ABSTRACT

The equilibrium moisture content (EMC) of food material is defined as the moisture content of the material after it has been exposed to a particular environment for an infinitely long period of time. Equilibrium moisture characteristics of Egg White Powder (EWP) was studied at higher temperatures of 50, 60, 70, 80 and 90°C in the equilibrium relative humidity (ERH) range of 10-78% (at seven levels). The standard gravimetric method was used to determine the EMC-ERH relationships of EWP by employing the various saturated inorganic salt solutions. The EMC of EWP at any particular ERH decreased with an increase in environmental temperature. The EMC of EWP ranged from 2.17 to 3.35 at lower ERH value of about 10% whereas the EMC values ranged from 12.07 to 14.80 at higher ERH value of about 78% when the environmental temperature increased from 50°C to 90°C. EMC values of EWP are different than that of the values obtained at lower
temperatures, hence useful to understand the thermal processing of EWP at higher temperatures. EMC-ERH data obtained in this study was fit into seven EMC-ERH models namely Henderson, modified Henderson, modified Chung-Pfost, modified Oswin, modified Halsey, modified GAB (Guggenheim, Ander-son and de-Boer) and Chen-Clayton models and were evaluated using mean relative percent error ($P_e$), standard error of estimate (SEE) and residual plots. The modified Henderson equation described the EMC-ERH relationship of EWP the best, Henderson, and Chen-Clayton equations gave good fit. The heat of vaporization ($h_{fg}$) of EWP at different moisture contents and temperatures was calculated from EMC-ERH data with the help of the Clausius-Clapeyron equation. The $h_{fg}$ values indicate that the heat of vaporization of EWP increases as the moisture content and temperature decreases and the values are higher than that of the pure water.

Keywords: Modelling; ingredient; standard gravimetric method; moisture content; relative humidity; heat of vaporization.

ABBREVIATIONS

| Acronym | Description |
|---------|-------------|
| BET     | Brunauer-Emmett-Teller |
| DEW     | Dried egg white powder |
| EMC     | Equilibrium moisture content |
| ERH     | Equilibrium relative humidity |
| EWP     | Egg white powder |
| FDEW    | Freeze dried egg white powder |
| GAB     | Guggenheim, Ander-son and de-Boer |
| GRG     | Generalized reduced gradient |
| HEW     | Hydrolysed egg white powder |
| HV      | Heat of vaporization |
| MC      | Moisture Content |
| MW      | Microwave |
| RF      | Radio frequency |
| RH      | Relative humidity |
| SEE     | Standard error of the estimate |
| d.b.    | Dry basis |
| w.b.    | Wet basis |
| $P_e$   | Mean relative percent error |
| $P_{vs}$| Saturated vapour pressure |
| $h^1_{fg}$ | Heat of vaporization of product |
| $h_{fg}$ | Heat of vaporization of pure water |

1. INTRODUCTION

The equilibrium moisture content (EMC) of food material is defined as the moisture content of the material after it has been exposed to a particular environment for an infinitely long period of time. EMC of food materials depends on the relative humidity and temperature conditions of the environment as well as on the species, variety, and maturity. Moisture sorption isotherms describe the relationship between EMC and equilibrium relative humidity (ERH). The isotherms are useful in drying and storage of the products. Isotherms help in designing the thermal processes. Egg white powder (EWP) is an ingredient in many of the bakery foods, bakery mixes, mayonnaise and salad dressings, confections, ice cream, pastas, and many convenience foods [1]. Besides nutritional and functional properties of the dried egg powder, the other advantages such as convenience in storage, transportation, handling, uniformity and microbiological safety make the dried egg powder a versatile ingredient in various food products. However, the physico-chemical properties (turbidity, solubility, residual denaturation enthalpy, surface hydrophobicity, sulfhydryl content and aggregation) of EWP changes during storage, if not handled properly. The important factors that contribute to the changes in EWP during storage are the temperature and relative humidity (water activity, $a_w$). At low moisture content the EWP exhibit the amorphous glassy state. As the moisture content
increases, the water molecules plasticize the amorphous structure of EWP leading to stickiness of EWP [2, 3].

EWP is traditionally pasteurized in a hot room maintained at 58-60°C for 10-14 days in order to enhance the microbiological safety and also to improve the foaming and gelling functionalities [4]. Novel thermal processing methods such as radiofrequency (RF)-assisted and Microwave (MW) assisted methods were found to reduce the processing times to less than a day without compromising the safety and functionality [5, 6]. In this novel methods (RF and MW assisted), processing temperatures up to 90°C for shorter periods were found suitable for the purpose.

EWP is generally processed in 9.0 kg packages kept in the corrugated paper board boxes in order to avoid cross-contamination and to also reduce moisture loss. While processing with the RF, initially RF power is used to increase the EWP temperature inside the package to the desired level (up to 90°C) very rapidly (in few minutes) throughout the mass. This faster heating rate results in desorption of moisture from the product when the product reaches higher temperature and adsorption of moisture during subsequent equilibration with the room temperature. To understand the phenomenon of moisture desorption and absorption at higher processing temperatures, equilibrium moisture characteristics of EWP is needed at higher temperatures. Keeping in view the need, the present study was undertaken (i) to determine the equilibrium moisture content (EMC) and equilibrium relative humidity (ERH) relationships of EWP at higher temperatures, (ii) to evaluate the applicability of general isotherm models for description of EMC-ERH relationship of EWP, and (iii) to calculate the heat of vaporization (h_v) of EWP as a function of moisture content and temperature.

2. MATERIALS AND METHODS

2.1 Materials

Spray dried egg white powder (Advance Nutratech, New Delhi) was purchased from local super market and is used in the study. The moisture content of EWP was determined by hot-air oven method by keeping the triplicated samples in convection hot air oven at 105°C for 24 h. The measured initial moisture content of EWP was 7.72% d.b.

2.2 Determination of Equilibrium Moisture Content of EWP at Various Equilibrium Relative Humidities

The EMC data of EWP at various relative humidities and temperatures was determined by employing a traditional static gravimetric method. Eleven different saturated solutions of various inorganic salts were employed to generate the controlled humidities ranging from 10% to 78% (seven levels except at 90°C) in a closed chamber. The different saturated salt solutions and their corresponding relative humidities at various temperatures [7] are presented in Table 1. EMC-ERH study of EWP was conducted at five different high temperatures of 50, 60, 70, 80 and 90°C.

Seven desiccators to maintain seven different relative humidities at each temperature were taken. About seven different saturated salt solutions were prepared in the desiccators to have seven different levels of ERH ranging from 10% to 78%. The level of saturated salt solutions in each desiccator was maintained below the perforated and raised platform in order to avoid contact of the salt solution with the bottom of a sample holder. About 3 g of EWP was taken into a glass sample holder (weighing bottles). Triplicated sample holders with EWP were placed in each desiccator after taking the initial weights. The desiccators with the samples were placed in an incubator set at a desired constant temperature. The samples were allowed to equilibrate with the environment (ERH) inside the desiccators until there was no discernible weight change. At the end of the experiment, the EMC of the samples was calculated from the initial moisture content, initial sample weight and final equilibrated weight of the samples. The averages of triplicated EMC value were presented during the discussion and raw individual data was used to fit the isotherm model. The above experiment was repeated for all the selected temperatures of 50, 60, 70, 80 and 90°C. The environmental temperatures were chosen randomly and set the temperature in an incubator.

2.3 Isotherm Model Fitting

Seven general forms of EMC/ERH (Isotherm) models namely, Henderson, Modified Henderson, Modified Chung-Pfost, Modified Halsey, Modified Oswin, Modified GAB, and Chen-Clayton as shown in Table 2 were chosen to fit the experimental EMC-ERH data of EWP determined in this study.
Table 1. Saturated salt solutions and their corresponding relative humidities at different temperatures

| Relative humidity level | Salt / relative humidity at five different temperatures, °C |
|-------------------------|-----------------------------------------------------------|
|                         | 50°C | 60°C | 70°C | 80°C | 90°C |
| **Level 1**             |      |      |      |      |      |
| Lithium Chloride        | 11.10| 10.75| 10.51| 10.23|      |
| **Level 2**             |      |      |      |      |      |
| Potassium Fluoride      | 20.80| 20.77| 22.85| 21.74| 23.25|
| **Level 3**             |      |      |      |      |      |
| Magnesium Chloride      | 30.54| 29.26| 27.77| 23.27|      |
| **Level 4**             |      |      |      |      |      |
| Magnesium Nitrate       | 45.44| 46.74| 46.97| 51.43|      |
| **Level 5**             |      |      |      |      |      |
| Cobalt Chloride         | 50.01| 49.66| 49.70| 52.01| 60.21|
| **Level 6**             |      |      |      |      |      |
| Potassium Iodide        | 64.49| 63.11| 61.93| 60.97| 65.00|
| **Level 7**             |      |      |      |      |      |
| Sodium Chloride         | 74.43| 75.50| 75.06| 76.29| 78.68|

The experimental EMC data at all temperatures and relative humidities determined was fitted into a chosen model once at a time with the help of Microsoft Excel Solver. "GRG Nonlinear" solving method was selected for solving the general isotherm equations with the experimental data. The other parameters set while using solver were: constants and sum of squares of residuals. The constants that were yielded after fitting the data by the solver and their respective models were used to calculate the predicted EMC values of EWP.

Then, the EMC-ERH models were evaluated for their suitability in predicting EMC-ERH relationships of the EWP on the basis of mean relative percent error (P<sub>e</sub>), standard error of estimate (SEE) and residual plots. The mean relative percent error (P<sub>e</sub>) was calculated as:

\[ P_e(\%) = \frac{\sum_{i=1}^{n} \frac{|\text{Predicted EMC} - \text{Measured EMC}|}{\text{Measured EMC}}}{\text{No. of data points}} \times 10 \]  

(1)

Standard error of estimate (SEE) was calculated as:

\[ \text{SEE} = \sqrt{\frac{\sum (\text{Predicted EMC} - \text{Measured EMC})^2}{\text{Degrees of freedom of regression model}}} \]  

(2)

The differences between measured and predicted equilibrium moisture content values at various ERHs were defined as residuals. The residuals were plotted against predicted values of EMC. The residual plots were assessed for their appearance as pattern or randomness. If the residuals in the plot are dispersed randomly the model is considered as suitable and then looked for SEE and P<sub>e</sub> values to judge the best fit.

2.4 Heat of Vaporization

The following equation derived from the Clausius-Clapeyron equation was used to calculate the heat of vaporization of EWP [16].

\[ \frac{h_i}{h_g} = \frac{\ln(R_iP_{v1}) - \ln(R_iP_{v2})}{\ln(R_1P_{v1} - \ln(R_1P_{v2}))} \]  

(3)

where, \( h_i \) is heat of vaporization of product in kJ kg<sup>-1</sup>, \( h_g \) is heat of vaporization of pure water in kJ kg<sup>-1</sup>, \( P_{v1} \) and \( P_{v2} \) are saturated vapour
pressures at temperatures $T_1$ and $T_2$, respectively. In computing the heat of vaporization from this equation the values of $RH_1$ and $RH_2$ at a given equilibrium moisture levels were obtained from the best fit isotherm equation for the two temperatures $T_1$ and $T_2$, respectively. The above equation expresses the ratio of the heat of vaporization of the moisture in a product at particular moisture content to that of the heat of vaporization of pure water at the same temperature. The heat of vaporization of EWP at different moisture contents were calculated in the temperatures range from 50°C to 90°C.

3. RESULTS AND DISCUSSION

3.1 Equilibrium Moisture Content of EWP

EMC of EWP in the ERH range from 10.23% to 78.68% at five different temperatures of 50, 60, 70, 80, and 90°C was determined. The moisture content of EWP reached equilibrium at different ERHs in 15-30 days depending upon the maintained environmental temperature. The EWP samples placed at higher temperature of 90°C took minimum of about 15 days to reach equilibrium, whereas samples placed at 50°C took maximum duration of about 30 days to reach equilibrium moisture content. The times taken to reach equilibrium condition for EWP at other temperatures fall in between the range. The EMC-ERH equilibrium conditions for EWP reached in 56 days at 23°C and in 52 days at 35°C depending on the ERH values as reported [17,18]. Further, the time taken to reach the equilibrium of raw and parboiled paddy, brown rice, and bran at different temperatures was 35-40 days at environmental temperatures of 13, 30 and 40°C [19].

The EMC of EWP at any particular ERH decreased with an increase in environmental temperature. The relationship between experimental values of EMC and ERH of EWP at 50, 60, 70, 80 and 90°C is plotted in Fig. 1. As the environmental temperature increased the EMC of EWP decreased. The EMC of EWP ranged from 2.17 to 3.35% d.b. at lower ERH value of about 10% whereas the EMC values ranged from 12.07 to 14.80% at higher ERH value of about 78% when the environmental temperature increased from 50°C to 90°C. From the figure it may be seen the adsorption and desorption behaviour of the EWP from its initial moisture content of 7.72% d.b. The EWP absorbed the moisture at ERH values beyond 47% and desorbed below. The rate of desorption (72% from its initial moisture content) is higher at higher temperature of 90°C while the rate of adsorption (92% from its initial moisture content) is higher at lower temperature of 50°C. The EMC of EWP is higher at lower temperature of 50°C throughout the ERH range than the EMC of EWP at higher temperatures. The EMC of EWP ranged from 3.35 to 14.80% d.b. at 50°C, whereas it increased from 2.17 to 12.07% d.b. at 90°C. As the environmental temperature increases, the water vapour pressure of the moisture within the EWP sample increases that hastens the diffusion of water vapour from the product to the surrounding environment thus resulting in lower EMC values at higher temperatures [20]. The similar trend is exhibited throughout the ERH range.

The EMC-ERH relationships of EWP at lower temperature of 23°C were determined [17]; freeze dried egg white powder (FDEM) at 20°C and 80°C[21]; at lower temperatures of 30, 35 and 40°C[22]; and at 25°C[18]. EMC values of EWP in all these studies were higher than the EMC values of EWP obtained in the present study. It is true that lower temperatures lead to higher EMC values i.e., an increase in temperature causes decrease in amount of water adsorbed, indicating that the material becomes less hygroscopic at higher temperatures when compared to that of the lower temperatures. The results in the present study are in accordance with the general truth. However, the EMC values of EWP are different than that of the values obtained at lower temperatures, hence useful for the thermal processing of EWP at higher temperatures.

The plot of equilibrium relative humidity and corresponding moisture content at a specific temperature is referred as an isotherm. The isotherms of EWP as shown in the Fig. 1 demonstrates the Type 2 sorption isotherm at all the environmental temperatures. In other words, the Type 2 isotherm is a sigmoidal in shape and curves are concave upwards. The Type 2 isotherm takes into account of filling moisture in multi-layers of material’s internal surfaces [23, 24]. The type of isotherms found for most of the food products are of Type 2 similar to the curves found for EWP [23, 24, 25, 26].
Table 2. EMC-ERH (Isotherms) models used to fit the experimental EMC data of EWP

| S. No. | Name of the model    | General form                                                                 | References |
|--------|----------------------|-----------------------------------------------------------------------------|------------|
| 1      | Henderson            | $1-\text{ERH} = \exp(-AM^H)$                                               | [8]        |
| 2      | Modified Henderson   | $1-\text{ERH} = \exp[-A(T + B)M^C]$                                         | [9]        |
| 3      | Modified Chung-Pfost | $\text{ERH} = \frac{-A}{T+B} \exp(-CM)$                                    | [10]       |
| 4      | Modified Halsey      | $\text{ERH} = \exp\left(-\frac{A+BT}{M^C}\right)$                         | [11]       |
| 5      | Modified Oswin       | $\text{ERH} = \frac{1}{1+(A+BT)^C}$                                        | [12]       |
| 6      | Modified GAB         | $\text{EMC} = \frac{A(\frac{C-1}{T})+\text{ERH}}{(1-B+\text{ERH})(1-B+\text{ERH}+\left(\frac{C}{T}\right)^B+\text{ERH})}$ | [13, 14]  |
| 7      | Chen-Clayton         | $\text{ERH} = \exp\left(-\frac{A}{T^B} \exp(-CT^D M)\right)$              | [15]       |

Fig. 1. Experimental values of equilibrium relative humidity and equilibrium moisture contents of egg white powder at different temperatures

3.2 Isotherm Model Fitting

The experimental EMC-ERH data of EWP was fitted in to the seven sorption isotherm models as described in the Table 2. The coefficients of the EMC-ERH models namely, Henderson, modified Henderson, modified Chung-Pfost, modified Halsey, modified Oswin, modified GAB and Chen-Clayton were determined by fitting the experimental EMC-ERH data at all the experimental ERH and environmental temperatures (used all triplicated values). The suitability of the each chosen model to predict the EMC-ERH relationship of EWP was determined from the values of standard error of estimate (SEE), mean relative percent error ($P_e$) and residual plots.

Among these suitable models, the modified Henderson model is the best fit to describe the EMC-ERH relationships of EWP in the studied temperatures range from 50 to 90°C. The other studies found that the GAB model is satisfactory to describe the EMC-ERH relationship of EWP at lower temperature of 23°C [17]. Whereas some other research work reported suitability of BET model as satisfactory to describe the EMC-ERH data of freeze dried egg white powder (FDEM) at 80°C and GAB model at 20°C [21]. The authors speculated about the GAB model in handling the temperature dependency. As the interaction between the solid and the water molecules is negligible at the higher temperatures, the GAB model may not give a good fit when the higher environmental temperatures are involved.
Nonetheless, both the BET and GAB models do not handle the temperature dependence.

It may also be observed that at higher temperature the kinetic energy of the water molecules is high, therefore less water is absorbed at a given water activity [27]. The quantity of sorbed water at a given relative humidity increased as the temperature decreased [23]. The plots of predicted EMC from the suitable models in the ERH range were plotted at each temperature separately from Fig. 2 to Fig. 6. From the figures it may be observed that the closer predictions of EMC values through modified Henderson and Chen-Clayton models when compared with the experimental EMC values. The prediction of EMC for EWP through Henderson model shows high deviation from the experimental values at higher ERH values at both the lowest (50°C) and the highest (90°C) temperatures. However, as described earlier the modified Henderson and Chen-Clayton models described the EMC-ERH relationships of EWP the best throughout the ERH range at all environmental temperatures.

The performance of the each EMC-ERH model found suitable to predict the EMC-ERH relationship of EWP at all the experimental temperatures (50, 60, 70, 80 and 90°C) are plotted in Figs. from 7 to 9. The Figs. from 7 to 9 demonstrate the adequacy of best fit models to describe the EMC-ERH behaviour of EWP in the experimental range.

### Table 3. The EMC-ERH model co-efficients, standard error of estimate (SEE), mean relative percent error (Pₑ) and pattern of residual plots (random / patterned) for egg white powder

| Name of the Model | Model Coefficients | SEE | Pₑ (%) | Pattern of Residual plot | Rank |
|-------------------|---------------------|-----|---------|--------------------------|------|
| Henderson         | $2.5772\times10^{-4}$ | 1.716597 | - | - | 0.039 | 5.57 | Random | 2 |
| Modified Henderson | $2.20739\times10^{-4}$ | 17.46582 | $1.678967$ | - | 0.035 | 5.52 | Random | 1 |
| Modified Chung-Pfost | 408.0855 | 28.32574 | $0.216317$ | - | 0.040 | 5.07 | Patterned | - |
| Modified GAB      | 12.48347           | 0.406105 | 341.6202 | - | 0.057 | 8.37 | Patterned | - |
| Modified Halsey   | $3.203721\times10^{-6}$ | 9.8 $\times10^{-6}$ | 1.721249 | - | 0.093 | 14.86 | Random | 5 |
| Modified Oswin    | $8.01695 \times 10^{-4}$ | 1.42 $\times10^{-4}$ | 2.363855 | - | 0.086 | 12.08 | Random | 4 |
| Chen-Clayton      | $4.074463 \times 10^{-4}$ | 7.2 $\times10^{-4}$ | $0.029513$ | $0.461458$ | 0.038 | 5.91 | Random | 3 |

**Fig. 2.** Experimental and predicted equilibrium moisture contents of egg white powder at 50 °C
Fig. 3. Experimental and predicted (Henderson, modified Henderson and Chen-Clayton) equilibrium moisture contents of egg white powder at 60 °C

Fig. 4. Experimental and predicted (Henderson, modified Henderson and Chen-Clayton) equilibrium moisture contents of egg white powder at 70 °C

Fig. 5. Experimental and predicted (Henderson, modified Henderson and Chen-Clayton) equilibrium moisture contents of egg white powder at 80 °C
Fig. 6. Experimental and predicted (Henderson, modified Henderson and Chen-Clayton) equilibrium moisture contents of egg white powder at 90 °C

Fig. 7. Experimental and predicted equilibrium moisture contents from fitted Henderson model for egg white powder at 50, 60, 70, 80 and 90 °C

Fig. 8. Experimental and predicted equilibrium moisture contents from fitted modified Henderson model for egg white powder at 50, 60, 70, 80 and 90 °C
The predicted EMC-ERH values of EWP at all the temperatures from the suitable models namely Henderson, modified Henderson, and Chen-Clayton were plotted in the Fig. 10 to see the type of Isotherm. The figure shows the obtained isotherms for EWP are of Type 2. This is in consistent with the Type obtained while plotting the experimental EMC-ERH values as in Fig. 1.

Some other researchers reported sigmoidal shapes of the Isotherms drawn for EMC data of solar dried whole egg powder, egg white powder and egg yolk powder at 25°C[18]. Authors also reported the Type II sorption isotherm for both the Hydrolysed Egg White powder (HEW) and Dried Egg White powder (DEW) at 23°C [17].

3.3 Heat of Vaporization of EWP

The heat of vaporization (HV) for EWP was calculated with the help of Clausius-Clapeyron equation (3.5). While calculating the HV values, the relative humidity values (RH1 and RH2) at given EMC levels were obtained from the modified Henderson equation (which was found to be the best to predict the EMC-ERH values for EWP) at the two temperatures T1 and T2. The HV values of EWP were determined for moisture contents ranging from 3 to 15% d.b. at the temperatures of 50, 60, 70, 80 and 90°C. The values of HV of EWP along with the HV of free (pure) water are presented in the Table 4. The data in the table shows that the heat of vaporization of EWP increased as the moisture content and temperature decreased and these values were always higher than that of the pure water. At constant temperature, as the moisture content decreases the HV increases indicating that the removal of moisture is more difficult from the inner monolayers of the powders and needs to expedite more energy to do so. The HV (%) value increased almost the same (~13%) at all the temperatures when the moisture content decreased from 15 to 3% d.b. With reference to the HV of pure water, the increase in HV values at 50°C and 90°C were 31% and 27%, respectively with decrease in moisture content from 15 to 3% d.b. It indicates that heat utilized at higher process temperatures is less when compared to that of the lower process temperatures.
One of the most important thermodynamic grain properties in the drying process is the heat of vaporization ($h_{fg}$). In most of the drying applications the heat of vaporization (HV) is determined by considering the evaporation of free water as given in the steam tables. The use of HV of free water data, especially with grains or lower moisture foods such as EWP at low moisture content levels present considerable error. The moisture content at which the HV approaches the HV of pure water is often taken as an indication of the level of ‘bound’ water in the product.

Drying processes, which usually lower the moisture content to 12-14%, apparently involve the removal of both free and bound or adsorbed water. The equilibrium moisture data may be used as a basis for determining the HV with the help of Clausius-Clapeyron equation [28]. The heat required for a phase change may be predicted from thermodynamic considerations.

Final moisture content of Pasteurised EWP was about 8% d.b. Moisture contents of the study were lower than the safe storage moisture content. Hence these moisture contents obtained in the present study was relevant to the thermal processing not related to the storage [6].

4. CONCLUSIONS

The EMC data of EWP at various relative humidities and temperatures was determined by employing a traditional static gravimetric method. Eleven different saturated solutions of various inorganic salts were employed to generate the controlled humidities ranging from 10% to 78% (seven levels) in a closed chamber at five different high temperatures of 50, 60, 70, 80 and 90°C. The EMC of EWP increased with an increase in ERH at all the experimental temperatures. The shape of the isotherms obtained for the EWP was in sigmoidal shape.
i.e., Type 2, which is a characteristic for most of the food products. EMC values of EWP were decreased with an increase in temperature at all the levels of ERH. Also, the EMC values obtained for EWP in this study were different and lower than that of the literature values which were reported at lower environmental temperatures. The models namely Henderson, Modified Henderson and Chen-Clayton were found suitable to describe the EMC-ERH relationships of EWP and the best among the three is modified Henderson model. The heat of vaporization of EWP increased as the moisture content and product temperature decreased, and these values were always higher than that of the pure (free) water. The moisture isotherms of EWP at higher temperatures are important to understand the desorption and adsorption processes, in design and optimization of drying equipment, in predicting the shelf-life under various storage conditions, and in calculating moisture changes which may occur during storage and in selecting appropriate packaging material.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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