Silver-Gelatine Metal-Dielectric Composites Made From Developed X-Ray Films

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***Abstract*—The artificial dielectric properties of developed silver-gelatine X-ray film have been evaluated at microwave frequencies. The density of the silver particles in the thin films on the surface of the plastic substrate determines the real and imaginary parts of the complex permittivity. The resulting metamaterial was evaluated between 5 and 20 GHz using the split-post dielectric resonator technique. The real part of the relative permittivity of silver-gelatine mixtures varies in the range 4–170 depending on the concentration of silver. The dielectric loss tangent for such materials is in the range 0.06–0.25 and is largest for the highest permittivity samples.**

***Index Terms*—Effective complex permittivity, metal-dielectric composites, metamaterials.**

I. Introduction

METAL-DIELECTRIC metamaterials and metal-dielectric composites exhibit unusual electromagnetic properties from microwave frequencies through terahertz up to infrared frequencies [1], [2]. The most interesting properties of metamaterials (negative effective permittivity and negative effective permeability) typically appear in the vicinity of the resonance frequency of the constituent metal particles, whereas their practical applications often take place at frequencies well away from the particle resonances. One important class of metamaterials is the metal-dielectric composite, which is often labeled an artificial dielectric. Depending on the shape and orientation of the metal particles, composites of this kind exhibit a large dielectric anisotropy especially when the metal particles, in the form of thin flakes or wires, are regularly oriented. So-called planar metamaterials have a large in-plane effective permittivity and a diamagnetic (smaller than unity) effective permeability [3]–[9]. The first effect results in an enhanced dielectric constant related to the large capacitances between adjacent conductive islands. The second effect results from the eddy currents induced in the conductive islands. In some cases, metal-dielectric materials (e.g., consisting of split metal rings) exhibit simultaneously negative permittivity and negative permeability [1], [2], [9]. Silver-gelatine films may find applications, e.g., in construction of flat microwave lenses [10], as well as may serve as reference samples in near-field scanning microwave microscopes [11]. The manufacturing of metal dielectric mixtures with a uniform distribution of metal particles is rather difficult. We have noticed that X-ray films exhibit significant differences in their dielectric properties depending on their transparency as observed in grayscale. Because the light-sensitive layers of such films are made from silver-gelatine mixtures, they in fact create artificial dielectrics. The more silver particles in the gelatine, the darker the color of the film. Therefore, by developing X-ray films subjected to different X-ray exposures, it is possible to create metal-dielectric composite with well-controlled silver content.

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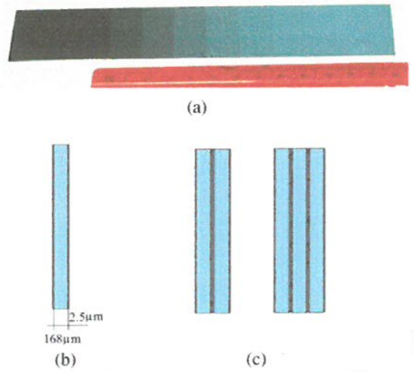

Fig. 1. (a) Test sample in the form of a rectangular sheet made from developed X-ray film with several density steps shown in grayscale. (b) Cross section of the same sample showing silver-gelatine films deposited on both sides of a polyethylene terephthalate (PET) substrate. (c) Stacked samples.

II. Measurements

Fig. 1 shows a test sample of such a material with different levels of silver content shown by several grayscale steps.

The silver-gelatine film thickness was determined by measurement of the height of the step after removing a small area of gelatine from the X-ray film as shown in Fig. 2. The thickness of the single silver-gelatine layers was found to be almost the same for the different steps and in the range 2.2–2.8 μm. The size of the quasi-spherical silver particles in the gelatine layer is in the range 1–2.5 μm. The particles are approximately uniformly distributed within each specific step (as shown in Fig. 3 for three different steps).

The complex permittivity was measured for a few particular density steps using split-post dielectric resonators (SPDRs) [7], [8] at both 12 and 21.5 GHz. The 12-GHz resonator permits the measurement of samples with minimum lateral dimensions of 10×10 mm^2, while the 21.5-GHz

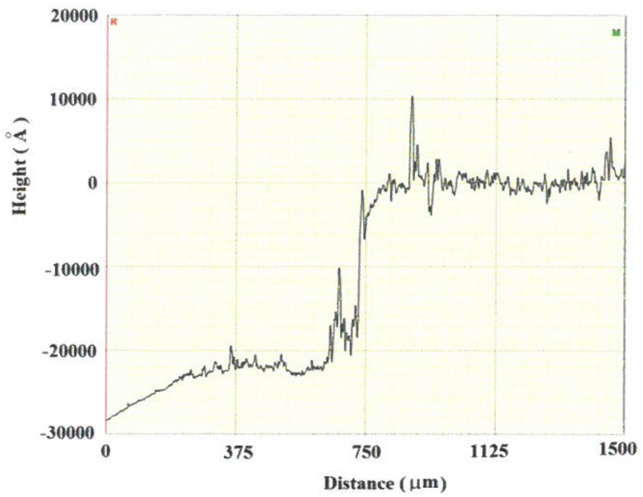

Fig. 2. Thickness measurement of the silver-gelatine layer employing Dektak 150 Surface Profiler (Veeco).

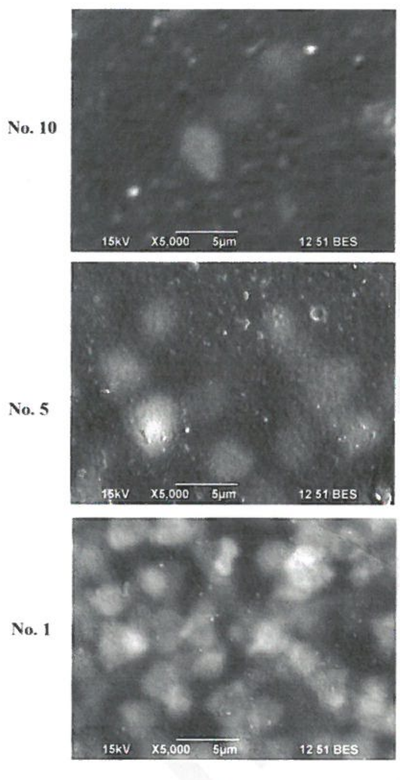

Fig. 3. Scanning electron microscope (SEM) picture of three steps of the X-ray film from Fig. 1(b). The darkest step has number 1, the middle number 5, and the brightest number 10. The white color regions correspond to the Ag particles. In these pictures, silver particles have white color.

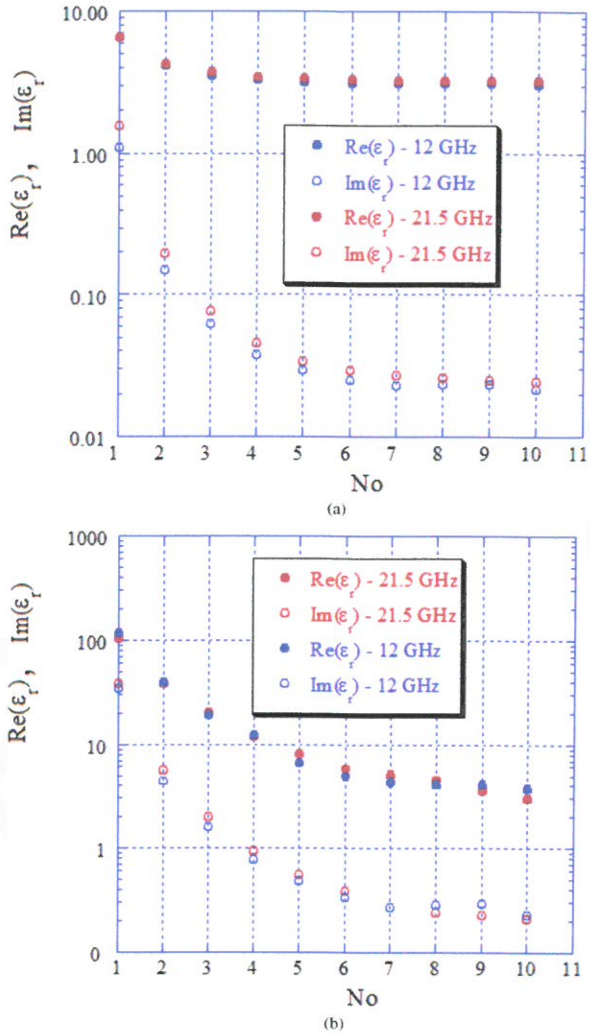

Fig. 4. Permittivity of the rectangular test sample shown in Fig. 1, determined for samples with an effective thickness of (a) 172 μm (the total thickness of the PET substrate and the gelatine films) and (b) 5 μm (the thickness of two gelatine films), respectively.

resonator permits measurement of samples as small as 8×8 mm^2. The results are presented in Figs. 4 and 5. The permittivity values shown in Fig. 4(a) were evaluated assuming that the whole sample, with a total thickness of 172 μm, creates a uniform metamaterial, whereas the permittivity values shown in Fig. 4(b) were evaluated assuming a thickness of 5 μm. This is the thickness of the two silver-gelatine films deposited on each side of the PET substrate. In this case, reference values of the resonance frequency and the Q-factor were measured for a piece of blank PET substrate with gelatine layers removed using hot water. It should be mentioned that the electric field in the SPDR has only an azimuthal component that is tangential to the plane of the film, so the PET substrate and the two silver-gelatine films act as three capacitors connected in parallel. For the silver-gelatine film where the silver concentration is largest, the real part of the permittivity of films exceeds 100 and is slightly frequency-dependent, as seen in Fig. 5. The imaginary part of the permittivity is about 10 times smaller than the real part for low concentrations of silver. The losses substantially increase for the samples with larger concentrations of silver. Measurements that were performed at 5 GHz were only possible on the very darkest sample because its dimensions were sufficiently large for our 5-GHz SPDR (samples for 5-GHz SPDR must cover a circle of 26 mm diameter). A width of 15 mm for the gray steps of our original sample was too small

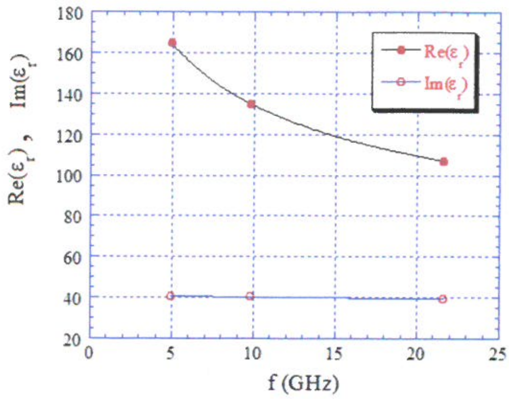

Fig. 5. Permittivity versus frequency for the very darkest sample of X-ray film (i.e., the film with the largest concentration of silver particles) evaluated with an effective thickness of 5 μm.

TABLE I
PERMITTIVITY AT 4.9 GHz FOR THE STACKED FILMS WITH THE LARGEST CONCENTRATION OF SILVER

	PET+silver+gelatine			Silver+gelatine		
Sample	h (μm)	Re(ε_r)	Im(ε_r)	h (μm)	Re(ε_r)	Im(ε_r)
S1	172	8.07	1.28	5	165.1	40.4
S2	172	7.46	0.94	5	145.3	29.8
S3	172	8.24	1.39	5	170.8	43.6
S1/S3	344	8.07	1.52	10	165.2	47.7
S1/S2/S3	516	7.94	1.07	15	159.3	33.4

for performing measurements at 5 GHz, so in Fig. 4, there are only two frequency data.

Additional measurements were also performed on stacked darkest films as shown in Fig. 1(c). The complex permittivities for both single and stacked films are listed in Table I. It is apparent that the permittivity of the stacked films is almost the same as the average permittivity of the individual films within experimental error. We have also attempted to measure the magnetic properties of the darkest films cutting from them small 6-mm-diameter samples and performing subsequent measurements on the small and the large samples in 4.9-GHz SPDR. Details of the measurement technique are described in [7]. These measurements proved that the real part of the relative permeability of our films is approximately equal to unity (to within experimental errors) and that the imaginary part of the relative permeability is equal to 0.01 (+/0.005). Such result is not unexpected because, as it is seen in Fig. 3, silver particles of our composites are very small (average diameter of 2 μm), and therefore the magnetic moments induced in them by an external magnetic field are also small, which in turns does not result in a significant diamagnetism.

III. CONCLUSION

It has been shown that ordinary X-ray film with silver-gelatine mixtures exhibits properties of an artificial dielectric at microwave frequencies. Microscopic inspection of such films shows that silver particles have quasi-spherical shape and average size the order of 2 μm. The real part of the relative permittivity for such mixtures varies in the range 4–170 depending on the concentration of silver. The dielectric loss tangent for such materials is in the range 0.06–0.25 and is largest for the highest permittivity samples. The main advantage of such materials is their simplicity and the low costs of their preparation. Furthermore, it is easy to control permittivity profiles of such films by different X-ray exposures. Due to small size of silver particles, films having thickness of a few microns behave rather as bulk artificial dielectrics than as metasurfaces. This has been experimentally confirmed by closely stacking films together. The effective permittivity of stacked films proved to be the same, to within experimental errors, as the effective permittivity of individual films. This means that the local electromagnetic fields do not extend noticeably beyond film surfaces, and therefore the effective permittivity of closely stacked films remains almost unchanged with respect to the effective permittivity of individual films. Silver particles in the our films are not regularly distributed, but they exhibit some degree of order in the sense of similar size and shape of the particles as well as similar distances between them for the fixed X-ray exposure.

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