. TGA thermogram of MF-PEG and MF-PEG encapsulated in PLGA. The inset shows differential thermogravimetric measurement.

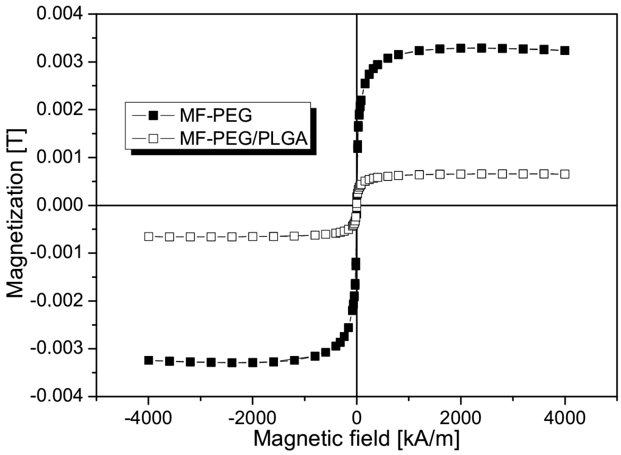


Fig. 5. Hysteresis cycles of MF-PEG and MF-PEG/PLGA at 300 K.

To investigate the magnetic properties of encapsulated MF-PEG and MF-PEG/PLGA nanospheres, the SQUID was used. From Fig. 5 it is seen that both types of materials exhibit similar overall superparamagnetic behavior. The presence of the non-magnetic shell is evident by the fact that M_S of MF-PEG/PLGA nanospheres is lower than MF-PEG. The saturation magnetization value of the MF-PEG was 3.4 mT and the MF-PEG/PLGA approximately 0.8 mT, resulting in calculated 14.5 wt% coating of PLGA, which is in good agreement with PLGA concentration obtained from TGA.

The surface modification of magnetite with PEG was a useful approach to prepare biocompatible magnetic fluid suitable for the entrapment into a hydrophobic polymer PLGA by nanoprecipitation method. The prepared magnetic nanoparticles with entrapment efficiency 22 wt% are superparamagnetic and from SQUID measurements we can calculate that the prepared nanosphere contains approximatelly ≈ 10 magnetite particles inside polymer matrix. This result was also confirmed by TEM observations. Nevertheless, the prepared magnetite nanoparticles still show sufficient magnetization for their magnetic properties to be useful from the point of view of magnetic targeting.

Acknowledgments

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References

[1] R.B. Galindo, R.S. Guerrero, O.S. Rodriguez-Fernandez, L.A. Garcia-Cerda, J. Matutes-Aquino, *J. Alloys Comp.* **369**, 87 (2004).

[2] K. Lanfester, L. Ramirez, *J. Phys., Condens. Matter* **15**, S1345 (2003).

[3] V. Závišová, M. Koneracká, O. Štrbák, N. Tomašovičová, P. Kopčanský, M. Timko, I. Vávra, *J. Magn. Magn. Mater.* **311**, 379 (2007).

[4] H. Fessi, F. Puisieux, J.P. Devissaguet, N. Ammoury, S. Benita, *Int. J. Pharm.* **55**, R1 (1989).