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Green Nanocomposite-Based Metamaterial Electromagnetic Absorbers: Potential, Current Developments and Future Perspectives

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ABSTRACT The use of the natural materials instead of conventional materials as electromagnetic absorbers promotes environmental sustainability, cost-effectiveness, and ease of accessibility. Furthermore, these materials may also be designed as absorbers and as reinforcements in building materials in a lightweight form. The absorbing ability of composite materials can be customized based on the chosen fillers. Specifically, magnetic and dielectric fillers can be incorporated to improve the absorption of a composite material compared to traditional materials. This work aims to review recent developments of electromagnetic absorbers enabled by nanocomposites, metamaterial and metasurface-based, as well as green composite alternatives. First, the background concepts of electromagnetic wave absorption and reflection will be presented, followed by the assessment techniques in determining electromagnetic properties of absorbing materials. Next, the state-of-the-art absorbers utilizing different materials will be presented and their performances are compared. This review concludes with a special focus on the future perspective of the potential of metamaterial based nanocellulose composites as ultrathin and broadband electromagnetic absorbers.

INDEX TERMS Electromagnetic, microwave absorber, nanocomposites, metamaterial, nanocellulose.

I. INTRODUCTION

In recent years, the development of wireless technologies with the particular aim at achieving higher communication throughput and wider bandwidths for high-speed communication have been developed rapidly to accommodate consumers' needs. However, the wide use of wireless communication systems has also resulted in an increasing electromagnetic interference (EMI) to the environment. The conducted or radiated electromagnetic signals interferes with the operation of other electronic devices [1] which can cause malfunction of the devices and even can be harmful to human [2]. This is becoming a major concern, raising the need for mechanisms which shields, absorbs or protects human, environment, devices and etc. against these unwanted electromagnetic signals.

To address this problem, several research and development efforts on electromagnetic absorbers has been reported, mainly to attenuate or absorb these unwanted electromagnetic signals [3], [4]. Besides that, absorbers are also used in a wide range of applications to recreate a free space environment by eliminating reflection in an anechoic chamber. The

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proper choice of absorbing material is an effective way to improve the performance of the absorber [3]–[5].

Several reviews have reported electromagnetic wave absorber materials based polymer composites [4] and [3] and metamaterial structures focused on the optical frequencies [6]. However, to the best of our knowledge, none of previous reviews focused on the different state-of-the-art green electromagnetic absorbers or the combination of metamaterial-based structures and green absorbers as alternatives electromagnetic absorbing materials in microwave region. Prior to that, a review of the theoretical aspects of absorption and its assessment techniques will be presented, followed by the recent developments of nanocomposite absorbing materials, metamaterial and metasurface-based absorbers and green composite absorbers, and applications. Microwave absorbing material is the material with microwave absorbing capabilities mainly defines by its electromagnetic parameters and focuses on the material's chemical compositions. Whereas microwave absorber is a structure normally integrate with absorbing material to absorb microwave. Both functions to absorb microwave with the latter usually focuses on the shape or design of the structure to improve the absorption. This paper is organized as follows. First, an overview of the theory and mechanism of electromagnetic wave absorption will be described. Next, the recent development of emerging absorbing materials/techniques such as nanocomposites and metamaterial-based absorbers will be discussed. This is followed by the presentation of the recent methods towards enhancing microwave absorption based on alternative green and organic materials in electromagnetic absorption. A special focused discussion will be provided on nanocellulose-based composites. Its production methods and potential applications will be discussed, prior to the discussion on the future perspective of such electromagnetic absorbing material.

II. THEORY OF ELECTROMAGNETIC ABSORPTION

In this section, the fundamentals of electromagnetic wave absorption are first presented. This includes the theory of absorption and electromagnetic parameters, configuration of a microwave absorber and principle of skin depth effect.

A. ABSORPTION AND ELECTROMAGNETIC PARAMETERS

Absorption of an electromagnetic wave is defined as the ability of materials to attenuate or absorb the electromagnetic radiation inside the materials. Fig. 1 shows the electromagnetic wave attenuation through a material with different mechanism of shielding.

Defined in terms of S parameters, the transmittance (T), reflectance (R), and absorbance (A) through the shielding material can be written as follows;

$$\mathrm{T} = S_{12}^2 = S_{21}^2 \tag{1}$$

$$\mathrm{R} = S_{11}^2 = S_{22}^2 \tag{2}$$

$$\mathrm{A} = 1 - \mathrm{R} - \mathrm{T}. \tag{3}$$

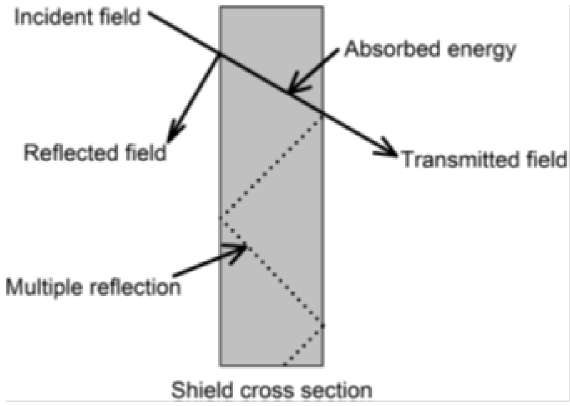


FIGURE 1. Reflection and transmission mechanisms of a material.

As an electromagnetic wave travelling in free space penetrates into a material, the wave will be reflected, transmitted or absorbed. Wave interaction and consequently, absorption properties of a material are usually determined by two material parameters; permittivity and permeability. Permittivity and permeability relate to a material's ability to transmit an electric fields and magnetic fields, respectively [3], [5]. Based on these parameters, materials are classified as either dielectric or magnetic. The complex permittivity ($\varepsilon^* = \varepsilon' - j\varepsilon''$) and complex permeability ($\mu^* = \mu' - j\mu''$) describe the interactions of electromagnetic wave through a material. The real parts of complex permittivity and permeability (ε', μ') are associated with electric and magnetic energy storage, whereas the imaginary parts (ε'', μ'') represent the dielectric and magnetic loss or energy dissipated within a material. The ratio of imaginary parts (energy loss) to the real parts (energy stored) is defined as the loss tangent of the material. The electric loss tangent or dielectric loss is defined by [3];

$$\tan \delta_\varepsilon = \frac{\varepsilon''}{\varepsilon'} \tag{4}$$

Meanwhile the magnetic loss is defined by [3];

$$\tan \delta_\mu = \frac{\mu''}{\mu'} \tag{5}$$

where, δ is the loss angle of the material. Thus, for an electromagnetic wave absorber, high imaginary parts of complex permittivity and permeability will enable greater absorption.

B. LAYER CONFIGURATIONS IN EM ABSORBERS

Most researches performed on absorbers have been aimed at increasing the absorption range and bandwidth by using multilayered material structures. Electromagnetic wave absorber can be designed in a multilayered configuration, depending on its application. In recent years, electromagnetic wave absorbing materials with broader frequency range and lower reflection losses have been realized using such structures. It is also well-known that single layer absorbing materials are hardly able to absorb in a wide frequency range. Fig. 2 and 3 illustrate the structure of a single layer and

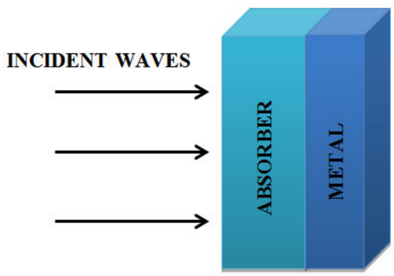


FIGURE 2. Structure of single layer absorber.

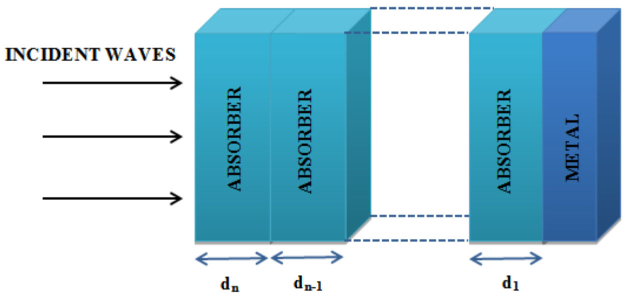


FIGURE 3. Structure of a multilayered absorber.

multilayered electromagnetic wave absorbing material, respectively. In dual- or multilayered structures, a matching layer and an absorbing layer are used in combination to satisfy both the impedance matching characteristic and attenuation characteristics. Therefore, many researches have utilized more than a layer of absorbing materials to enhance these aspects.

Based on the transmission line theory; the impedance for a single layer electromagnetic wave absorbing material is given by [7], [8]:

$$Z_1 = \eta_1 \tanh(\gamma_1 d_1) \tag{6}$$

where η_1 is the effective input impedance from;

$$\eta_1 = \eta_0 \sqrt{\frac{\mu_1}{\varepsilon_1}} \tag{7}$$

and γ_1 propagation constant can be written as;

$$\gamma_1 = j\frac{2\pi}{c} f \sqrt{\mu_{r1}\varepsilon_{r1}} \tag{8}$$

where d_1 is the thickness of absorbing material, c is the speed of light in air, η_0 is the characteristic impedance of the free space, ε_{r1} is the complex permittivity and μ_{r1} is the complex permeability.

For multi-layered electromagnetic absorbing material with$n$ layers of different materials, the impedance of the nth layer is defined by [4], [9];

$$Z_n = \eta_n \frac{Z_{n-1} + \eta_n \tanh(\gamma_n d_n)}{\eta_n + Z_{n-1}\tanh(\gamma_n d_n)} \tag{9}$$

where η_n and γ_n are given by

$$\eta_n = \eta_0 \sqrt{\frac{\mu_{rn}}{\varepsilon_{rn}}} \tag{10}$$

$$\gamma_n = j\frac{2\pi}{c} f \sqrt{\mu_{rn}\varepsilon_{rn}} \tag{11}$$

Reflection loss (RL) is commonly used to evaluate the absorption capacity of materials/absorbers. The RL of electromagnetic wave at the absorbing material surface is given by [4];

$$RL = 20 \log |\Gamma| = 20 \log |\frac{Z_n - \eta_0}{Z_n + \eta_0}| \tag{12}$$

where Γ is the reflection coefficient. Combination of both the magnetic permeability and electric permittivity satisfying the impedance matching condition is the key to produce a high-performance absorber. The desirable absorbing material properties should exhibit impedance matching with equal permeability and permittivity [9].

C. PRINCIPLE OF SKIN DEPTH EFFECT

Skin depth is a measure the depth of electromagnetic wave propagation and penetration into a material. Skin depth decreases with the increase of the frequency and the effective dielectric and magnetic loss factor. The higher the skin depth value, the lower the microwave electrical conductivity. This indicates the higher ability of the electromagnetic field to propagate across the material. Skin depth (δ) is a function of frequency (f), resistivity (ρ) and relative permeability (μ_r) define as [3], [10];

$$\delta = \sqrt{\frac{2\rho}{2\pi f \mu_0 \mu_r}} \tag{13}$$

where μ_0 is the permeability of free space (4π x 10^{-7} H/m). For a perfect conductor, resistivity is zero and therefore the skin depth is zero. Resistivity is a measure of how resistive a material is and it is the reciprocal of conductivity. Thus the equation before can be expressed as [3], [10];

$$\delta = \frac{1}{\sqrt{\pi f \mu_0 \mu_r \sigma}} \tag{14}$$

On the other hand, electromagnetic field that propagates within the material can be expressed as [4];

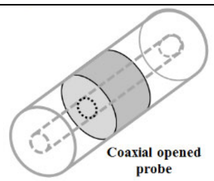
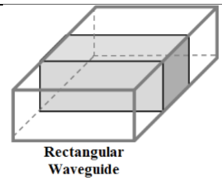
$$\lambda = \frac{\lambda_0}{\sqrt{|\varepsilon||\mu|}} \tag{15}$$

where $\boldsymbol{\lambda_0}$ is the wavelength in free space and $|\varepsilon|$ and $|\mu|$ are the modulus of ε and μ, respectively. The maximum reflection loss is associated with a quarter wavelength (0.25λ) thickness of the material.

III. MEASUREMENTS OF THE EM PROPERTIES OF MATERIALS

There are several methods to measure electromagnetic properties of a material, specifically permittivity and permeability. Accurate measurements of these parameters are critical in modelling the performance of microwave absorbers. This paper presents three most common methods applied for this purpose, namely the transmission line method, coaxial probe method and free space method. These methods involve the transmission of waves into the material and measuring the material response within a selected range of frequencies.

TABLE 1. Comparison between coaxial and waveguide transmission line methods [11].

Transmission line	Coaxial	Waveguide
Structure		
Sample holder	Sample holder has precision connector, port and a precision center conductor	Sample holder is a section of waveguide transmission line
Sample	Complex sample fabrication	Sample is brick-shaped; easier sample fabrication
Frequency	Broadband	Limited frequency band

A. TRANSMISSION LINE METHOD

Transmission line method requires the material under test (MUT) to be placed inside a portion of an enclosed transmission line. The line is usually a section of rectangular waveguide or coaxial airline. The complex permittivity (ε_r*) and permeability (μ_r*) are then computed from the measurement of the reflected signal (S_{11}) and transmitted signal (S_{21}).

B. COAXIAL PROBE METHOD

An open-ended coaxial probe is built using a section of a transmission line. The material is measured by immersing the probe into a liquid or by bringing in contact onto a flat surface of a material in solid or powder forms. The fields at the probe end "infringe" into the material and change as they come into contact with the MUT. The reflected signal (S_{11}) can be measured and related to permittivity.

C. FREE SPACE METHOD

Free-space method uses antennas to focus microwave energy at a slab or sheet of material. This method is non-contacting and can be applied to materials to be tested under high temperatures and hostile environments. The material properties measurement using the free space method is based on the scattering parameter measurement of the MUT. This measurement method is broadband and usually unaffected by air gaps.

Table 2 compares the advantages and disadvantages of each measurement technique in terms of their frequency range, accuracy, sensitivity, sample types and types of parameters which can be collected. It shows that each technique has its own strengths and limitations, which need to be considered when deciding on the type of measurement.

IV. MICROWAVE ABSORBERS

A. NANOCOMPOSITE ABSORBING MATERIAL

Nanocomposite absorbers have received much attention in recent years due to its light weight, thin thickness, high absorption, and wide operating frequency band [19]. To achieve such features, the composite such as carbon-based

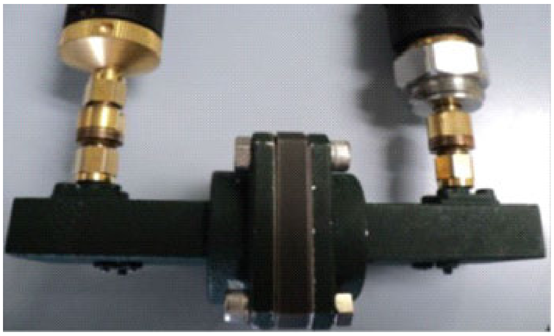


FIGURE 4. Actual waveguide transmission line measurement setup [12].

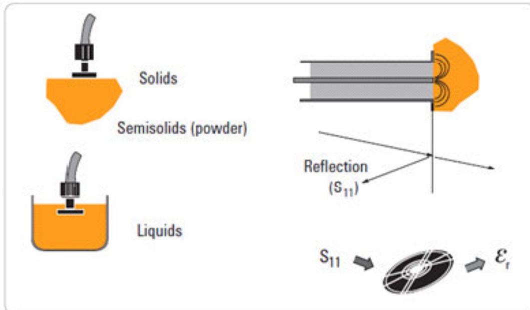


FIGURE 5. Illustration of an open-ended coaxial probe with material under test (MUT) [11].

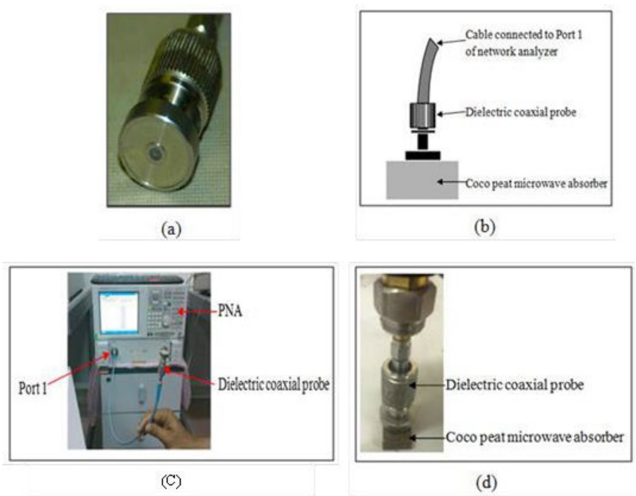


FIGURE 6. (a) Dielectric coaxial probe (b) Sketch of dielectric coaxial probe position and material under test (c) Connection of dielectric coaxial probe connection to port 1 of PNA (d) Actual dielectric properties measurement [13].

composite materials are the popular choice among researchers which offer excellent properties of absorber. They include graphite, graphene and carbon nanotube, as illustrated in Fig. 8. Similar to conventional absorbers, the electrical and magnetic capacities of such nanomaterial absorbers are determined mainly by the relative permittivity, the relative permeability, the electromagnetic impedance match and the microstructure of the absorbing material. The absorbing

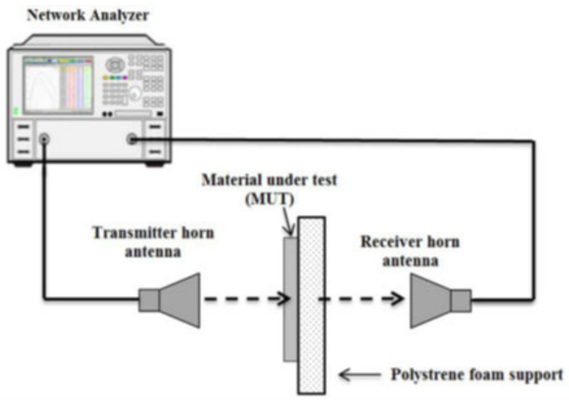


FIGURE 7. Illustration of a free space measurement method setup.

TABLE 2. A comparison of different measurement technique.

Method	Advantages	Disadvantages
Transmission line method	- Sensitive - High accuracy - Both permittivity and permeability	- Narrower range of frequency - Difficult sample preparation - Destructive to sample as the sample need to fit inside a sample holder during measurement
Open ended probe method	- Wide range of frequency - Easy to apply - Non-destructive to sample as the probe only in contact with the surface of sample	- Limited accuracy - Errors in measurement at very low and very high frequency
Free space measurement method	- Accurate - Wide range of frequency - No sample preparation - Both permittivity and permeability	- Errors in measurement due to diffraction effect at the edges of sample

ability of composites is dependent on its geometry, particles distribution, morphology and loading level of the fillers particles [4]. The type of fillers chosen to be incorporated in a composite is a critical parameter to achieve better and higher absorption of electromagnetic wave absorbers. This is mainly due to their intrinsic materials properties in combination with the main absorbing materials which result in an improved electromagnetic absorbing composite. An ideal electromagnetic impedance matching condition is met when a beam of wave propagates through the surface of an absorber and produces zero reflectivity. Thus, adjusting the electromagnetic parameters of materials by combining and balancing the dielectric losses with magnetic losses will improve absorption performance.

Carbon nanotube (CNT) is a promising candidate for composite absorbers due to the existence of carbon nanostructures [4], [14]. Zhou *et al.* [15] presented a lanthanum nitrate doped amorphous carbon nanotubes (ACNTs) (with

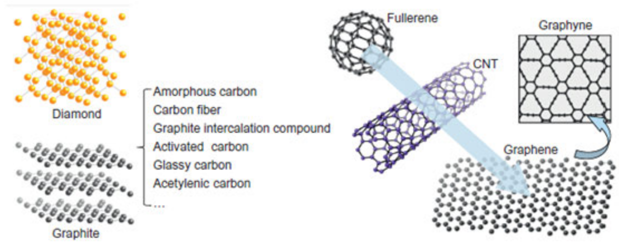


FIGURE 8. Carbon allotropes and related material [14].

diameters in the range of 7-50 *nm*) and polyvinyl chloride (PVC) composite electromagnetic waves absorber. The results exhibited the minimum reflection loss was −25.02 dB at 14.44 GHz with a wide bandwidth of 5.8 GHz. To further enhance the absorption bandwidth, Meng *et al.* [21] proposed new nanocomposites material incorporating magnetic nanoparticles, island-like nickel/carbon nanocomposites electromagnetic wave absorber in a broad frequency range of 4.5 – 18 GHz (reflection loss < -20 dB), leading to an ultra-wide absorption bandwidth of 13.5 GHz. However, such material suffers from the thickness, where it exhibited higher thickness than [15].

Another attractive option for lightweight and wide frequency band is graphene. Graphene is simply a one atomic layer of graphite; one of the three naturally occurring allotropes of carbon. The unique structure of graphene is its sp^2 hybridization and very thin atomic thickness [14]. The thickness of graphene is only 0.35 nm and it is also very light at 0.77 mg/m^2. These enable graphene to have many outstanding properties in terms of optical transparency, electric conductivity, mechanical strength and thermal conductivity [14], [16]. Whereas CNT can be considered as rolled-up graphene in the form of coaxial tubular structure with several micrometers length as shown in Fig. 8 [4], [14], [16]. It has strong mechanical strength and displays a good semi-conducting properties [4], [16]. Incorporating the ferrite nanoparticles to the graphene is also an effective method for lightweight and wide frequency bandwidth. An example of such is the graphene-coated ferrite nanocomposites (Fe/G) proposed in [17]. This resulted in a wide operating bandwidth of more than 4.6 GHz with a thickness of only 1.7 mm. Ding *et al.* [18] improved the fabrication for such absorber by adding Cobalt (Co) into ferrite, achieving broader absorption bandwidth of 7.17 GHz (see Fig. 9). Several similar works presented in [21], [22] and [24] which added metallic nano-oxide nanomaterials, i.e., tin sulfide (SnS_2), zinc sulfide (ZnS) quantum dots and etc also indicated satisfactory broad frequency bandwidths, see Table 3.

Double-layering structures of nanocomposites absorber is another effective technique for broadening the bandwidth and lightweight. Ni *et al.* [19] proposed the use of a double layered barium titanate (BTO)/CNT nanocomposites resulted in an absorption bandwidth of 1.7 GHz with a minimum reflection loss up to −63.7 dB at 13.7 GHz This absorber has a small thickness of only 1.3 mm. However, the

TABLE 3. Reflection loss performance state-of-the-art composite and nanocomposite materials.

Ref.	Materials	Thickness (mm)	Target Frequency (GHz)	Reflection loss RL_{min} (dB) at (f (GHz))	-10 dB Bandwidth (GHz)
[15]	Lanthanum nitrate doped ACNT/PVC nanocomposite	2.0	2.0-18.0	-25.02 (14.44 GHz)	5.8 (12.0-17.8)
[17]	Fe_3O_4/RGO nanocomposites	1.7	2.0-18.0	-65.10 (15.2 GHz)	>4.6 (13.4-18.0)
[18]	CoFe/RGO nanocomposites	2.7	2.0-18.0	-21.64 (13.41 GHz)	7.17 (10.87-18.04)
[19]	Double layer BTO/CNT nanocomposite	1.3	0.5-13.8	-63.7 (13.7 GHz)	1.7 (12.1-13.8)
[8]	Double layer PANI and PANI/Fe_3O_4 composite	1.0	26.5-40.0	-54.00 (33.72 GHz)	11.28 (27.24-38.52)
[20]	Island-like nickel/carbon nanocomposite	2.3-7.0	2.0-18.0	-	13.5 (4.5-18)
[21]	RGO/$CoFe_2O_4$ nanocomposites	2.5	2.0-18.0	-53.6 (11.4 GHz)	4 (10.0-14.0)
[22]	RGO/ $CoFe_2O_4$/ZnS nanocomposite	1.8	2.0-18.0	-43.2 (13.7 GHz)	4.5
[23]	Double layer Ag/CNT nanocomposite	3.3	0.5-14.0	-52.9 (6.3 GHz)	3 regions; 3.5 (4.7-8.2) 0.8 (9.0-9.8) and 1.5 (12.5-14.0)
[24]	RGO/ $CoFe_2O_4$/SnS_2 nanocomposite	1.6	2.0-18.0	-54.4 (16.5 GHz)	4.9 (12.4-17.3)

frequency range covered in this study can only operate in a narrower band than the single layered nanocomposite materials [15], [18]–[22] and [24]. This caused the final design to only operate from 12.1 to 13.8 GHz. Similar work was conducted by Melvin *et al.* [23], who proposed CNT composites incorporating silver nanoparticles (Ag) and evaluated its microwave absorbing properties (see Fig. 10). The use of Ag nanocomposites showed enhanced absorbing capability of electromagnetic wave where it managed to achieve multi frequency regions, up to 3 regions of wide bandwidths with a maximum up to 3.5 GHz.

On the other hand, Xu *et al.* further improved the −10 dB bandwidth by proposing a double layered polyaniline (PANI) and polyaniline/magnetite (PANI/Fe_3O_4) that produced an excellent 11.28 GHz bandwidth with a minimum reflection loss reaching −54 dB in the millimeter wave range centered at 33.72 GHz. This method enabled the absorber to have very low profile and very thin 1 mm thickness while featuring an ultra-wide bandwidth [8]. Table 3 summarizes the performance of current state-of-the-art types of composite and nanocomposite materials as absorbers. In general, these composites and nanocomposite-based absorbers indicated satisfactory absorption performance and their suitability as wave absorbing materials. Fig. 9 and 10 show examples of SEM and TEM images of nanocomposites.

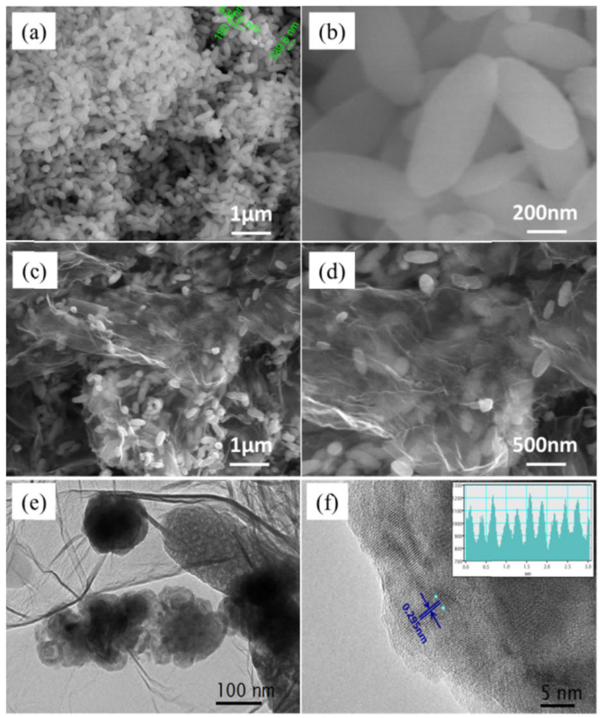


FIGURE 9. Typical SEM images of $Co_xFe_{2-x}O_3$ (a,b) and CoFe@rGO (c,d) and typical TEM images of CoFe@rGO (e,f) [18].

Another recent trend on very thin green material incorporating nanoparticles can be observed in [25]–[28]. These materials exhibited comparable absorption bandwidth with thicker non-green materials. Firstly, various types of carbon gels were investigated by Gutierrez et al. [25]. The broadband electromagnetic properties of these gels and their dependence on bulk density and pore size were studied across a wide frequency range (from 20 Hz to 36 GHz). Maximum microwave absorption in this work was observed at lower densities (0.2 g/cm^3 and 0.4 g/cm^3). At 30 GHz, however, layers of these carbon gels with thicknesses of 2 mm and 4 mm, and with low density of 0.2 g/cm^3 showed absorption of around 0.5, lower than the ideal absorption rate value of

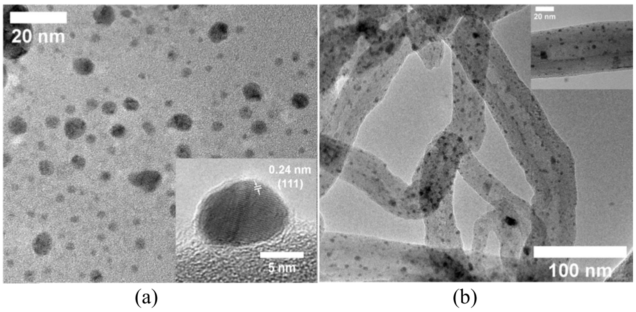


FIGURE 10. TEM images of (a) Ag nanoparticles and (b) Ag/CNT nanoparticles [23].

0.9. It is also observed that carbon gels with higher thickness (4 mm) showed better absorption compared the lower thickness (2 mm).

Besides that, the application of low-cost precursors such as biomass or waste residue in carbon nanomaterials have indicated promising potentials. Both are good candidates as they are enriched sources of carbon and easily available as well as renewable. For example, Zhao et al. [26] introduced a novel carbon aerogel coated co-composite fabricated from biomass based alginate aerogel as precursor. The Co/carbon aerogel composite showed nano-porous morphology with yolk-shell structure. Good microwave absorption ability was exhibited with coating thicknesses of between 1.5 and 5 mm. For a thickness of 1.7 mm, its minimum RL was −34 dB at 15.6 GHz, with an absorption bandwidth of 4.6 GHz. It was also demonstrated that the minimum RL frequency shifted towards lower frequencies with increasing coating thicknesses.

Next, Xu et al. [27] introduced a porous wood aerogel (WA) modified with Fe_3O_4/ZIF-67 nanoparticles for the microwave absorption. The natural delignified WA was used as the porous low-density compressible matrix, and Fe_3O_4/ZIF-67 dodecahedrons as the absorbing agents. By adjusting the amount of Fe_3O_4 in the composites, its magnetic characteristic can be controlled, hence a better microwave absorption can be achieved. The fabricated composites with 2.0 mmol Fe_3O_4 exhibited the best microwave absorption performance with minimum RL of − 23.4 dB with a thickness of 1.5 mm, and absorption bandwidth of 4.5 GHz, similar to [26].

Apart from that, porous jute biomass carbon (PJBC) composited by Fe_3O_4 nanoparticles were successfully prepared using chemical coprecipitation method at 60°C by Wang et al. [28]. They used jute as the carbon source because it is low cost and easy to make porous carbon matrix. The prepared PJBC/ Fe_3O_4 composites indicated excellent microwave absorption performance in comparison to Fe_3O_4 magnetic nanoparticles due to their porous structure and large interfaces between PJBC and Fe_3O_4. The minimum RL value of −35.7 dB with absorption bandwidth of 5 GHz is achieved with a thickness of 1.6 mm. In conclusion, the aforementioned green-based composite absorbers infused with nanocomposite demonstrated great potential towards realizing ultra-wideband absorption characteristics while featuring very small thicknesses.

B. METAMATERIAL AND METASURFACE-BASED ABSORBERS

Metamaterials based absorbers is another emerging type of new absorbing materials. They are arrays of structured or periodically arranged materials which exhibit exceptional physical properties [29]–[31]. Metamaterials feature extraordinary properties such as evanescent wave amplification and negative-refractive index which not readily observed in natural materials [29]–[33]. The properties of metamaterials enable them to potentially be applied in many types of applications including perfect absorbers [29], [30], [34]–[36], sensors [31], [36], photovoltaic cells [32], electromagnetic filters and others [31].

In contrast to conventional absorbing materials, metamaterials their absorbing properties originate from their structure rather than the material of which they are composed of. They are often engineered by arranging a set of small scatterers such as metallic rings and rods, or spherical magneto-dielectric particles in a regular array throughout a region of space. For instance, split rings may be formed into one unit cell, or may consist of several sub-units, and then arrayed to fill space in one, two and three dimensions. Conductive metals such as copper, gold, or silver may be used as their components [31]. On the other hand, three-dimensional metamaterials can be extended by arranging electrically small scatterers or holes into two-dimensional pattern at the surface or interface. This surface version of a metamaterial, also known as metasurface can be used as an alternative. They require less physical space compared to a full three-dimensional metamaterial structures, offering the possibility of less lossy structures. Metasurfaces have a wide range of potential applications in electromagnetics including novel wave-guiding structures, absorbers, biomedical devices, terahertz switches and etc.

Besides being more flexible in terms of design, simpler manufacturing procedures, low profile, ultra-thin thickness, near unity absorption properties, metamaterial or metasurface-based absorbers also feature tunability and ease of absorption characteristics control compared to conventional absorbers. However, a critical limitation of the current metamaterial and metasurface absorbers they suffer from narrow absorption bandwidth due to their resonant absorbing mechanisms [37]–[39].

Different types of metamaterial or metasurface structure have been studied in the past years. Fig. 11 illustrates the design of a unit cell of one of the first metamaterial based absorber by Landy et al [29]. Each unit cell consists of two metallic layers and a dielectric spacer, which resulted in a maximum narrow band absorbance of 88% at 11.5 GHz. The design of such absorber was fabricated on a rigid FR4 substrate with a very small thickness of 0.2 mm and 17 μm copper thickness. Another example is a simple design of inclined metallic hexagonal patch printed on grounded dielectric

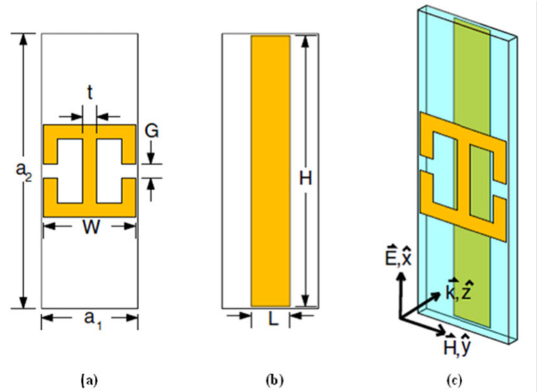


FIGURE 11. Design of metamaterial absorber, (a) Electric resonator and (b) cut wire component of unit cell, (c) a unit cell with direction of wave propagation [29].

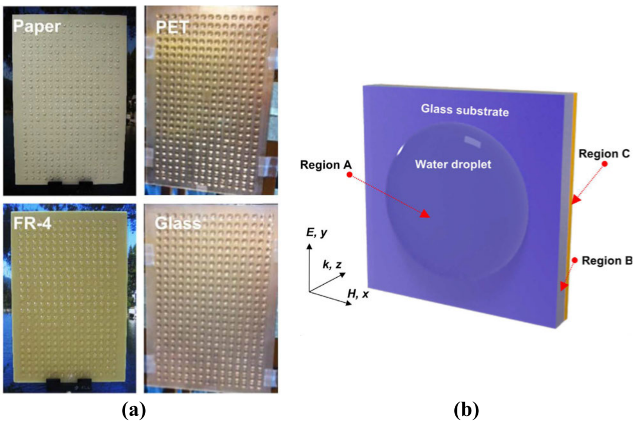


FIGURE 12. Design of metamaterial absorber, (a) Images of water droplets are formed periodically on the surface of 4 different substrates (b) the structure of metamaterial with water droplet (Region A) on the glass substrate (Region B) backed by metallic ground plate (Region C) [41].

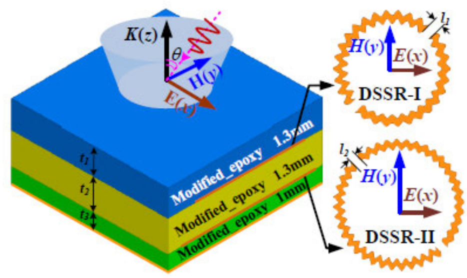


FIGURE 13. Schematic geometry of a unit cell of three layers metamaterial microwave absorber based on DSSRs [44].

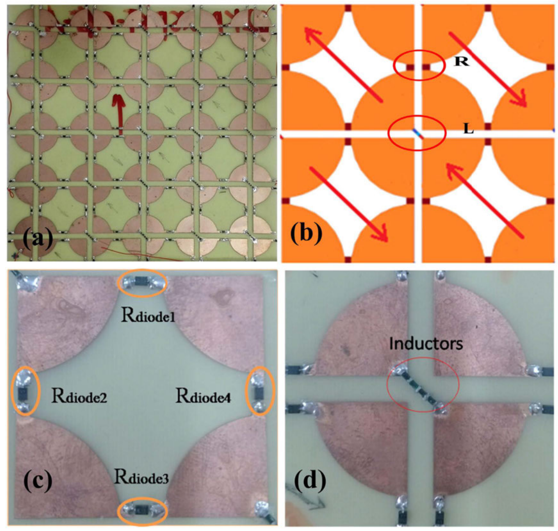


FIGURE 14. Images of (a) sample of tunable and polarization insensitive absorber (b) diagram of current flow (c,d) magnified images of the pattern [45].

substrate was presented by Sood et al [40]. The metamaterial absorber has been fabricated on a 1.6 mm FR-4 substrate with 36 x 36 unit cells. This design was able to achieve a 10 dB absorption bandwidth of 4.91 GHz from 8.96 to 13.87 GHz. Besides that, Yoo et al introduced a new type of water droplet-based perfect metamaterial absorber. Water droplets unit cells were arranged on the surface of various substrate such as FR-4, PET, paper and glass by controlling the surface wettability as shown in Fig. 12 [41]. Metamaterial absorber with paper as substrate presented the best performance with 7.3 GHz bandwidth followed by FR-4, PET and glass. However, the water droplets can be influenced by the strong vibration, extreme temperature and dirty environment.

To increase the absorption bandwidth, several methods have been proposed, including stacked multilayered structure [42]–[45]. Multilayered structure is formed by stacking two or more layers to enhance the bandwidth where normally different layers features their own resonant frequency [46]. In the earlier work, Xiong et al presented a metamaterial absorber consists of a periodic array of loop-dielectric multilayer structure with thickness of 3.65 mm [43]. The numerical simulations indicated a wide absorption bandwidth of 12.63 GHz. This is a significant improvement in comparison to the work by Landy, at the expense of increased thickness to 3.45 mm. Similarly, Li et al designed a three-layered metamaterial-inspired absorber based on the double split separation rings (DSSRs) structure. The result shows a wide operating bandwidth of 9.3 GHz with a structure thickness of 3.6 mm [44].

Another recent technique in performance enhancement is by loading the absorbers using lumped elements [38], [47]–[49]. Tunable or adaptive MMAs are proposed to enable microwave absorbers to work in a wide frequency range. Li et al proposed a tunable MMA with a near-perfect absorption peak, shifting within a frequency range of 0.2 to 7.6 GHz by modulating external magnetic field [35]. The metamaterial absorber consists of a metal-backed Garnet-type ferrite with a metal-strip-arrayed metastructure on the surface, where the ferrite acts as the tuning medium. Alternatively, Wang et al presented a tunable and polarization insensitive absorber based on an array of PIN diodes with biasing lines including inductors. The absorber performed with satisfactory absorption levels from 1.6 to 4.5 GHz, and from 5.4 to 8 GHz with total thickness of 11.9 mm [45].

Generally, multilayered structures are observed to be capable of widening the bandwidth range of absorption, but at the same time, increases the design complexity compared with single-layered structures. This is due to the need for each layer in the multilayered structure need to be precisely stacked (or precision in soldering, if applicable) to produce the intended performance during fabrication [50]. Besides that, increasing the number of layers also results in increased thicknesses of the absorbers. Meanwhile, loading lumped elements can be effective in enabling multi-band or tunable features in the operation of absorbers [47], [51]. Despite that, such absorbers typically result in a narrower absorption bandwidth.

In the recent years, flexible or stretchable metamaterials absorbers are gaining attention among researchers [52]–[54]. Unit cells for most metamaterial absorbers are typically metallic patches formed on rigid substrates, and such structures provide durable mechanical support. While being acceptable in most applications, these structures difficult to be used in portable or wearable applications. This brings about the need to design them on flexible substrates/materials. One of the main challenges of flexible or stretchable metamaterial microwave absorbers is to maintain their absorption performance when operating in the states of deformation such as when bent, stretched and etc. It is of prime importance that the influence of such deformation on the performance of the absorber be characterized to understand its variation of absorption, and preferably, to minimize it. For example, a wearable metamaterial microwave absorber using felt as substrate for indoor radar clear applications was presented in [52]. The fabricated absorber exhibits two absorptivity peaks of more than 90% at 9 GHz and 9.8 GHz. Besides that, the absorber was insensitive of the polarization angle and the frequency shift for different absorber bending radii was also minimal. In addition to that, Zhou et al. proposed a stretchable slotted cross shaped microwave absorber with deformation compensation for resonant frequency [53]. The unit cell schematic for both conventional and compensation absorbers are shown in Fig, 15.

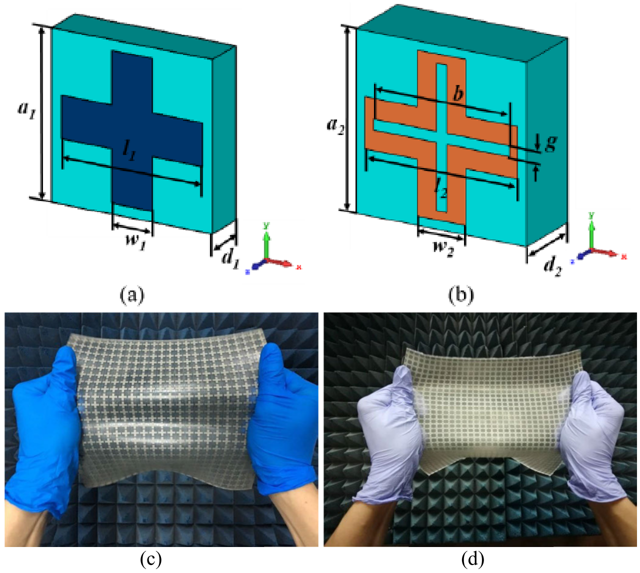


FIGURE 15. The schematic of a unit cell of stretchable microwave absorber for (a) conventional structure and (b) compensation structure with photos of fabricated absorbers in stretch; (c) the conventional structure and (d) the compensation structure [53].

At 10% of stretching, the absorption frequency of the proposed compensation absorber only shifted by 1.1% compared to 3.2 % shift in the conventional absorber for TM polarization. For TE polarization, this shift level is 3.2 % for the compensated version, and 5.2 % for the conventional one. Table 4 presents a summary of previous literature on metamaterials and metasurface-based absorbers.

C. GREEN MATERIAL-BASED COMPOSITES ABSORBERS

Due to the increasing need to ensure sustainability, absorbing materials are increasingly adapting green or organic materials. They include materials such as agricultural waste as an alternative to ensure environmental-friendliness, with the potential of overcoming the limitations of traditional electromagnetic absorbers. Moreover, agricultural waste is abundantly available and possesses good microwave absorption properties. EM absorbers based on agricultural waste includes the likes of rice husk [57], [58], sugar cane bagasse [59], dried banana leaves [60] and etc, have exhibited promising absorption rates.

The first example is an 8 × 8 array pyramidal microwave absorber using rice husk is presented in [57]. The absorber was designed using Computer Simulation Technology Microwave Studio (CST MWS) and the radar cross section method was used to measure the reflection loss performance of the fabricated absorber from 7 to 13 GHz. The results obtained were in the range of −28 dB to −58 dB. To improve the design of the pyramidal microwave absorbers, Malek et al. [61] combined rice husk and rubber tire dust. Different percentages of rubber tire dust and rice husk were investigated and the result showed that the highest percentage of rubber tire dust had the best reflection loss. In addition to that, the performance of rice husk and carbon nanotubes were used to design multilayered flat microwave absorbers in [58]. Such structure resulted in a microwave absorption (or reflection loss) of more than −20 dB. Besides that, a set of pyramidal microwave absorbers using sugar cane bagasse has been fabricated and measured in [59]. The average reflection loss of the fabricated absorber is −45.9 dB between 1.8 and 18.0 GHz. The high amount of carbon contents, particularly in sugar cane bagasse increases its absorption level.

Table 5 compares the reflection losses for different types of microwave absorbers made using agricultural waste materials. Despite its many advantages, as of now, these absorbers are typically bulky, heavy, and are limited in terms of durability and mechanical strength. This is due to the limited interfacial interaction between the agriculture waste and the resin or carbon-based filler materials such as CNTs, carbon black (CB) and carbon nanofibers (CNFs). This resulted in

TABLE 4. Summary of metamaterial- and metasurface-based absorbers.

Ref.	Characteristics	Number of layer	Thickness (mm)	Target Frequency (GHz)	-10 dB Bandwidth (GHz)
[40]	Composed of inclined metallic hexagonal patch printed on a grounded dielectric substrate	1	1.6	4-18	4.91 (8.96-13.87)
[41]	Arranged water droplets on the surface of different substrates (FR-4, PET, Paper, Glass)	1	4	6-18	FR-4: 3.77 (8.3-12.07) PET: 1.13 (11.23-12.36) Paper: 7.3 (9.2 -16.5) Glass: 0.6 (12.05-12.65)
[42]	Consists of two layers of dielectric substrates. The sub-cell of a unit cell with two concentric Closed Ring Resonator (CRR) structure is designed on the top surface of both layers of the dielectric.	2	3.2	4-12	Dual band; 1.48 and 1.86
[43]	Composed of periodic array of loop-dielectric multilayer structure with three metallic films and a metal ground to reflect EM wave. Every two adjacent metal sheets (copper) are separated by dielectric substrate (TMM4).	3	3.65	0-25	12.63 (8.37-21)
[44]	Based on the double-split-separation-rings (DSSRs) structure	3	3.6	2-18	9.3 (5.4-14.7)
[45]	An array of PIN diodes with biasing lines including inductors in 4-axial symmetric form	2	11.9	1-8	2.7 (1.6-4.3) and 2.6 (5.4-8)
[47]	Consists of an outer circular ring and an inner Jerusalem cross with two pin diodes mounted in between, etched on FR-4.	1	1	0-7	< 0.5
[48]	Consist of a conductor-backed magnetic sheet (CMBS) coated by a non-Foster metasurface (NFMS).	1	7.35	50MHz-1200MHz	< 0.5
[49]	Eight-resistive-arm (ERA) cell as metamaterial unit cell on a FR-4 substrate.	1	3	7-15	5 (8-13)
[38]	Consist of metallic square patch and square loop structure inserted with resistors etched on Rogers 5880 substrate with a foam spacer.	1	~6.6	2-19	Dual-band; 0.8 (2.7-3.5) and 7.5 (10.5-18)
[35]	Metal-backed Garnet-type ferrite with a metal-strip-arrayed metastructure on the surface.	1	1.2	0-12	Tunable in frequency range of 0.2 - 7.6 GHz
[55]	Comprises conductive crossed dipole loaded with split ring resonators	1	1.83	2-16	Multiband at: 3.68, 8.58, 10.17 and 14.93
[37]	Periodic arrays of honeycomb core as the constituent dielectric, onto which resistive paint has been deposited in square loop patterns	1	5.5	2-18	11.4 (5.52-16.92)
[56]	High impedance surface metamaterial absorber based on octagonal ring-shaped resistive patches	1	8	2-18	10.28 (3.65-13.93)
[39]	Hybrid unit cells with each comprising four circular sectors on FR-4 substrate	1	0.8	9.5-11.5	0.45 (10.15-10.6)

their porous structure which may easily degreade in terms of structural integrity when applied in a real environment. These issues are hindering their employment as a credible alternative to commercial microwave absorbers. Fig. 16 displays a couple of fabricated electromagnetic absorbers from agricultural waste.

V. NANOCELLULOSE-BASED COMPOSITES

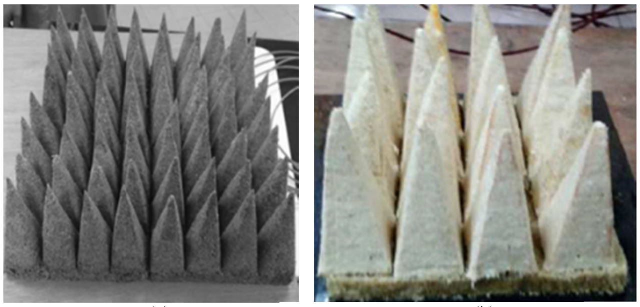
A. NANOCELLULOSE COMPOSITES: RECENT APPLICATIONS

Cellulose, an organic compound easily obtained from nature, is a structural component of the cell walls of many plants. It is used mainly for making paper and cardboards, and have recently been used in various industrial applications [63]. This is due to its availability, sustainability and flexibility [63]–[65]. In the past years, cellulose has received special attention due to their application in the development of conducting materials. For example, an earlier work in [66] featured the technique to produce graphite/carbon fiber/cellulose fiber composites papers with tunable conductivity and good mechanical properties. The produced conductive papers are flexible and extremely conductive. Another work implementing a similar concept is presented in [67]. In this work, Carbon nanotube (CNT) / cellulose composites material was fabricated using a similar papermaking process. A higher electric conductivity was obtained compared to the one usually obtained using polymer-based composite material without affecting the paper strength.

Cellulosic fibers in the micro- and nano-scales have also been proven to potentially serve as an environmental-friendly composite reinforcement. The production of nanocellulose (NC) with crystalline structure is actively pursued due to its improved material properties [68]. A few examples of biodegradable polymer–cellulose nanofiber (CNF) based nanocomposites and their mechanical properties are presented in [64]. This includes Poly(vinyl alcohol) – CNF, Poly(lactic acid) – CNF, Poly(ethylene oxide) – CNF, Chitosan – CNF, Starch – CNF and Soy protein – CNF. It has been discovered that NCs can be applied to energy

TABLE 5. Reflection loss performance of different green material for absorber.

Ref.	Material	Shape / Thickness (cm)	Target frequency (GHz)	Reflection loss (dB)
[58]	Rice husk and CNT	Multilayered flat absorber/1.8	2 – 18	Better than -20dB
[57]	Rice husks	Pyramidal absorber/15	7 – 13	Better than -20dB
[59]	Sugar cane bagasse	Pyramidal absorber/ 15	0.1 – 20	Better than -30dB
[13]	Coco peat	N/A	8 – 12.4	Better than -30dB
[62]	Rubber wood sawdust	Flat absorber/ 2.4 and 4.8	1-3	Best at -16.4dB
[60]	Dried banana leaves and coal	Pyramidal absorber/13	8.2-12.4	Average -45.2dB



(a) (b)

FIGURE 16. Electromagnetic absorbers made from agricultural waste, (a) rice husk [57] and (b) sugar cane baggase [59].

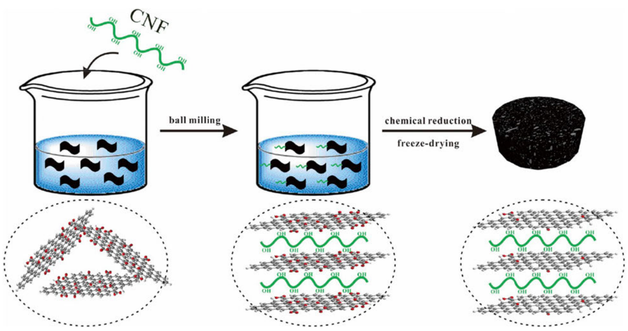


FIGURE 17. The schematic illustration for self-assembly process of CNF/rGO composites [69].

devices such as paper batteries, supercapacitors and paper displays.

Recently, their application in microwave absorption have also been investigated [69]. Kuang et al. developed a natural biopolymer cellulose nanofiber (CNF) and reduced graphene oxide (rGO) composites as microwave absorptive materials. In this work, different compositions of CNF/rGO were fabricated to evaluate the absorption performance. The microwave absorption performance with maximum reflection loss of −40.64 dB and absorption bandwidth of up to 7.72 GHz was achieved with a thickness of 2.5 mm, see Fig. 17 and 18. Another recent study, Xu et al. reported a lightweight and flexible periodic cellular CNF/CNT foam [70]. The microwave absorption bandwidth is reported to be 15.7 GHz, from 2.3 to 18 GHz. However, a thickness of 20 mm is needed to interconnect the porous structure and to enable the strong hydrogen bonding among CNF-CNF and CNF-CNTs, as illustrated in Fig. 19.

These results indicate that nanocellulose features promising absorbing characteristic, besides being a potentially

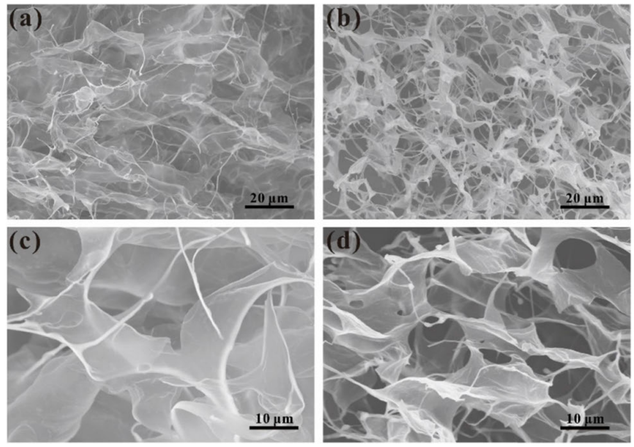


FIGURE 18. SEM images of CNF (a) (c) and CNF/rGO aerogels (10rGO) (b) (d) [69].

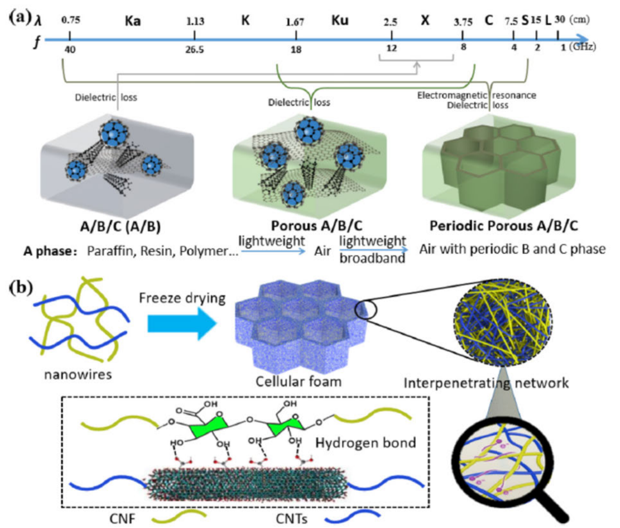


FIGURE 19. (a) Schematic illustration of the microwave absorption materials for broad-band microwave absorption (b) Schematic illustration of the preparation of a periodic CNF/CNT foam [70].

suitable as a thin and flexible electromagnetic absorbing material in near future. Although cellulose can be used to build highly conducting materials as electromagnetic shielding materials, is possible that its high carbon content also increases its ability to be used as electromagnetic absorbers. Moreover, different types of filler could be incorporated into nanocellulose composites to offer better electromagnetic absorption. These includes CNTs, CB and other carbon-based, as well as magnetic-based materials [3], [4]. This is

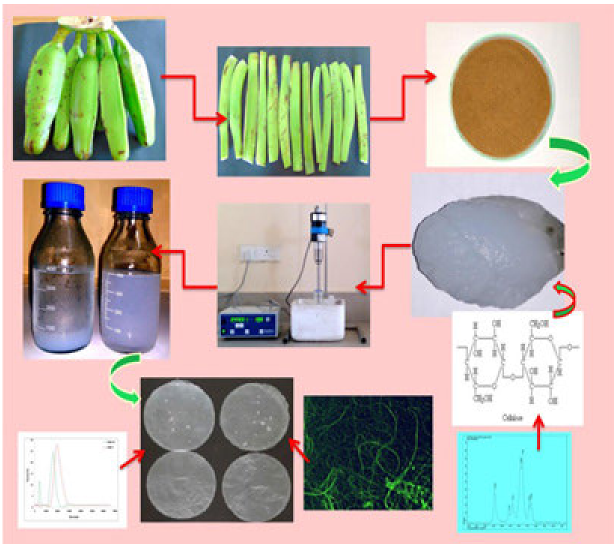


FIGURE 20. Steps in development of cellulose nanopaper from banana peels [71].

because the type and concentration of these fillers loaded in a composite affects the absorption performance, as proven in [69]. However, currently its potential ability as electromagnetic absorbers is not widely studied.

B. METHODS OF PRODUCTION

There are two distinctive methods to produce nanocellulose; by means of mechanical and chemical methods [63], [64]. Chemical pre-treatment includes alkali and acid hydrolysis [7], [71], [72], [74]–[77]. Alkali solutions such as sodium hydroxide, potassium hydroxide or caustic soda facilitate the removal of lignin, hemicellulose and pectin. Kraft process (or sulphate process) is the most commonly used method of lignin removal, which uses hot sodium bydroxide and sodium sulfide solutions in a digester. Meanwhile acid solutions such as sodium chlorite, hydrochloric acid or oxalic acid are able to leach out non-cellulosic components. On the other hand, the mechanical method includes the delamination of interfibrillar hydrogen bonding of cellulose microfibers under intense mechanical forces. For instance, fibers are mechanically separated into nanofibers using high intensity ultrasonication [72], [73], [76], [78]. The ultrasonic treatment is performed in an ice bath and ice is sustained throughout the sonication period. The nanofibers obtained after the ultrasonic treatment will be vacuum-filtered and vacuum-dried to produce a fine sheet of cellulose nanopaper as shown in Fig. 20.

Another example is shown in Fig. 21 where nanocellulose fibers from pinecones using chemical and mechanical treatment were produced [74]. The chemical pre-treatment and mechanical grinding processes were optimized with respect to the tensile properties of films prepared using the cellulose fibers. This study showed that chemical pre-treatment followed by mechanical process is a simple and efficient technique for the production of nanocellulose. In general,

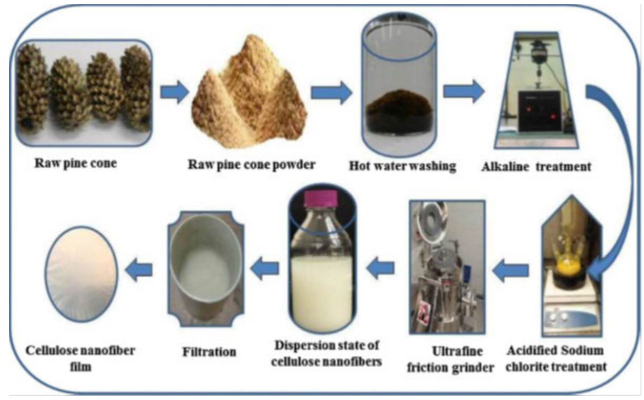


FIGURE 21. Schematic view of production of nanocellulose from pinecone biomass using chemical and mechanical treatments [74].

it is observed that chemical methods can break cellulose fiber nanocrystals or modify the cellulose surface properties [63]. On the other hand, mechanical methods are unsuitable for precise material processes, but are eco-friendly and chemical free. These factors need to be considered when deciding on the choice of methods in the process of producing nanocellulose for the desired output.

VI. CONCLUSION

In this review, different types of electromagnetic absorbing materials have been studied and developed over the years with new features and improved performance. They should ideally be lightweight, low profile, enable ease of fabrication, operate in wider bandwidth and preferably be mechanically flexible. These features are highly desirable in emerging applications of electromagnetic absorbing materials. In achieving this aim, the selection of material type and their structure are critical factors in ensuring functional future absorbers. Moreover, the choice of green materials based on natural fibers is also significant to contribute towards sustainability efforts.

Despite featuring acceptable levels of absorption, current green absorbers from agricultural waste are physically bulky, thick, rigid and heavy. To overcome this, such green materials need to be produced and applied in a different form. One of the best methods of doing this is by extracting the nanofiber or nanocellulose from the agricultural waste, which is highly possible due to their origin as part of natural plants. From there, a thin, flexible and strong mechanical properties material could be achieved. Moreover, the application of nanocellulose to develop nanocomposite-based metamaterial may potentially also serve as good reinforcements with effective microwave absorption characteristics in building materials, for instance. They feature very attractive characteristics as future microwave absorbers, i.e., ultrathin, lightweight, ultra-wideband and flexible, while maintaining a minimal variation of absorption performance when implemented in bent conditions.

While nanocellulose composites are of great potential, there are still some limitations which need to be resolved.

One of them is in controlling the sizes and properties of nanocellulose composites produced, which can be easily affected during the production process. Specifically, excessive chemical and mechanical treatments during its production could alter or change the properties of the nanocellulose produced. This potentially opens up the possibility of seamless integration of both components in smart buildings and cities.

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