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Journal of Aerospace Technology and Management, vol. 2, núm. 1, enero-abril, 2010, pp. 63-70
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Available in: http://www.redalyc.org/articulo.oa?id=309426552008
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Abstract: This paper presents the processing and characterization of electromagnetic radiation absorbing paints and sheets based on magnetic and dielectric materials dispersed in polymeric matrices. Two different paint formulations containing carbonyl iron and/or polyaniline, using polyurethane as matrix, were prepared. Silicone sheets were also produced with polyaniline conducting polymer as filler. Measurements of the electric permittivity and magnetic permeability of the materials were also carried out. Simulations for the silicone sheets were performed in order to correlate the electromagnetic parameters with the material thickness. The paints absorbed 60 to 80% of the incident electromagnetic radiation and the silicone sheets absorbed 90%, indicating the material’s radar absorbing potential.

Keywords: Absorbing media, Radar absorbing material, Conducting polymer, Dielectric materials, Magnetic materials.

INTRODUCTION

Electromagnetic radiation absorbing materials or Radar Absorbing Materials (RAMs) have been the focus of much research due to increasing regulation to control the levels of electromagnetic radiation emitted by electronic equipment, and also to new norms and standards issued regarding compatibility and electromagnetic interference produced by this type of equipment. RAMs are also important tools in electronic warfare, since they can be used to camouflage potential targets from radar detection. Furthermore, microwave absorbers have been widely used to prevent or minimize electromagnetic reflections from large structures such as aircraft, ships, and tanks and to cover the walls of anechoic chambers (Stepanov, 1968; Emerson, 1973; Lawrence, 2000; Hemming, 2002). RAMs can be produced in different forms such as paints, sheets, and thin films (Lee, 1991; Olmedo, Hourquebie, Jousse, 1997; Skotheim, Elsenbaumer, Reynolds, 1998; Chandrasekhar, 1999; Folgueras, Rezende, 2008). Usually, these materials are obtained by the dispersion of one or more types of absorbing fillers in a polymeric matrix, which is then applied onto a substrate. Understanding the methods to produce RAMs by combining components, additives and polymeric matrices is decisive on the final application of the resulting material. Depending on the electromagnetic properties, the material can be either used as an absorber or a reflector of electromagnetic radiation (Afsar et al. 1986; Knott, Shaffer, Tuley, 2004). The need for RAMs as paints has increased as a result of the new civilian and military applications found for these materials. The use of materials with specific characteristics and new processes enable developing RAMs with special physical properties, resulting in paints that respond differently to electromagnetic radiation. Materials used as RAMs have dielectric and magnetic losses, and the dependence of these losses on frequency is responsible for their performance, resulting in the absorption and/or scattering of electromagnetic waves. An ideal absorber might comprise a layer of material with numerically equal values of complex permeability and permittivity and high loss tangents over a wide range of frequencies. The former ensures a perfect impedance match with air, thus enabling incident signals to enter the material without front-face reflection, and the latter promotes rapid attenuation afterwards. In ferrites, the complex permeability is frequency-dependent; the dispersion is caused by the high-frequency magnetization reversal processes: the rotation of the magnetization vector and movement of domain boundaries. In dielectric materials, such as polyaniline, the complex permittivity of a material is related to its dielectric conductive properties (Naito, Suetake, 1971; Lee, 1991; McCurrie, 1994; Olmedo, Hourquebie, Jousse, 1997).

Electric permittivity ($\varepsilon$) and magnetic permeability ($\mu$) are parameters related to a material’s dielectric and magnetic properties; they are among the most important characteristics of absorbing materials, and are directly associated with their absorbing properties (Hippel, 1954; Balani, 1989; Clark et al. 1995; Chen et al., 2004). The
relative permittivity and permeability are represented by Equations 1 and 2, respectively; the values of these parameters are obtained from the experimental values of the transmission and reflection coefficients of the material.

\[ \varepsilon_r = \varepsilon' - i\varepsilon'' \]  \hspace{1cm} (1)

\[ \mu_r = \mu' - i\mu'' \]  \hspace{1cm} (2)

When the material is lossy, some of the incident electromagnetic energy is dissipated, its permittivity and permeability are complex: Equations 1 and 2 (Sucher, Fox, 1963; Kovetz, 2000; Daniels, 2007; Orfanidis, 2009) show the real \((\varepsilon', \mu')\) and imaginary components \((\varepsilon'', \mu'')\).

In the case of a magnetic material, losses are produced by changes in the alignment and rotation of the magnetization spin (Jarem, Johnson, Scott, 1995; Thostenson, Chou, 1999; Johnson, 2004).

The determination of \(\mu\) and \(\varepsilon\) of a material is usually based on measurements of complex electromagnetic parameters (S parameters), the reflection and transmission coefficients \((S_{11}/S_{22}\) and \(S_{12}/S_{21}\), respectively), using a vector network analyzer. To analyze a material based on the value of these parameters, it is necessary to take into account other variables such as frequency band, intrinsic electrical properties of the material and its thickness (Fig. 1).

![Diagram of a RAM showing the influence of thickness and other parameters.](image)

Figure 1: Diagram of a RAM showing the influence of thickness and other parameters.

The amplitude of an electromagnetic wave that propagates through a conducting medium is attenuated as the wave advances into the medium (Fig. 1) (Balanis, 1989; Lima, 2005). The electromagnetic penetration depth (called skin depth) and electric field attenuation at the surface of the material are important parameters in the production of RAMs (Quéffelec, Le Floc’h, Gelin, 1998; Guru, Hiziroglu, 2004).

Materials with high conductivity and limited ability to store energy, such as metals, have high dielectric losses. In this case, the penetration depth approaches zero and the material has reflector-like characteristics. In materials with low dielectric losses, the penetration depth is larger and, as a result, little energy is absorbed by the medium, rendering the material transparent to electromagnetic radiation. In the case of absorbing materials, most of the electromagnetic energy is attenuated. The attenuation results from the interplay of several factors such as electric conductivity, dielectric loss and electromagnetic penetration depth, which need to be less than the thickness of the material (Thostenson, Chou, 1999; Oh et al., 2004; Rmili et al, 2005).

Organic polymers that conduct electricity are a class of polymers often referred to as “synthetic metals” due to their ability to combine the chemical and mechanical properties of polymers with the electrical properties of metals and semiconductors (Anand, Palaniappan, Sathyanarayana, 1998; Falcou et al., 2005).

Absorbing materials can be grouped into two categories: narrowband (or resonant) and wideband absorbers. Resonant materials are more common, while the wideband ones are produced by the combination of different materials (Ruck et al., 1970). The main application of both materials is essentially the same, i.e., the absorption of electromagnetic radiation. Thin sheet absorbers are prepared by dispersing a lossy material over a matrix. The absorption obtained depends on the thickness and the absorption mechanisms are independent of each other. By choosing a thickness that is a match to complex permittivity and permeability, the absorption bandwidth can be considerably increased, both at normal and oblique incidence of the electromagnetic wave (Naito, 1970; Naito, Suetake, 1970; Nicolson, Ross, 1970; Miller, 1986; Musal, Hahn, 1989).

The absorption of electromagnetic energy of a single-layer RAM as a function of frequency can be calculated analytically using Equations 3 and 4 (Balanis, 1989; Chen et al., 2004).

\[ R(dB) = 20 \log_{10} \left( \frac{iA\tan(kd) - 1}{iA\tan(kd) + 1} \right) \]  \hspace{1cm} (3)

where

\[ A = \sqrt{\mu/\varepsilon}, \]  \hspace{1cm} \[ k = \frac{2\pi f}{c} \sqrt{\mu\varepsilon}, \]  \hspace{1cm} and \( i = \sqrt{-1} \).  \hspace{1cm} (4)

In (3) and (4), \(\mu\) and \(\varepsilon\) are, respectively, the complex permeability and permittivity of the absorbing material,
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$k$ is the wave number, $f$ is the frequency of the incident wave, $c$ is the speed of light in vacuum, and $d$ is the thickness of the absorbing layer. Both the permeability and permittivity vary along with the frequency.

In this context, the main objective of this work was to describe how we produce RAMs in the form of paints and sheets to absorb electromagnetic radiation in the frequency range of 8-12 GHz (X-band).

**EXPERIMENTAL**

**A. Production of the paints**

Two different absorbing centers were used to produce the absorbing paints: conducting polyaniline, which behaves as a dielectric; and commercial-grade carbonyl iron powder, a magnetic material. Polyaniline was synthesized at laboratory scale in the Materials Division of IAE. Briefly, the process consisted of the oxidization of aniline by ammonium persulfate in an acidic medium (dodecylbenzenesulfonic acid). The resulting polyaniline was obtained as a conducting powder (MacDiarmid et al., 1984; Skotheim, Elsenbaumer, Reynolds, 1998; Rannou et al., 1999; Mattoso, MacDiarmid, Epstein, 1994; Folgueras, Rezende, 2008).

Two paint formulations were prepared: one consisting of carbonyl iron powder (90% w/w) dispersed in the polyurethane matrix, and the other of carbonyl iron powder and polyaniline (15% w/w) dispersed in the same polymeric matrix. The absorbing fillers were mixed with the matrix material by mechanical agitation for 30 minutes. Next, the resulting material was applied to flat aluminum plates (20 x 20 cm) with a brush. The thickness of the paint layer containing only carbonyl iron was 1.10 mm, the paint layer containing carbonyl iron and polyaniline had a thickness of 1.85 mm. In order to evaluate some of the properties of these paints, they were also applied to a polymeric substrate, which facilitated their removal for further analyses. All paints were cured at room temperature.

**B. Production of the silicone sheets**

The doped polyaniline powder (17% w/w) was added to a matrix composed of two types of silicone rubber, L9000 and RTV630 (GE Silicones). The mixture was homogenized by mechanical agitation, and the processed materials were poured into (30 x 30 cm) molds and dried at 70°C. After this, two types of sheet forms were obtained, each corresponding to the different types of silicone rubber used. The sheet produced with L9000 silicone rubber had a thickness of 2.80 mm, and the one produced with RTV630 silicone rubber was 4.40 mm thick.

**C. Electromagnetic measurements**

For the electromagnetic characterization of the paints, reflection/absorption measurements in the frequency range of 8-12 GHz were carried out using the Naval Research Laboratory (NRL) arch method (Skolnik, 1970; Knott, Shaffer, Tuley, 2004). The NRL arch (Fig. 2) consists of a wooden structure in the shape of a semicircular arch, enabling the proper positioning for emitting and receiving antennas (horn type). The samples are placed at the center of the arch curvature; first, the antennas are positioned at the highest position in the arch, and then each antenna is moved 10° to each side of this position. The antennas always pointed to the center of the sample. The setup also included a spectrum analyzer (Anritsu, model MS 2668C) and a frequency generator (Agilent Technologies, model 83752A). A flat aluminum plate was used as a reference for the reflection/absorption measurements; its reflectivity and absorptivity were considered to be 100% and 0%, respectively. The main advantage of the NRL method with respect to the others such as the waveguide method is that it allows measuring the properties of relatively large samples in free space conditions.

The transmission line technique (with a waveguide) was used to measure the complex electric permittivity and magnetic permeability of the processed paints in the microwave frequency range of 8-12 GHz (X-band). A closed waveguide (rectangular cross section) was coupled to a network vector analyzer (Agilent Technologies, model 8510C), an S-parameter tester (Hewlett Packard, model 8510A) and a synthesized frequency generator, both operating in the frequency range of 45 MHz – 26 GHz. This setup measured the S-parameters of the material;
the transmission and reflection coefficients $S_{12}$ / $S_{21}$ and $S_{11}$ / $S_{22}$, respectively. Commercial software (Agilent Technologies – 85071E) was used to calculate the values of the complex permittivity and magnetic permeability as functions of the frequency. Figure 3 shows the setup used in the measurements.

Figure 3: The transmission line technique setup: (a) vector network analyzer, (b) closed system rectangular waveguide and the position of the sample within the waveguide.

RESULTS AND DISCUSSION

Figure 4 shows the appearance of the processed paints applied to a polymeric substrate. The polymeric substrate was used to enable removing the layer of paint undamaged for further analyses. It was observed that the paint containing polyaniline and carbonyl iron reflected more light and was darker than the paint produced with only carbonyl iron. After their removal from the substrates, the paint samples were weighed, being verified that the polyaniline paint had a mass smaller than the one containing carbonyl iron. This was expected since the specific mass of carbonyl iron (7.8 g/cm$^3$) is larger than that of polyaniline (~1.2 g/cm$^3$).

Figure 5 and 6 show the measurements of the attenuation of electromagnetic radiation and the complex electric permittivity, $\varepsilon$, and magnetic permeability, $\mu$, of the paints produced. Figures 5(a) and 6(a) show that both paints acted as RAMs in the frequency range used in this study. The paint containing only carbonyl iron (Fig. 5a) attenuated the incident wave by about 7 dB, which corresponds to an absorption of 80% of the electromagnetic energy. The paint containing both polyaniline and carbonyl iron (Fig. 6a) attenuated the wave by about 4 dB, corresponding to about 60% of absorption of the electromagnetic energy.

Figure 5b shows the measured values of the electric permittivity of the carbonyl iron paint, about 16.0 and 10.0 for the real and imaginary parts, respectively. The permeability values for this paint were 4.0 and approximately zero for the real and imaginary parts, respectively. It is known that the larger the value of the imaginary component of permittivity ($\varepsilon''$), the larger the losses of the material. Thus, a material with low dielectric loss can store energy, but will not dissipate much of the stored energy. On the other hand, a material with high electric losses does not store energy efficiently; a certain amount of energy will be transformed into heat within the material. In general, the smaller the magnetic permeability, the larger the resonance frequency, in which the material exhibits good absorption properties; but for frequencies higher than 2 GHz, the permeability is related to the energy anisotropy in the material (Bady, 1969; Vinoy, Jha, 1996; Jiu et al., 2004).

Figure 4: Paints on a plastic substrate. Formulations containing (a) carbonyl iron and polyaniline, (b) thickness of the paint layer, (c) carbonyl iron only and (d) paint peeled off the metal plate for measurements.

Figure 5: Electromagnetic characteristics of the paint produced with carbonyl iron. (a) Attenuation of the incident radiation, and (b) relative electric permittivity, $\varepsilon$, and magnetic permeability, $\mu$. The prime and double primes refer to real and imaginary values, respectively.
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Figure 6b shows the relative values of permittivity and permeability of the paint produced with polyaniline and carbonyl iron. The measured values of the real and imaginary parts of the electric permittivity were 5.5 and 1.5, respectively; and the measured values of the real and imaginary parts of the magnetic permeability were 1.2 and 0.3, respectively. This paint can be considered a hybrid absorbing material since it has dielectric and magnetic characteristics (Lee, 1991; Knott, Shaffer, Tuley, 2004). For this paint, the permittivity values are smaller than those measured for the carbonyl iron paint. This is due to a superposition of effects from the magnetic phenomena associated with the presence of the carbonyl iron and the electronic polarization of the polyaniline molecules, which act on the incident electromagnetic wave.

When this material was evaluated using the back metal plate, it absorbed 88% of the incident energy; thus, 64% of the incident energy was attenuated due to physical processes occurring within the material, and 29% of the attenuation was caused by the intrinsic properties of the absorbing material. Similarly, Figure 9 (material with 4.40 mm) shows that the reflected and transmitted energy values were 42% and 35%, respectively. When this material was applied to a metal plate, 71% of the energy was absorbed; in this case 23% of the energy was absorbed intrinsically.

Figure 7 shows the aspect of the processed silicon sheets absorbers. Both types of different substrates (L9000 and RTV 630) used for processing the absorbing material demonstrated similar aspects.

Figure 8 depicts the results derived from the S-parameters measurements (reflected, transmitted and absorbed energy) for the two silicone sheets produced in this study. By this coefficient, its possible energy value was absorbed intrinsically by the material. It can be observed (material with 2.80 mm) that the reflected and transmitted energy were, on average, equal to 45% and 26%, respectively,
The dielectric properties of the absorbing silicone sheets are shown in Figures 10 and 11. It is obvious that these materials have distinct electromagnetic properties. The average real and imaginary values of the relative permittivity are close to 6.0 and 2.0, respectively, for the material produced with the silicone rubber L9000 (Fig. 9). For the material produced with the silicone rubber RTV630 (Fig. 10), the average real and imaginary permittivity values are 3.5 and 5.0, respectively.

**CONCLUSIONS**

Based on the results obtained in this study, we conclude that the paints produced have the potential to be used as RAMs, since they attenuated 60 to 85% of the incident electromagnetic radiation. The attenuation measured for the paint containing conducting polyaniline can be explained by the fact that when this polymer is surrounded by a matrix, conduction paths are formed in the material, allowing the dissipation of energy due to electrical losses. The carbonyl iron in the paints also contributed to the dissipation of electromagnetic energy due to magnetic anisotropy effects, a characteristic of this material for frequencies larger than 2 GHz.

The sheets produced with a silicone matrix attenuated the incident radiation to approximately 90%, demonstrating that these materials can be used as absorbers of electromagnetic radiation. Also, the analytical calculations demonstrated the importance of optimization tools to produce absorbing materials with the required properties.

The attenuation behavior of the paints suggests that their electric conductivity is related to the type of absorbing center and insulating material (matrix) used in the paint formulation, which modify the impedance of the RAM and the ability to attenuate the incident radiation.

An important characteristic of the materials produced is their low density (no larger than 1 g/cm$^3$) compared to conventional absorbers based on ferrites (absorption 10 dB), with densities ranging from 4 to 5 g/cm$^3$.

**ACKNOWLEDGMENTS**

The authors wish to thank the Departamento de Ciência e Tecnologia Aeroespacial (DCTA)/Instituto de Aeronáutica e Espaço for the technical and financial support and also the Brazilian government funding agencies CNPq (Projects numbers: 559246/2008-0, 151803/2008-0 and 150048/2010-6) and FINEP (Project number: 1757/03) for the financial support.
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