

## REVIEWER'S REPORT

Manuscript No.: **IJAR- 59312**

**Title: Behavior of an Agromaterial Based on Neem Seed Extrusion Raffinate in relation to water - Modeling of the Adsorption Isotherm.**

### Recommendation:

Accept as it is .....

**Accept after Minor revision.....**

Accept after Major revision

Do not accept (*Reasons below*)

| Rating         | Excel. | Good | Fair | Poor |
|----------------|--------|------|------|------|
| Originality    |        | ✓    |      |      |
| Techn. Quality |        | ✓    |      |      |
| Clarity        |        |      | ✓    |      |
| Significance   |        |      | ✓    |      |

Reviewer Name: **Dr. ANAPANA GOPAL**

### Reviewer's Comment for Publication.

#### 1. General Comments

This manuscript investigates the water adsorption behavior of a thermopressed agromaterial produced from neem seed extrusion raffinate. The authors determine an adsorption isotherm at 25°C and compare several mathematical models, including BET, GAB, Peleg, Oswin, Hasley, and Henderson. The study reports a Type III adsorption isotherm and identifies the Peleg model as providing the best statistical fit, while also highlighting the practical interpretability of the GAB model.

The topic is relevant to the development, storage and utilization of bio-based materials derived from agro-industrial residues. The manuscript has a potentially useful experimental dataset, particularly because neem seed extrusion raffinate is relatively underexplored as a material source.

However, substantial methodological, statistical, interpretive and editorial revisions are required. In particular, the experimental design and number of replicates are insufficiently described, the moisture-adsorption methodology is ambiguous, the model-selection procedure needs statistical strengthening, several inconsistencies occur in the number of models and statistical terminology, and some conclusions regarding microbial stability and storage conditions are stronger than the presented data support.

**Overall Recommendation: MINOR REVISION**

#### 2. Minor Comments

##### 2.1 Experimental design and number of replicates are not reported

The manuscript provides the preparation conditions of the thermopressed material in considerable detail—200°C, 200 kg/cm<sup>2</sup>, 4 minutes and a 5 × 5 cm mold—and states that the sheets were stabilized at 60% RH and 25°C.

However, the authors do not clearly state:

- number of sheets produced;

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- number of specimens tested;
- number of independent experimental replicates;
- whether adsorption measurements were repeated;
- whether the values in Table 5 are means or individual measurements;
- measurement uncertainty or standard deviation.

This is a major omission for an experimental adsorption study. The authors should clearly describe the experimental unit and provide replicate measurements wherever possible.

### **2.2 The water-adsorption methodology is ambiguous**

The manuscript states that the analysis can be performed “manually or automatically using the Dynamic Vapor Sorption device,” but it does not clearly indicate which method was actually used in this study.

The authors should explicitly state:

- whether a DVS instrument was used;
- model and manufacturer;
- sample mass;
- number of samples;
- equilibrium criterion;
- balance sensitivity;
- equilibration time;
- temperature control;
- RH step sequence;
- whether adsorption was measured continuously or gravimetrically.

If the experiment was conducted using saturated salt solutions, the authors should describe the complete procedure rather than referring alternatively to a DVS device.

### **2.3 The manuscript reports an adsorption isotherm but uses only a limited number of RH points**

The experimental dataset presented in Table 5 contains measurements at 0, 15, 30, 45, 60, 75 and 90% RH.

Seven experimental points are relatively few for fitting and comparing six nonlinear models, especially when some models contain several adjustable parameters.

The authors should explain:

- why these RH points were selected;
- whether intermediate RH levels were tested;
- whether equilibrium was independently confirmed at each point;
- whether the same samples were sequentially exposed to increasing RH.

If only seven points were available, the limitations of fitting multiple nonlinear models should be explicitly acknowledged.

### **2.4 There is an important inconsistency in the number of models evaluated**

The manuscript states that **six** models were selected: GAB, BET, Peleg, Oswin, Hasley and Henderson.

Table 2 also contains these six models.

However, the Results section states:

“Statistical analysis of the seven models...”

and Table 6 contains only **six models**.

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This must be corrected throughout the manuscript. The authors should verify whether six or seven models were actually evaluated.

### **2.5 The manuscript inconsistently describes the number of models in the abstract**

The abstract states that the isotherms were modeled using “several commonly used” models, while the Introduction/Methods effectively describes six models.

The manuscript should consistently state the exact number of models evaluated and name them.

### **2.6 The model-selection criteria require stronger statistical justification**

The manuscript states that the correlation coefficient  $r$  is the primary criterion for selecting the best model.

Peleg gives  $r = 1.000$ , while GAB and Oswin give  $r = 0.999$ .

However, relying primarily on correlation coefficient is not sufficient for selecting among nonlinear sorption models. A very high correlation can occur even when systematic deviations exist.

The authors should consider reporting and comparing:

- $R^2$  or an appropriately defined goodness-of-fit statistic;
- RMSE;
- MAE;
- mean relative error;
- chi-square, where appropriate;
- AIC or another model-comparison criterion if possible;
- residual plots.

Most importantly, the authors should explain why Peleg is considered definitively superior rather than simply having the highest reported  $r$ .

### **2.7 The reported $r = 1$ for the Peleg model should be treated cautiously**

The manuscript describes Peleg's model as having a correlation coefficient equal to 1 and states that its predicted curve “perfectly overlaps” the experimental curve.

With only seven experimental points and a **four-parameter model**, an almost perfect fit is not surprising and does not necessarily demonstrate superior predictive performance.

The authors should discuss possible overfitting and distinguish:

- goodness of fit to the current dataset;
- predictive ability;
- physical interpretability.

If possible, independent validation data or cross-validation should be considered.

### **2.8 The use of “correlation coefficient” should be clarified**

The manuscript presents the formula for “Correlation ( $r$ )” in Table 3 and uses it as a major model-selection criterion.

The authors should clarify exactly what statistical measure is being calculated and whether it is:

- Pearson's correlation coefficient;
- coefficient of determination;
- a custom goodness-of-fit statistic.

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The terminology should be standardized throughout the manuscript.

### **2.9 The MAE/RME terminology is inconsistent**

The Methods section mentions **Relative Mean Error (RME)**, while Table 3 labels the statistic **Mean Absolute Error (MAE)**.

Later, the Results again use "RME" while Table 6 reports "MAE (%)".

The authors must determine exactly which statistic was calculated and use one correct definition and terminology consistently.

### **2.10 Table 3 should be checked mathematically**

The equations presented for the statistical parameters in Table 3 are difficult to interpret because of formatting problems and incomplete mathematical notation.

The authors should carefully verify:

- correlation equation;
- MAE equation;
- SEM equation;
- residual-error equation;
- denominator definitions;
- degrees of freedom;
- units.

The equations should be typeset clearly and checked against the cited references.

## **3. Important Scientific/Interpretive Issues**

### **3.1 The claim that the material can be stored at 90% RH without significant microbial degradation is insufficiently supported**

The Results state:

"This material can be stored at relative humidity levels up to 90% without a significant risk of degradation by microorganisms..."

because the water content remains below 12%.

This conclusion is too strong based solely on adsorption data.

The study did **not** report:

- microbial growth experiments;
- fungal counts;
- bacterial counts;
- water activity measurements associated with microbial thresholds;
- storage trials;
- chemical degradation;
- mechanical degradation.

Therefore, the authors should remove or substantially qualify this statement. Water content alone does not establish microbial stability.

A more defensible statement would be that the measured equilibrium moisture content provides information relevant to storage conditions, while microbial stability requires independent validation.

### **3.2 Water content and water activity should not be treated as interchangeable**

The manuscript discusses relative humidity, water activity and moisture content. Although the relationship between RH and water activity is stated as  **$a_w = RH/100$** , the implications for microbial stability are not sufficiently distinguished.

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The authors should carefully distinguish:

- equilibrium moisture content;
- relative humidity;
- water activity;
- microbial growth thresholds.

This distinction is particularly important because storage recommendations are one of the claimed practical outcomes.

### **3.3 The Type III classification needs more careful justification**

The manuscript concludes that the isotherm is Type III based on the GAB parameter  $C = 7.202$  and states that  $C \leq 10$  corresponds to Type III, while  $C \geq 10$  corresponds to Type II.

This classification should be supported more carefully.

The authors should distinguish between:

- classification based on the **shape of the experimental isotherm**;
- interpretation based on the **GAB parameter C**.

The actual experimental curve in Figure 2 should be discussed in conjunction with the fitted GAB parameters rather than using  $C$  alone as definitive evidence of isotherm type.

### **3.4 The manuscript sometimes overinterprets the adsorption mechanism**

The authors interpret the curve as evidence of multilayer adsorption and relate the behavior to the fibrous composition of the material.

This interpretation is plausible, but adsorption isotherm shape alone does not establish the precise molecular mechanism.

Statements regarding:

- monolayer formation;
- multilayer completion;
- specific polar binding sites;
- hydrogen bonding;
- Van der Waals interactions

should be presented cautiously unless supported by independent physicochemical characterization.

### **3.5 The interpretation of inflection points is too speculative**

The manuscript states that an inflection point below 30% RH may correspond to monolayer formation and that a second inflection between 60 and 80% RH represents completion of hydration multilayers.

These interpretations should be softened unless supported by independent evidence.

The authors could write that the observed curvature is **consistent with** changes in water-binding behavior rather than claiming that specific molecular events have been directly demonstrated.

### **3.6 The discussion of GAB parameters needs greater precision**

The manuscript interprets  $X_0$ ,  $C$  and  $K$  in terms of water affinity and the state of water in multilayers.

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The physical interpretation of GAB parameters can be model- and system-dependent. The authors should avoid presenting these parameters as direct measurements of molecular states without appropriate qualification.

In particular, the statement that the closer  $K$  is to 1, the more multilayer water behaves like liquid water should be carefully referenced and contextualized.

### **4. Methodological Concerns Regarding Material Preparation**

#### **4.1 The source and preprocessing of neem raffinate need additional information**

The manuscript identifies the raffinate as a product of oil pressing followed by aqueous extraction of azadirachtin using twin-screw extrusion.

However, important information is missing regarding:

- neem seed origin;
- seed maturity;
- moisture content before processing;
- pretreatment;
- particle size;
- extrusion operating conditions;
- screw configuration;
- feed rate;
- water/feed ratio;
- residence time.

Because the final material properties may depend strongly on processing history, these parameters should be reported.

#### **4.2 Thermopressing conditions require better reproducibility**

Although temperature, pressure, duration and mold dimensions are given, the authors should clarify:

- whether 200°C refers to plate temperature or actual material temperature;
- pressure application profile;
- heating time;
- cooling conditions;
- sample thickness;
- density;
- final dimensions;
- number of sheets produced.

These parameters are important for reproducing the agromaterial.

### **5. Experimental Moisture-Conditioning Procedure**

#### **5.1 Equilibration criteria are not adequately defined**

The manuscript states that samples were equilibrated for approximately three weeks and then weighed.

However, equilibrium should preferably be defined quantitatively, for example by a mass-change criterion over a specified time interval.

The authors should state the exact criterion used to determine that equilibrium had been reached.

#### **5.2 The use of thymol requires clarification**

The authors placed crystallized thymol in the chamber to prevent microorganisms.

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The potential effect of thymol vapor on the material should be considered, especially because the material is being characterized for adsorption behavior.

The authors should explain:

- amount of thymol used;
- whether samples were physically separated from thymol;
- whether thymol could interact with or be adsorbed by the material;
- whether a control experiment was performed.

### **6. Results and Data Presentation**

#### **6.1 Figure 2 should provide more experimental information**

Figure 2 presents the water adsorption curve against relative humidity.

However, the figure does not appear to show:

- error bars;
- replicate variability;
- number of observations;
- fitted model confidence intervals.

If replicate data exist, error bars should be included.

#### **6.2 Figure 3 is useful but needs improved presentation**

Figure 3 compares experimental observations with six model curves.

The figure would be clearer if:

- axes were standardized;
- units were clearly defined;
- experimental uncertainty were shown;
- model equations were identified in the captions or legend;
- the number of experimental observations were stated.

#### **6.3 Table 5 contains a potentially confusing treatment of the BET model**

The BET model has predicted values only through 45% RH, with dashes at 60, 75 and 90% RH.

This is consistent with the stated BET applicability range below approximately 0.4 water activity, but the manuscript should explicitly explain why the BET model is not extrapolated beyond this range.

The text should also avoid comparing BET performance across the full RH range if the model was deliberately restricted to a smaller range.

#### **6.4 The manuscript refers to “both isotherms” although only one experimental isotherm is presented**

The Results state:

“the correlation coefficients of these models for both isotherms...”

Only one adsorption isotherm appears to have been determined in the manuscript.

This should be corrected to **“the adsorption isotherm”** unless a second isotherm exists but has been omitted.

#### **6.5 Table 6 should include all relevant statistical parameters**

Table 6 currently reports R and, on the following page, MAE and SEM.

It would be clearer to combine all statistical indicators into one table:

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### **Model R/R<sup>2</sup> MAE SEM/RMSE MRE/RME Parameters**

This would allow direct comparison among models.

### **7. Interpretation of Storage and Industrial Applications**

#### **7.1 Industrial suitability is not demonstrated by adsorption data alone**

The Introduction states that the study aims partly to evaluate suitability for preservation and potential industrial applications.

However, adsorption behavior alone cannot establish industrial suitability.

The authors should distinguish between:

- moisture-storage behavior;
- mechanical performance;
- dimensional stability;
- biodegradation;
- thermal stability;
- microbial stability;
- durability;
- processing performance.

The Conclusion appropriately identifies mechanical properties and durability as future work. Therefore, the present manuscript should avoid implying that industrial suitability has already been established.

### **8. Statistical and Model-Validation Issues**

#### **8.1 The authors should provide residual diagnostics**

The manuscript presents seven residual values for Peleg's model and states that their distribution appears random.

However, seven residual values alone are insufficient to establish model adequacy robustly.

A residual plot against:

- water activity;
- predicted moisture content;
- RH

would provide a much stronger assessment.

#### **8.2 Model complexity should be considered**

The Peleg model has four adjustable parameters, whereas GAB has three. The manuscript emphasizes the superior fit of Peleg but does not discuss the effect of parameter number.

Because the dataset contains only seven experimental points, model complexity is important.

The authors should discuss whether the improvement in fit justifies the additional parameters.

#### **8.3 Confidence intervals for model parameters should be considered**

Table 4 provides only point estimates of model parameters.

Confidence intervals or standard errors for the fitted parameters would improve the statistical reliability of the model comparison.

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### 9. References

The reference list is reasonably relevant and includes classical sources such as Brunauer et al., Peleg, Henderson and Oswin, as well as recent studies.

However, several bibliographic and formatting problems need correction.

Examples include:

- "112)-118" in the Aguirre-Loredo reference;
- "419)-421" in the Oswin reference;
- inconsistent spacing between authors and "&";
- inconsistent journal capitalization;
- inconsistent formatting of thesis and conference references;
- inconsistent use of italics;
- possible incomplete bibliographic information.

These should be corrected according to the journal's reference style.

### 10. Language and Presentation

The manuscript requires **substantial English-language editing**.

Examples include:

- "The search for bio-based materials derived from the recovery...";
- "The results of the analysis of the mass of water adsorbed...";
- "The curve from Peleg's model matches the experimental curve perfectly...";
- "This validates both the estimation method and the selected model."

There are also numerous spacing, punctuation and formatting problems, for example "watereven," inconsistent capitalization, and inconsistent terminology.

The manuscript should be professionally edited by a fluent scientific English speaker.

### 11. Specific Terminology Issues

The authors use both:

- **agromaterial**
- **agro-material**
- **adsorbent**
- **material**
- **thermopressed material**
- **hot-pressed material**

These terms should be standardized.

Similarly, the manuscript alternates between **water adsorption**, **water absorption**, and **water behavior**. For an adsorption-isotherm study, the terminology should be technically consistent.

For example, the sentence:

"The GAB model is the second-best model for water absorption..."

should preferably refer to **water adsorption/sorption**, depending on the precise phenomenon being measured.

### 12. Strengths of the Manuscript

The manuscript has several positive aspects:

1. The study addresses an underutilized agro-industrial byproduct.
2. The chemical composition of the starting raffinate is provided, including cellulose, hemicellulose and lignin contents.
3. The thermopressing conditions are reported.

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4. The study uses established adsorption-isotherm models.
5. GAB and Peleg models are appropriately given particular attention because of their relevance to bio-based materials.
6. Experimental and predicted values are presented together in Table 5.
7. Residual errors for the Peleg model are provided rather than reporting only a correlation coefficient.
8. The manuscript identifies future research directions, including desorption, hysteresis, thermodynamic parameters, mechanical properties and durability.
9. The study has potential practical relevance for storage and utilization of neem-derived agromaterials.

### 13. Minor Comments

1. The title should be grammatically revised; "in relation to water" is awkward.
2. Use **Keywords:** rather than "Key words:-".
3. Define **RH** and **aw** at first use.
4. Standardize "thermopressed" and "hot-pressed."
5. Correct spacing throughout the manuscript.
6. Improve Figure 1 resolution and provide a more informative caption.
7. Add sample thickness to the material description.
8. Report the number of independent samples.
9. State whether reported moisture values are wet basis or dry basis; the manuscript generally indicates % dry matter, but this should be explicitly defined.
10. Clarify the phrase "two adsorbents" in the Methods because the manuscript appears to investigate one agromaterial.
11. Correct "both isotherms" to "the isotherm" if only one is studied.
12. Correct "seven models" to "six models."
13. Correct the MAE/RME terminology.
14. Check all mathematical equations for formatting and transcription errors.
15. Explain the dash entries in Table 5.
16. Provide units for every model parameter where applicable.
17. Avoid describing the Peleg curve as "perfect" merely because  $r = 1$ .
18. Avoid unsupported statements about microbial safety.
19. Replace "water absorption" with "water adsorption" where appropriate.
20. Correct all reference formatting according to journal guidelines.

### 14. Final Assessment

The manuscript presents useful preliminary information on the moisture-sorption behavior of a neem seed extrusion-raffinate-based agromaterial. The experimental isotherm and comparison of several established mathematical models provide a potentially valuable contribution to the characterization of this emerging bio-based material. The reported data indicate relatively low moisture uptake at moderate RH and a strong increase at high RH, while the Peleg and GAB models provide close fits to the experimental observations.

Nevertheless, the manuscript in its present form requires **minor revision**. The most important issues are:

- unclear experimental replication and sample size;
- incomplete description of the adsorption-equilibrium procedure;

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- ambiguity regarding DVS versus manual/saturated-salt methodology;
- inconsistency in the reported number of models;
- inconsistent statistical terminology;
- overreliance on correlation coefficient for model selection;
- insufficient consideration of model complexity and overfitting;
- unsupported claim regarding microbial stability at 90% RH;
- overinterpretation of adsorption mechanisms from the isotherm alone;
- insufficient validation of the model;
- several errors and inconsistencies in tables, equations and references;
- extensive language and formatting problems.

### **Final Recommendation: MINOR REVISION**

The manuscript could become publishable after the authors substantially strengthen the experimental-method description, verify the statistical/modeling procedures, correct the internal inconsistencies, moderate unsupported interpretations, and undertake thorough language and reference editing.