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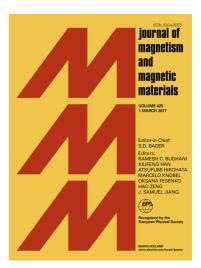
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3D-printing of novel magnetic composites based on magnetic nanoparticles and photopolymers

Norbert Löwa*, Josephine-Marie Fabert, Dirk Gutkelch, Hendrik Paysen, Olaf Kosch, Frank Wiekhorst

Physikalisch-Technische Bundesanstalt, Abbestr. 2-12, 10587 Berlin, Germany *Mail: norbert.loewa@ptb.de, Phone: +49(30) 3481 7736

A fast and cost-effective way to manufacture complex 3D structures out of numerous materials is the technique of 3D-printing employing vat photopolymerization where a liquid photopolymer in a vat is selectively cured layer by layer under radiation with light. To improve mechanical as well as functional properties different types of fillers can be added to the photopolymer. However, when fillers are added particular attention must be paid according to the quality of the resulting part (e.g. sedimentation, homogeneity, processability) which creates the need for appropriate quality evaluation. The aim of our work was to test the feasibility of printing magnetic composites which consist of photopolymers embedded with MNP. These magnetic composites are intended to be used as long-term stable phantoms in magnetic particle imaging. To assist the development of this novel imaging modality phantoms of defined geometry and magnetic properties are mandatory. With 3D printing of magnetic composites, it is possible to advance the characterization of MPI scanners with defined, long-term stable magnetic composite phantoms and opens an elegant way to print complex structures that could resemble body-like parts containing defined amounts of MNP.

Keywords: magnetic nanoparticles, magnetic particle spectroscopy, magnetic particle imaging, 3D printing, generative manufacturing, magnetic composites

1.Introduction

Magnetic nanoparticles (MNP) are of great interest in bio- and nanomedicine as they offer numerous promising therapeutic and diagnostic methods being intensively researched. A diagnostic method for the visualization of the spatial distribution of MNP, called magnetic particle imaging (MPI), has recently been introduced. MPI offers the specific measurement of MNP without any background signals from diamagnetic tissue. For MPI research, long-term stable phantoms with defined geometry and MNP content are required to determine the achievable resolution, to emulate anatomical structures, for cross-comparison of MPI scanners or as fiducial markers to verify the spatial position of the body under analysis.

A fast and cost-effective way to manufacture complex 3D structures out of numerous materials (e.g. polymers or ceramics) is the technique of generative printing, or commonly 3D-printing. This additive technique manufacturing of customized parts with complex shapes that have a voxel resolution (=smallest printable feature) in the micrometer range. Among other additive manufacturing processes, the vat photopolymerization provides the best results in terms of complexity and accuracy of resulting parts and components [STAN2016]. The process starts from a liquid photopolymer which is selectively solidified layer by layer under light radiation in a liquid vat. Using a digital micromirror device a whole photopolymer layer can be irradiated and cured within a few seconds. Thus, 3D printed polymers already have found potential applications in aerospace [KROL2011] [BOGU2013], architecture [LIPS2013], arts [SEQU2005], and medical fields [RENG2010] [MURP2014] with rather modest demands on the mechanical strength of the printed parts. To improve mechanical as well as functional properties, or even to introduce some new ones (e.g. mechanical, optical, electrical) many different fillers added to photopolymers have been tested

[WANG2017]. Even MNP deposited on microfibers have been reported as fillers. Here, these fillers act as programmable reinforcement fibers oriented by external magnetic fields [MART2015]. However, if fillers are added to a polymer, attention must be paid particularly to sedimentation phenomena, homogeneous dispersion, or alteration of the processability of the desired part.

The aim of our work was to investigate the feasibility of printing magnetic composites which consist of photopolymers embedded with MNP for manufacturing of sophisticated MPI phantoms. To this end, we developed a protocol for systematic quality evaluation of 3D-printed magnetic composites to generate complex, long-term stable MPI phantoms with defined magnetic properties (MNP amount per voxel, signal shape per voxel). Prior to the actual 3D-printing, essential parameters such as magnetic properties of basic materials, homogenization procedure, mixture ratio, polymer cross-linking, and long-term stability were characterized and optimized to improve the quality of the resultant mixture. To magnetically characterize the liquid and solidified photopolymers with embedded MNP, we measured the quasi-static and dynamic magnetization behavior by means of dc-magnetometry (M-H) and Magnetic Particle Spectroscopy (MPS), respectively. To analyze the formability and smallest printable feature of a 3D-printed magnetic composite an appropriate geometry demonstrator was developed which also considers the orientation of different structures with respect to the printing direction. Finally, preliminary MPI imaging measurements of magnetic composites with different geometries were conducted.

2. Materials and methods

2.1 Magnetic nanoparticles

We used two commercially available MNP types, namely Ferucarbotran (Meito Sangyo, JPN) as well as EFH3 (FerroTec,

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