



1 Article

2 EMI Shielding of Carbon Nanoparticles/Polymer 3 Composites at Terahertz Frequency Region

4 Eman Taha^{1*}, Asmaa El-Deeb², Usama Kandil¹, Mahmoud Reda Taha³

5 ¹ Egyptian Petroleum Research Institute, Nasr City 11727, Cairo, Egypt; eman@unm.edu

6 ² Housing and Building National Research Center, Giza 12622, Egypt; asmaphysics@yahoo.com

7 ¹ Egyptian Petroleum Research Institute, Nasr City 11727, Cairo, Egypt; alfa_olefins@yahoo.com

8 ³ Department of Civil, Construction and Environmental Engineering, University of New Mexico,
9 Albuquerque, NM 87131, USA ; mrtaha@unm.edu;

10 * Correspondence: eman@unm.edu; Tel.: +20-12-2573-7515; Fax: +202-22747433

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13 **Abstract:** Significant progress has taken place in the past few years in developing terahertz devices
14 to make use of their superior capabilities in radio astronomy, security screening, chemical analysis,
15 medical imaging and biological sensing, etc. With such advances in terahertz devices,
16 electromagnetic interference shielding (EMI) in the terahertz frequency region is becoming
17 significantly important. In this work, we examine the time-domain spectroscopy, THz-TDS,
18 performance of carbon nanoparticles/epoxy nanocomposites. Two types of carbon nanoparticles
19 (carbon nanofibers and multi-walled carbon nanotubes) with different carbon contents (0.5, 1.0, 2.0,
20 2.5 wt. %) were used to fabricate epoxy nanocomposites using both sonication and mechanical
21 stirring processes and were experimentally investigated at the terahertz (0.1-1.0 THz) frequency
22 range. Shielding efficiency, together with the dielectric properties of both types of carbon-epoxy
23 nanocomposites were studied and compared with that of neat epoxy as a reference. We demonstrate
24 the potential use of carbon nanofibers and multi-walled carbon nanotubes for THz-EMI applications
25 and show that the variation of carbon contents in epoxy matrix can greatly affect EMI shielding
26 efficiency.

27 **Keywords:** Terahertz; electromagnetic interference shielding; polymer nanocomposites

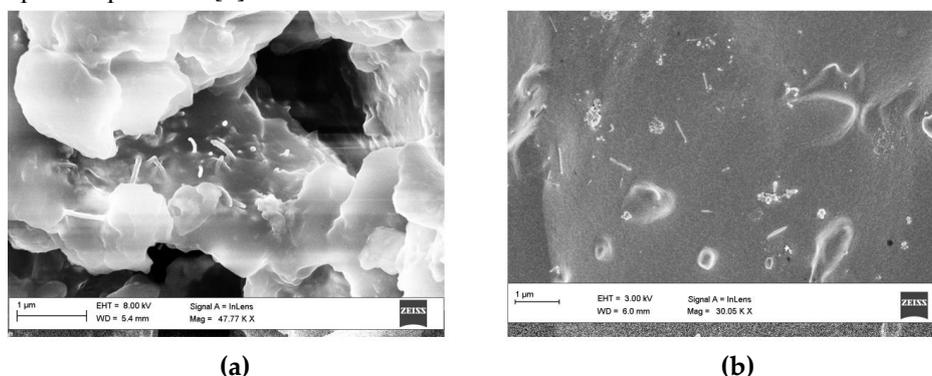
29 1. Introduction

30 Electromagnetic behavior of polymer composites based conductive nanoparticles have attracted
31 a lot of attention in the past two decades [1-2] owing to their properties that can be adjusted by
32 changing the content of nanoparticles content such as carbon nanotube, carbon nanofiber and
33 graphene. Unique properties of such nanoparticles, including electrical conductivity, high strength,
34 durability and low permeability, help in developing the field of composites nanotechnology [3].
35 Electromagnetic interference shielding (EMI) of polymer nanocomposites has
36 been intensively investigated at frequencies ranging from MHz to several GHz [4], but there are very
37 limited studies on EMI of polymer nanocomposites in the terahertz frequency range despite
38 the recent revolution in terahertz devices. For instance, Polley et al. [5] prepared poly-vinyl alcohol
39 (PVA) composites based single-walled carbon nanotubes (SWNT) and EMI shielding efficiency (SE)
40 was found to be 20 dB at 1.25 THz for composite with 1.6% SWNT and increases to 29 dB at 2.1 THz.
41 Seo et al. [6] studied the EMI properties of transparent SWNT-coated polyethylene terephthalate films
42 and SE was improved with increase coating thickness, SE for 300-time coating process was around 8
43 dB. Seo et al. [7] studied also the EMI properties of poly methylmethacrylate (PMMA)/graphite
44 composites and the highest SE was found to be 50 dB for 35.7 wt. % graphite content at 0.5 THz.

45 In this study, epoxy nanocomposites incorporated carbon nanotube and carbon nanofiber were
 46 fabricated and their EMI properties were investigated in terahertz (0.1-1.0 THz) frequency range. We
 47 aimed to improve the EMI shielding efficiency and dielectric properties of epoxy nanocomposites for
 48 using in THz-EMI applications.

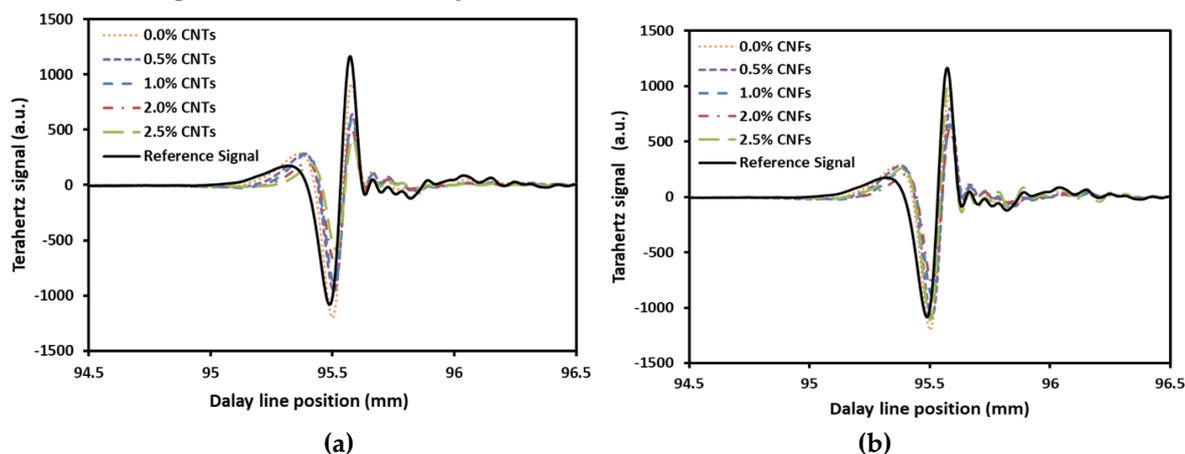
49 2. Results and Discussion

50 To examine the effectiveness of using both sonication and mechanical stirring processes in the
 51 dispersion of carbon nanotubes (CNTs) and carbon nanofibers (CNFs) in the epoxy matrix, Field
 52 Emission Scanning Electron Microscope, FESEM, images were taken for 2.0 wt.% CNTs/epoxy and
 53 2.0 wt.% CNFs/epoxy nanocomposites and are shown in Figure 1(a) and (b), respectively. The images
 54 demonstrate that both CNTs and CNFs were randomly and well dispersed in the epoxy matrix. It is
 55 well known that aggregation of carbonaceous particles during the fabrication process significantly
 56 affects the final properties of the polymer composite. Homogenous dispersion is crucial for formation
 57 of conductive networks through the matrix which govern the electrical properties. Therefore, the
 58 interaction of the electromagnetic wave with aggregated carbonaceous particles is weaker than with
 59 the well dispersed particles [8].



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62 **Figure 1.** Field Emission Scanning Electron Microscope (FESEM) images for (a) 2.0 wt. % CNTs/epoxy and (b)
63 2.0 wt. % CNFs/epoxy nanocomposites.

64 THz time domain signals passing through the CNTs/epoxy and CNFs/epoxy nanocomposites
 65 are shown in Figures 2(a) and (b), respectively. Figure 2 was recorded with and without the
 66 specimens by using the ATR unit with silicon substrate crystal which used as a reference signal. From
 67 Figures 2(a) and (b), it can be observed that the THz amplitude decreases with an increase in the
 68 concentrations of both CNTs and CNFs. The transmission spectra for CNTs/epoxy and CNFs/epoxy
 69 nanocomposites are obtained by applying a fast Fourier transform (FFT) and are then normalized by
 70 the reference spectrum as shown in Figures 3(a) and 4(a).



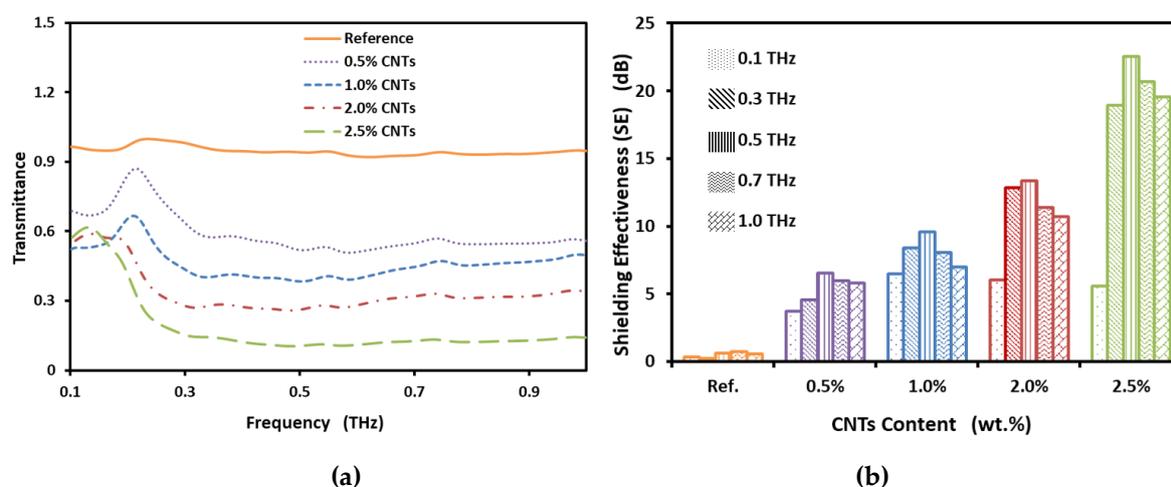
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73 **Figure 2.** Response of the THz electric-field waveforms attenuated through the silicon substrate crystal (black
74 line) and the silicon crystal with (a) CNTs/epoxy and (b) CNFs/epoxy nanocomposites.

75 Considering neat epoxy specimens with 0.0 wt. % carbon as a reference, one can understand the
 76 effect of different carbon contents on electromagnetic shielding. This technique is used here to
 77 demonstrate the significance of CNTs. Transmittance profile of CNTs/epoxy nanocomposites is
 78 shown in Figure 3(a). The transmittance for all the specimens decreases with increasing frequency
 79 from 0.1 THz to 1.0 THz due to high absorption of CNTs at high frequencies [5]. It is evident that
 80 CNTs/epoxy nanocomposites has better transmittance at lower frequencies, which indicate that the
 81 CNTs/epoxy nanocomposites, specifically at high CNTs content, are excellent low-bandpass filters
 82 that screen high THz wave reasonably well.

83 The THz shielding effectiveness (SE) of the nanocomposites is define as a function of the
 84 transmittance (T) using the following equation [9]:

$$SE = -10 \text{ Log } (T) \quad (1)$$

85 Figure 3(b) shows THz SE data for CNTs/epoxy nanocomposites at five different frequencies
 86 (0.1, 0.3, 0.5, 0.7 and 1.0 THz). It is evident that the shielding effectiveness (SE) increases with
 87 increasing frequency and with increasing CNTs content. The highest values of SE were found for
 88 nanocomposite with 2.5 wt. % CNTs which are 18.9 dB, 22.5 dB, 20.7 dB and 19.6 dB at frequencies
 89 0.3, 0.5, 0.7 and 1.0 THz, respectively. The target SE value needed for commercial EMI applications
 90 is around 20 dB, which approximately equal to or less than 1% transmission of the electromagnetic
 91 waves [10]. Further research is warranted to examine EMI shielding of CNTs/epoxy nanocomposite
 92 with CNTs content in the range of 2.5 wt. % and examine their practical use for THz-EMI applications.



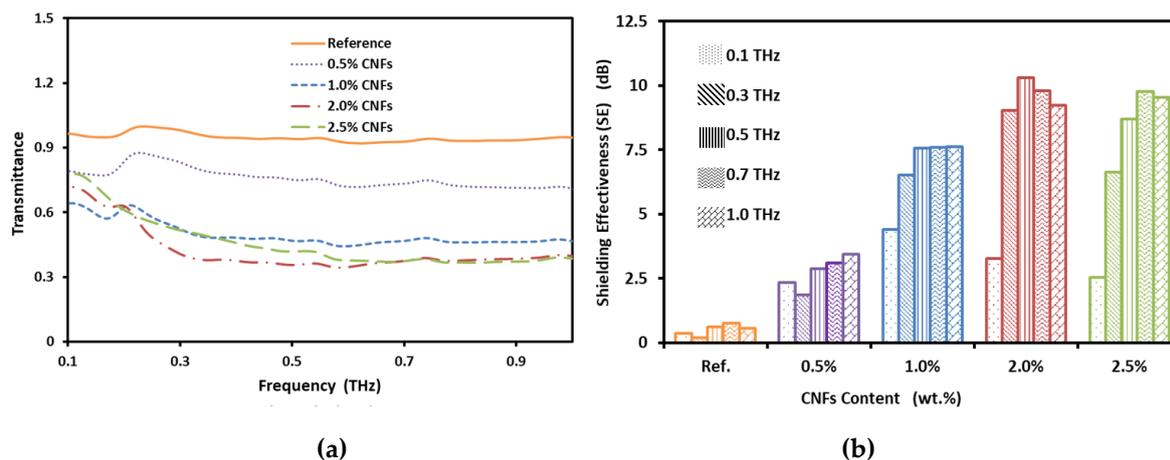
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95 **Figure 3.** (a) Transmission spectra of CNTs/epoxy nanocomposites. (b) SE at frequencies 0.1, 0.3, 0.5, 0.7 and 1.0
 96 THz as a function of CNTs content.

97 Figure 4 (a) shows transmission spectra of CNFs/epoxy nanocomposites. The spectra were
 98 shown the same behavior of CNTs/epoxy nanocomposites. The transmittance decreases with
 99 increasing frequency from 0.1 THz to 1.0 THz and the CNFs/epoxy nanocomposites exhibited a good
 100 low-bandpass filters and a good shield at high THz wave beyond 0.3 THz.

101 The THz SE data for CNFs/epoxy nanocomposites is shown in Figure 4 (b) at frequencies 0.1,
 102 0.3, 0.5, 0.7 and 1.0 THz. The SE increases with increasing frequency and with increasing CNFs
 103 content. The highest values of SE were found for nanocomposite with 2.0 wt. % CNFs which are 9.0 dB,
 104 10.3 dB, 9.8 dB and 9.5 dB at frequencies 0.3, 0.5, 0.7 and 1.0 THz, respectively. In general, the
 105 transmittance and SE of CNFs/epoxy nanocomposites is lower than that of CNTs/epoxy
 106 nanocomposites. This might be owing to a high aspect ratio of CNTs compared with CNFs. The aspect
 107 ratio of the nanoparticles controls the formation of conductive networks inside the polymer matrix
 108 [11] and consequently affects its shielding efficiency.



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Figure 4. (a) Transmission spectra of CNFs/epoxy nanocomposites. (b) SE at frequencies 0.1, 0.3, 0.5, 0.7 and 1.0 THz as a function of CNFs content.

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To understand the carbon nanoparticles contribution to the EMI response of the polymer nanocomposite material, it is useful to consider the dielectric properties. The frequency dependent dielectric constant (ϵ') and dielectric loss (ϵ'') are obtained from the experimentally determined the real (n) and imaginary (k) parts of the refractive index using the following relations [12]:

$$\epsilon' = n^2 - k^2, \quad \epsilon'' = 2nk \quad (2)$$

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Figures 5 (a, b) and 6 (a, b) show the frequency dependence of dielectric constant and dielectric loss for CNTs/epoxy and CNFs/epoxy nanocomposites, respectively. Both CNTs/epoxy and CNFs/epoxy nanocomposites have the same trend through the THz frequency range. The spectra of dielectric constant and dielectric loss significantly affected by carbon content and change in frequency. As frequency increases from 0.1 THz to 1.0 THz, the dielectric loss decreases, whereas the dielectric constant decreases until it becomes constant beyond 0.5 THz.

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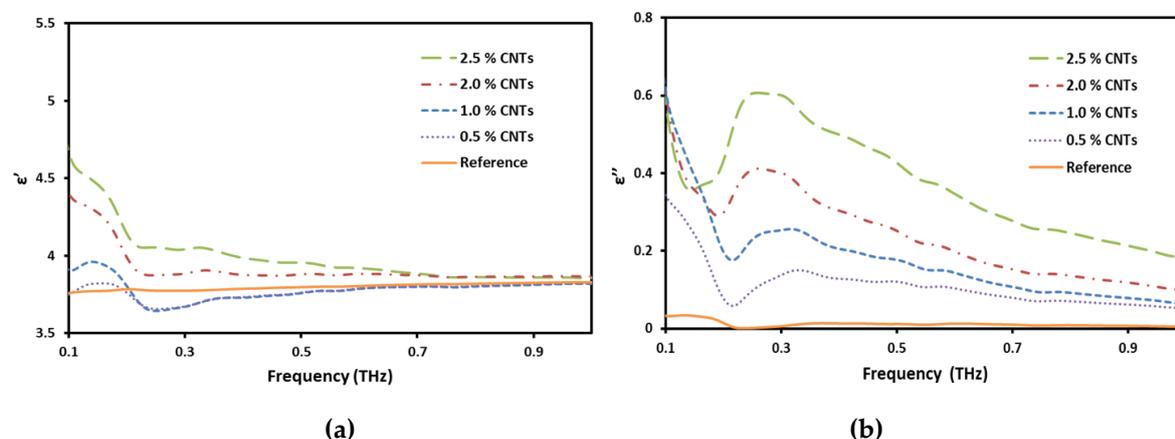
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High values of the dielectric constant at low THz frequencies mean that the polymer nanocomposite acts as a normal dielectric medium. While at high THz frequency beyond 0.3 THz, the low values of the dielectric constant mean that the polymer nanocomposite acts as a semiconductor and reflects the EM waves in this frequency domain proving that both CNTs/epoxy and CNFs/epoxy polymer nanocomposites are good shielding materials in the examined frequency domains.



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Figure 5. Calculated (a) dielectric constant, ϵ' , and (b) dielectric loss, ϵ'' , as a function of THz frequency for CNTs/epoxy nanocomposites.

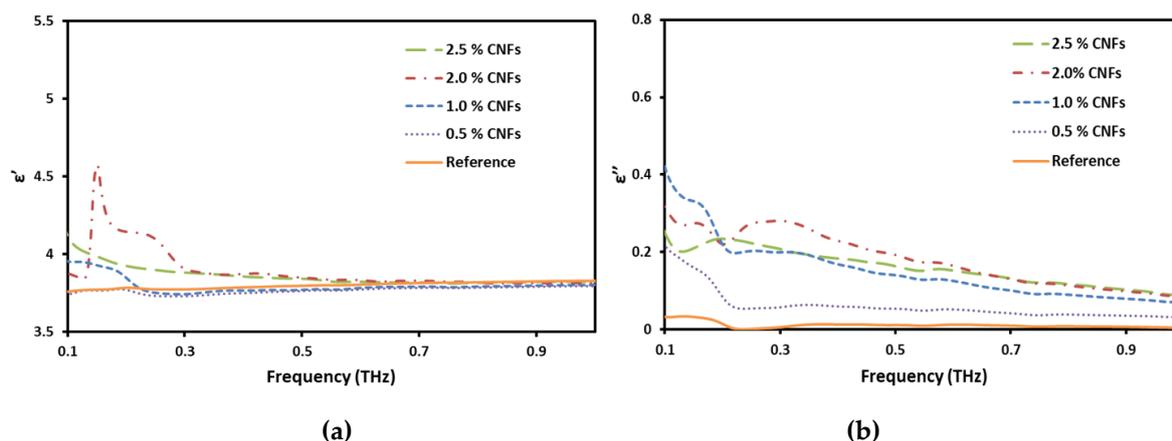


Figure 6. Calculated (a) dielectric constant, ϵ' , and (b) dielectric loss, ϵ'' , as a function of THz frequency for CNFs/epoxy nanocomposites.

4. Materials and Methods

Two types of carbon nanoparticles are used in this study; multi-walled carbon nanotubes (CNTs) supplied by Cheap Tubes, Inc. (Cambridgeport, VT, USA) with a length of 10–30 μm and with an inside and an outer diameter of 5–10 nm, 20–30 nm and carbon nanofibers (CNFs) supplied by Nanostructured & Amorphous Materials Inc. (Los Alamos, NM, USA) with a length of 0.5–20 μm and diameter of 80–200 nm, respectively. The epoxy system was EPOTUF® 37–127 epoxy and the hardener was Aliphatic Amine EPOTUF® 37–614 and were supplied by U.S. Composites, Inc. (West Palm Beach, FL, USA).

Different carbon contents (0.0, 0.5, 1.0, 2.0, 2.5 wt. %) were added into the epoxy resin and ultrasonicated for 1 hour at a temperature 40 $^{\circ}\text{C}$ and then the mixtures were mechanically stirred for 3 hours at a temperature of 90 $^{\circ}\text{C}$. The mixtures were degassed to remove the bubbles and the hardener was then added into mixtures and left overnight. Carbon nanoparticles/epoxy composites were then cured for 2 days at a temperature of 100 $^{\circ}\text{C}$ to ensure full curing. Dispersion of the carbon nanoparticles in the epoxy composites was examined using Field Emission Scanning Electron Microscope, FESEM, Leo Supra 55 (Carl Zeiss SMT, Oberkochen, Germany).

Terahertz measurements were carried out using Terahertz Time-Domain Spectroscopy, THz-TDS, (TPS spectra 3000 system, Teraview Ltd. England) model by using the Attenuated Total Reflectance, ATR, unit (350) with silicon crystal and under Nitrogen gas N_2 purging. The measuring range is in between 0.1 and 1.0 THz with spectral resolution 1.2 cm^{-1} and the number of scans were 1800 scans/s.

5. Conclusions

The transmission spectra, the shielding effectiveness and the dielectric properties of CNTs/epoxy and CNFs/epoxy nanocomposites were examined in terahertz (0.1–1.0 THz) frequency domain. The results show that the transmittance, the shielding efficiency and the dielectric properties are significantly affected by carbon content and frequency change. It is evident that both CNTs/epoxy and CNFs/epoxy nanocomposites exhibited a good EMI shielding at high THz wave beyond 0.3 THz. However, it is demonstrated that CNTs can significantly improve the shielding efficiency than CNFs. The highest shielding effectiveness was found in CNTs/Epoxy nanocomposite with 2.5 wt. % CNTs. Further research is warranted to examine the use of CNTs/epoxy nanocomposites in THz-EMI applications.

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169 **Author Contributions:** Eman Taha fabricated all specimens. Asmaa El-Deeb conducted Terahertz
170 measurements. Eman Taha and Mahmoud Reda Taha participated in data analysis and interpretation of the test
171 results. Asmaa El-Deeb and Usama Kandil participated in different parts of analysis and reviewed all the work.
172 Eman Taha and Mahmoud Reda Taha wrote the manuscript. Eman Taha led the team efforts.

173 **Conflicts of Interest:** The authors declare no conflict of interest.

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