Reviewer 2

Comments and Suggestions for Authors
The manuscript entitled, 'Enhanced Bioactive Properties of Halloysite Nanotubes via Polydopamine Coating' reported preparation of polydoapmine coated HNTs and their biological belongings. I am mentioning some loopholes of this work which should be accounted before publication;
-We thank the reviewer for takin time and suggesting some important points to improve the quality of the manuscript and we improve the manuscript taking into his/her suggestions accordingly.

1. There are lots of articles on bioactive coating based on catechol based monomers. What is the most significant novelty here?
   - The part that emphasize the significance of this work was added at the end of Introduction section as;

   "Various surface modification and coatings techniques such as grafting, etching surface functionalization via chemical treatment, irradiation, and so on are commonly used to obtain surface-modified material with new properties. Specifically, to acquire phenolic coatings of the surfaces, catechol-based monomers are commonly utilized to obtain materials with antioxidant properties. To determine the antioxidant properties of the materials tests such as total phenolic content (TPC), ABTS+ scavenging assays, total flavonoid content (TFC), and iron (II) ion chelating capacity methods are normally used [41]. In this study, in addition to the total phenol content (TPC), total flavonoid content (TFC), and iron (II) ion chelating capacity of PDOPA@HNTs, the AG and AChE enzyme inhibition capabilities of PDODA@HNTs were determined to assess the potentials of PDODA@HNTs materials as an alternative to the traditional drugs used in the treatment of diseases such as diabetes and Alzheimer's."

2. Why the authors choose HNTs irrespective of other anisotropic nanofillers?
   - In the literature, the potential of anisotropic nanofillers can be used in many fields, including environmental applications, biomedical applications, paint, and aerospace materials are being commonly investigated. When incorporating these nanofillers, a number of factors were considered, including their mechanical characteristics, electrical conductivity, optical transparency, and barrier properties (Bandyopadhyay et al. 2020) and so on depending on their application field. In this study, some degenerative diseases and inhibition of different enzymes for the prevention of these diseases e.g., Alzheimer's, Parkinson etc. were examined. Therefore, HNT is chosen because it is a biocompatible, natural, and inexpensive material that can be easily obtained.
   The following information is given on pp 1 of the revised manuscript as;

   "Scientific interest in HNTs has continuously increased as they are modifiable, inherently non-toxic, natural, biocompatible [3,4], and in most cases are inexpensive and reasonable in device application [5,6] as they are abundant in nature."

   -Bandyopadhyay, A.; Dasgupta, P.; Basak, S. *Anisotropic Nanofillers in TPE*; 1st ed.; Springer Singapore, 2020; ISBN 9789811590856.

2. In figure 1a, the structure of dopamine is overlapped. It should be clarified.
   - As suggested, Figure 1a has been rearranged as suggested.

4. How did the author ensure that the coating of PDA was tightly bound to the HNTs surfaces? Is there any cross-check?
   - As the number of PDOPA coatings increases on the HNT, the colloidal stability decreases. Aggregation and sedimentation of PDOPA@HNTs were observed in various solutions. As can be seen in the digital camera images of PDOPA@HNTs composite given below in various solution e.g., water, 0.1 M HCl, 1 M

HCl, 0.1 M NaOH, 1 M NaOH and sonication after 2 min, there is no elution was observed as PDOPA layers were tightly bound to HNT.

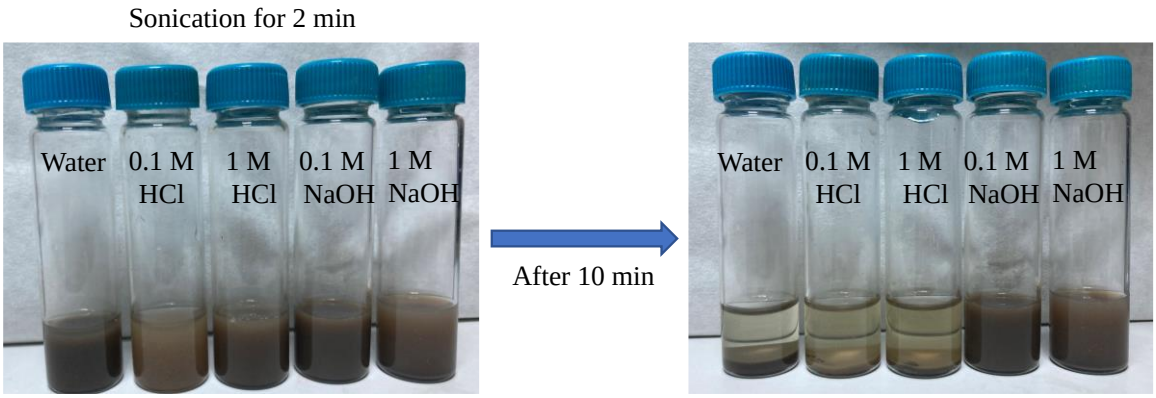


Figure 1. The digital camera images of PDOPA1@HNT composites suspended in various solvents and sonicated 2 min.

Also, as mentioned, the white color of the HNT turned into black as the number of PDOPA coatings was increased. This observation is also in agreement with the literature, and a reference #53 is cited and the following information on pp 6 of the revised manuscript is given as;

"After PDOPA coatings of HNTs, the composites structures e.g., PDOPA1@HNT sus-pended in water, 0.1 M HCl, 1 M HCl, 0.1 M NaOH, 1 M NaOH, and sonication for 2 min, and there was no elution of PDOPA was observed as PDOPA layers were tightly bound to HNTs (data is not shown). This observation is in agreement with the literature [53]."

5. Did the author check the dispersibility or colloidal stability of the systems? Make some arguments on it.

It cannot be said that PDOPA@HNTs show colloidal stability. For this reason, vortex and/or sonication was performed before each use to obtain the suspension for antioxidant and enzyme studies of PDOPA@HNT in.

The following information is provided on pp 9 of the revised manuscript as:

"...As the number of coatings was increased, the colloidal stability of the composites was decreased as each layer of the polymer coating of DOPA generated bigger particles which can easily precipitate under gravity. Also, the PDOPA coatings of HNTs can induce intra particle polymer-polymer interactions between the PDOPA coated HNTs."

6. Several articles related to catecholic coating have been reported in the literature which could be useful for reference: DOI https://doi.org/10.1039/C6RA24153K; https://doi.org/10.1021/acs.accounts.8b00583; https://doi.org/10.1021/acs.langmuir.2c00278; https://doi.org/10.1039/C1NR10969C.

The mentioned references were included on pp 2 and 6 the revised manuscript as;

"...As there is no steric hindrance by dopamine or its oxidized oligomers when they attach to narrow surfaces [34], DOPA can readily bind to surfaces in a single step [35]. DOPA and the similar chemical structure of norepinephrine can render new functionality to the materials by covering almost any surface [36]..."

"...After PDOPA coatings of HNTs, the composites structures e.g., PDOPA1@HNT sus-pended in water, 0.1 M HCl, 1 M HCl, 0.1 M NaOH, 1 M NaOH, and sonication for 2 min, and there was no elution of PDOPA was observed as PDOPA layers were tightly bound to HNTs (data is not shown). This observation is in agreement with the literature [53]..."