The left-hand behaviour of polymer composites with Fe-based microwires

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Received 24 September 2013, revised 10 November 2013, accepted 11 November 2013
Published online 14 April 2014

Keywords ferromagnetic microwires, microwave absorption, metacomposites

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In the present paper, we deal with the electromagnetic properties of Fe-rich microwire polymer metacomposites prepared by embedding glass-coated Fe-based microwires into the E-glass prepreg matrices in a parallel fashion. The free-space measurement shows that a transmission window was created by adjusting the wire spacing below 7mm, which indicates that double negative index had been obtained. The correlation between the left-handed feature with the local dielectric response has been established. The 'natural' double negative behaviour is attributed to the intrinsic domain structure of Fe-based ferromagnetic wires.

1 Introduction
Metamaterials have been of a topical topic since the seminal work by Sir Pendry in 1996 [1]. The relevant studies progress rapidly and the frequency of interest extends from microwave [2] to optical frequency [3]. Although metamaterials generally refer to materials with negative permittivity and/or permeability, the majority of efforts have been spent seeking a double-negative-index characteristic in a target frequency band by periodically arranging functional units. At this point, development of metamaterials microstructure has prospered based on designing of split ring resonator (SRR) [4] and fishnet structure [5], which suffices to induce negative magnetic response in addition to the negative permittivity. However, fabrication of conventional metamaterials is highly demanding due to the adoption of micro-/nano-fabrication techniques, the metamaterial thus made is more of a structure rather than a piece of material.

Recently, ferromagnetic microwires are considered feasible as building blocks of double-negative metamaterials due to their excellent soft magnetic properties and microwave tunability towards external fields [6,7]. Besides, embedding the microwires into structural polymer matrices to formulate a what we term here as ‘metacomposite’ is of particular interest for engineering applications. These merits ensure a much simpler structure and relatively easier manufacturing process compared to conventional metamaterials. It should be noted that, unlike conventional metamaterials, the ultimate properties of metacomposites are dependent on the local properties of microwires in addition to the geometrical parameters, which offer more degrees of freedom to optimize and manipulate the composite configuration to meet varying application requirements.

As Co-based microwires and their array have demonstrated the left-handed properties [8,9], we in the present paper deal with the Fe-based microwire composites and aim to understand the physics of associated microwave characteristics.

2 Experimental
2.1 Design principle
To obtain the negative permittivity, an array of conducting fibres or even a single fibre would be able to excite such negative response as shown by the blue curve of Fig. 1. Below the cut-off frequency, i.e., electrical plasma frequency, the permittivity of the material is negative, making the material opaque to the microwave. Realization of negative magnetic permeability relies on an analogous term ‘magnetic plasma frequency’ [10] as indicated by the black curve of Fig. 1. Between the magnetic plasma frequency and the ferromagnetic resonance frequency the permeability becomes negative. Therefore, as long as the electrical plasma frequency is larger than the ferromagnetic resonance frequency, the...
double negative region would be able to occur. The widest possible double negative region can be obtained in the case of the magnetic plasma frequency equal to or larger than the electrical plasma frequency.

The physics behind these interesting observations merit detailed discussions. As the spacing is narrowed, the plasma frequency is supposed to shift to higher frequency. Clearly, in the current case, 10 mm or 7 mm spacing cannot secure an electrical plasma frequency matching the magnetic plasma frequency, while \( d = 3 \) mm meets the condition. Further, the long range dipolar interactions would be generated when the wires are close enough. The neighbouring field thus generated can significantly enhance the magnetic plasma frequency and hence the transmission window level and broadness. The application of magnetic field will increase the transmission level due to the improved impedance match at the transmission region, while at higher frequency, the application of magnetic field will enhance the skin effect and reduce the transmission window. We should however note that the level of increase is not that significant considering the field applied. This may be attributed to the relatively poor magnetic softness. It may also be the reason that why little shift of window peak was observed with increasing external field. It is worth emphasizing that, with the involvement of matrix and autoclave curing process, the present work is significantly different from Ref. [9], which deals with free-standing wires. It has been reported that the hard crystalline phase on the surface of the microwires can drastically degrade their soft magnetic properties [13]. Also the amorphous phase is generally meta-stable and can be easily modified on the surface, thereby degrading the soft magnetic properties and consequently reducing the frequency shift of the FMR peaks and transmission windows.
The simulated stress distribution of a single microwire embedded in the epoxy matrix.

To verify this, we performed a finite element modelling (FEM) simulation on the case of a single microwire embedded in epoxy. The model was created by using MSC Patran software and solved by the LS-Dyna software. Figure 3 shows that there forms significant residual stress imposed on the wire by the matrix, which arises from the difference between thermal expansion efficient of epoxy matrix, glass-coat and metallic core. These induced stresses will degrade the soft magnetic properties of Fe-based microwires. Note that the matrix effect on the Fe-based microwires is different from that on the Co-based wires [15] due to the opposite sign of magnetostriction. In terms of the domain structure, the Fe-based microwire consists of predomainted longitudinal domains while the Co-based wire is occupied by predominated circumferential domains. The longitudinal stress imposed by the matrix on the wire will benefit the circumferential domains but does not favour the longitudinal domains. In one of our previous papers [13], we reported that the stress alone is capable of inducing the formation of nanocrystallites, not to mention that the possible aging effect with the long-time annealing at constant temperature. Based upon the above analyses, one can conclude that the matrix imposes significant impact on the microstructure and magnetic structure of microwires, leading to electromagnetic behaviour of their composites that cannot be predicted based on their properties in the free-standing state. We believe that it is necessary to further study matrix-associated effects since free standing wires can hardly represent the practical case. Much work remains to be done to quantitatively describe how the curing process in autoclave influences the local properties of microwires and their collective electromagnetic responses.

4 Conclusions

The microwave properties in presence of high external field of polymer matrix composite filled with Fe$_7$Si$_{10}$B$_{10}$C$_3$ microwires in a parallel arrangement have been investigated. The prepared composites display a remarkable transmission window in the frequency band of 1 to 7 GHz without external magnetic field, indicating that the double-negative-index metacomposites are constructed. It presents unusual field effect, which is attributed to the influence of matrix and curing process.

Acknowledgements FXQ is supported under the JSPS fellowship and Grants-in-Aid for Scientific Research No. 25-03205. Yang Luo would like to acknowledge the financial support from University of Bristol Postgraduate Scholarship and China Scholarship Council. The authors would also like to thank Mr. Wang-Chang Li and Dr. Xiangqian Li of Bristol University for the useful discussions.

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