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Research Article

Modelling Moisture Sorption Isotherms of Superabsorbent Polymer Fabric for Desiccant Drying of Crops

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Desiccants are important in the drying of temperature-sensitive grains such as seeds for planting. Superabsorbent polymers (SAPs) have previously been identified for possible application in the drying of crops. Applying the gravimetric technique to measure the sorption isotherms and the effectiveness of the desiccant in crop drying was determined at temperatures 20, 25, 30, 35, 40, 45, 50, and 55°C. Using R statistical software, eleven sorption isotherm models were fitted to the experimental data using the nonlinear regression functions. The coefficients of each model were obtained. The modified Freundlich, modified Henderson, and modified Oswin models best predicted the experimental data for the study temperature range. Using symbolic regression and nonlinear regression in R software, four mathematical models were obtained. R software codes were assembled for the analysis of sorption isotherm models. Compared to the existing models, the developed models were found to give a more statistically accurate association between the temperature, moisture content of SAP fabric, and relative humidity.

1. Introduction

The interaction of molecules of water and other materials, e.g., food, is usually studied by the use of the water sorption isotherms. The isotherms' shape would generally indicate the quantitative state of water for a given material and thus could aid in determining the suitable storage conditions or show if the material is susceptible to degradation due to its moisture content [1, 2].

Superabsorbent materials have a chemical structure that is cross-linked and has hydrophilic parts and thus can attract and hold significant amount of water molecules. Examples include the polyacrylate polymer and acrylate-vinyl alcohol which could be in liquid or powder form or embedded in a fabric to increase the surface area of the material. Their application is varied: in construction, consumer good packaging, and food material packaging. Desiccants adsorb water depending on the vapor pressure difference of air and surface to equilibrium point and can be regenerated using dry hot (50 and 260°C) air and therefore reused repetitively for a process [3].

Geographical regions with tropical climates are characterised by high relative humidity and high temperatures. These conditions catalyse microbial action not only in structural facilities but also in food materials. There are reports of high aflatoxin contamination in food materials especially cereals in those regions, which is contributed by poor storage and preservation mechanisms under the stated air conditions. The design of storage structures could be improved by including mechanisms for air conditioning [4].

When selecting or producing a SAP, it would be required to know the water quantitative state and the hydrophilic capacity; it would be desired that the polymer that would adsorb large quantities of water would not easily leak. For drying applications, the information would be crucial in

Table 1: ERH of saturated salt solutions [8].

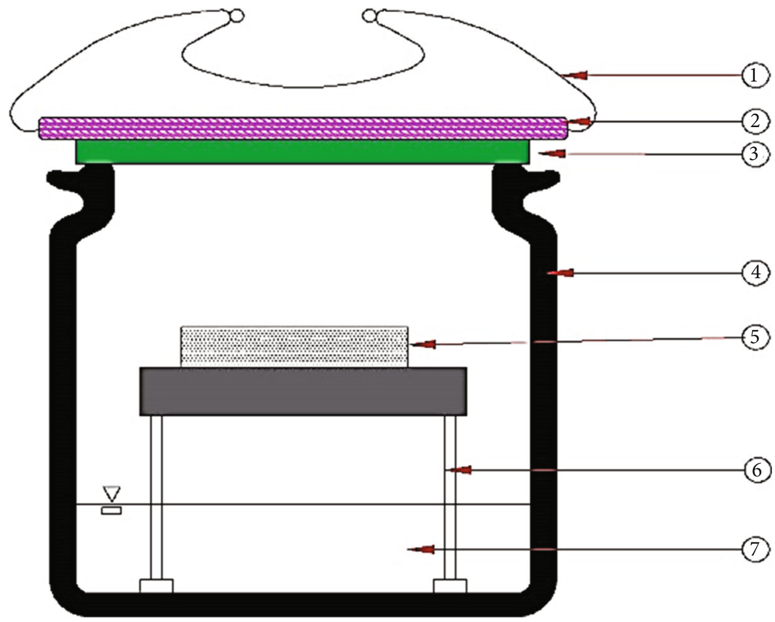
Salt solutions	Equilibrium temperature							
	20	25	30	35	40	45	50	55
Lithium chloride (LiCl)	0.1131	0.1135	0.1128	0.1125	0.1120	0.1116	0.1110	0.1103
Potassium acetate (CH_3COOK)	0.2300	0.2300	0.2200	0.2105	0.2010	0.1950	0.1890	0.1833
Magnesium chloride ($MgCl_2$)	0.3300	0.3300	0.3200	0.3200	0.3160	0.3110	0.3050	0.2993
Potassium carbonate (K_2CO_3)	0.4400	0.4316	0.4300	0.4310	0.4320	0.4290	0.4090	0.4090
Magnesium nitrate ($Mg(NO_3)_2$)	0.5400	0.5300	0.5100	0.5000	0.4840	0.4693	0.4544	0.4400
Sodium nitrite ($NaNO_2$)	0.6500	0.6400	0.6300	0.6200	0.6150	0.6065	0.5980	0.5900
Sodium chloride (NaCl)	0.7500	0.7500	0.7500	0.7500	0.7470	0.7452	0.7440	0.7441
Potassium chloride (KCl)	0.8500	0.8400	0.8400	0.8300	0.8230	0.8174	0.8120	0.8070

Figure 1: Experimental jars.

equipment and process design and optimization. Findings on food-grade SAP fabric lack in this information. This study sought to determine the quantitative SAP-water correlation and evaluate its affinity for water, and the following objectives were set:

(i) To obtain equilibrium moisture sorption isotherm of SAP fabric between 20°C and 55°C

(ii) Model sorption isotherms for predicting equilibrium moisture contents in SAP from experimental data



1- Metal locking ring,
2- Glass Cover Top;
3- Rubber ring,
4- glass jar,
5- SAP Fabric Sample,
6- Tripod Stand,
7- Saturated salt

Figure 2: Experimental jar.

Figure 3: Climate chamber.

Table 2: The isotherm model equations used.

No.	Model	Equation	Reference
1.	Modified Henderson	$W_e = \left[\frac{\ln(1-r)}{-A(T+B)}\right]^{1/C}$	[16]
2.	Modified Oswin	$W_e = (A+BT)\left[\frac{r}{1-r}\right]^{1/C}$	[17]
3.	Modified Halsey	$W_e = \left[\frac{\exp(A+BT)}{-\ln(r)}\right]^{1/C}$	[18]
4.	GAB (Guggenheim–Andersen–de Boer)	$W_e = \frac{ABCr}{(1-Br)(1-Br+BCr)}$	[19]
5.	Modified GAB	$W_e = \frac{AB(C/T)r}{(1-Br)(1-Br+B(C/t)r)}$	[20, 21]
6.	Modified Chung-Pfost	$W_e = \left(\frac{-1}{B}\right)\ln\left[\frac{-(T+C)}{A}\ln(r)\right]$	[22]
7.	Modified Freundlich	$W_e = (A+BT)r^{(C+DT)}$	[3, 23]
8.	Chen-Clayton	$W_e = \frac{-1}{CT^D}\ln\left(\frac{\ln(r)}{-AT^B}\right)$	[24]
9.	Copace	$W_e = \exp(A - BT + Cr$	[25, 26]
10.	Henderson-Thomson	$W_e = \left(\frac{\ln(1-r)}{-A(T+C)}\right)^{1/B}$	[27]
11.	Sabbah	$W_e = \frac{\mathrm{A}r^B}{T^C}$	[25, 28]

Note: A, B, C, and D are model parameters; r is relative humidity in decimal; T is the temperature in °C.

Table 3: EMC of SAP fabric at experimental temperatures (20°C-55°C).

Temperature	Parameter	Values							
20°C	ERH	0.1131	0.23	0.33	0.44	0.54	0.65	0.75	0.85
	EMC	0.009	0.0503	0.0843	0.13	0.1728	0.3733	0.6622	0.8567
25°C	ERH	0.1135	0.23	0.33	0.4316	0.53	0.64	0.75	0.84
	EMC	0.0061	0.0362	0.0588	0.1361	0.2099	0.3829	0.6741	0.817
30°C	ERH	0.1128	0.22	0.32	0.43	0.51	0.63	0.75	0.84
	EMC	0.0082	0.0866	0.0744	0.172	0.214	0.4075	0.6373	0.8686
35°C	ERH	0.1125	0.2105	0.32	0.431	0.5	0.62	0.75	0.83
	EMC	0.0047	0.0187	0.0698	0.1649	0.2481	0.4212	0.6712	0.8992
40°C	ERH	0.112	0.201	0.316	0.432	0.484	0.615	0.747	0.823
	EMC	0.0064	0.0149	0.0591	0.1538	0.2481	0.4394	0.6461	0.7414
45°C	ERH	0.1116	0.195	0.311	0.429	0.4693	0.6065	0.7452	0.8174
	EMC	0.0105	0.088	0.083	0.1637	0.2235	0.3733	0.6893	0.9842
50°C	ERH	0.111	0.189	0.305	0.409	0.4544	0.598	0.744	0.812
	EMC	0.0088	0.0618	0.096	0.161	0.2349	0.3915	0.6802	1.012
55°C	ERH	0.1103	0.1833	0.2993	0.409	0.44	0.59	0.7441	0.807
	EMC	0.0087	0.0737	0.0927	0.1523	0.248	0.48	0.702	1.0528

2. Methods

2.1. Sample Preparation. Sample circular nonwoven SAP fabric was cut of 5 cm diameter of approximately 8 g, the SAP fabric is manufactured by BASF, and it is made up of 100 g/m^2 nonwoven fabric coated with 300 g/m^2 of SAP (coating on both sides with 150 g/m^2). The cut samples were oven dried at 105°C for 24 hours before the start of each experiment.

2.2. Experimental Procedure. Gravimetric method was applied to determine the moisture equilibrium of SAP fabric using the prepared saturated salt solution of relative humidity range 0.111–0.851 as shown Table 1 and Figure 1 [5]. For

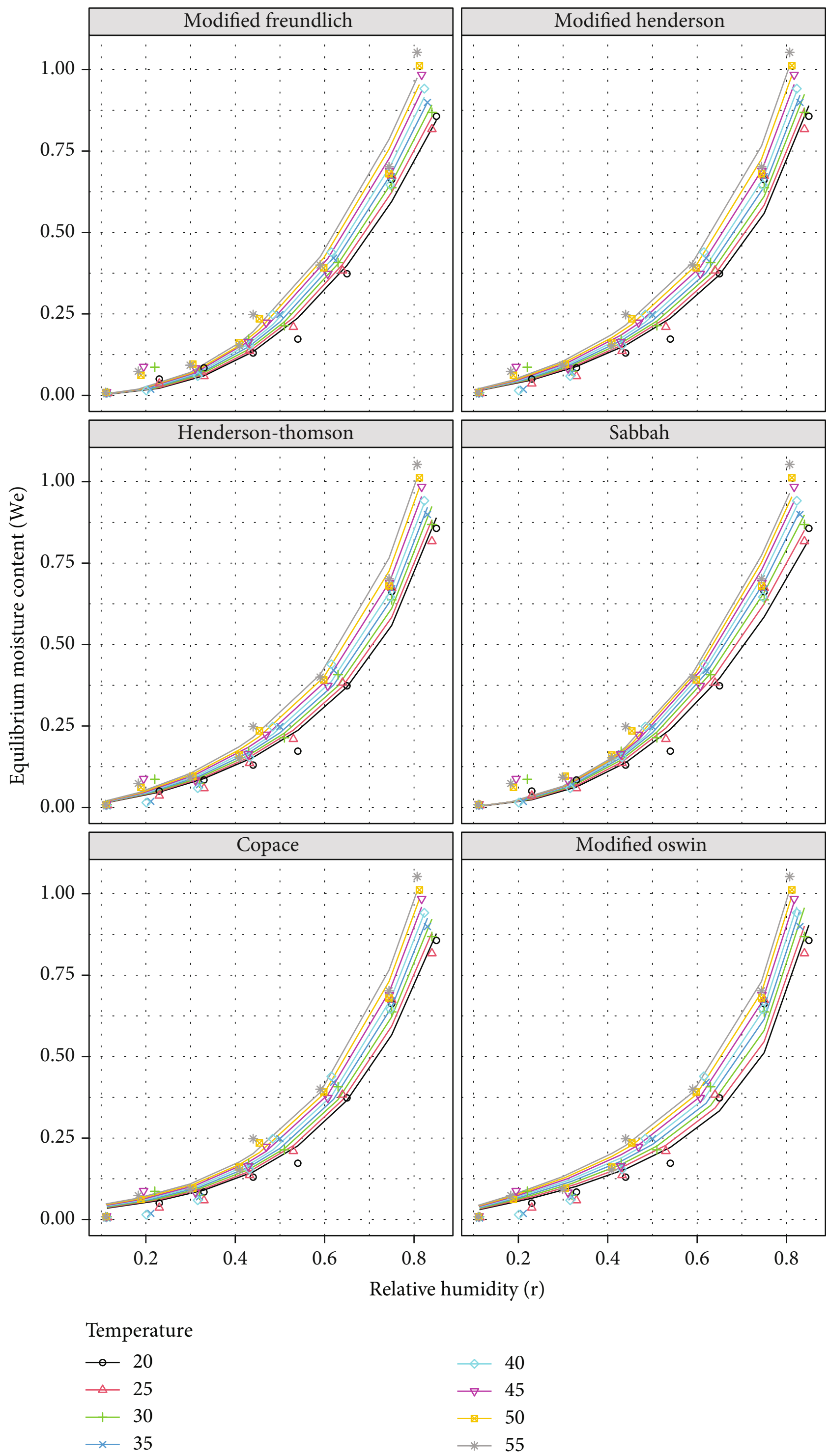


Figure 4: Graphs of EMC of model prediction (lines) and measured values (points) vs. relative humidity at different experimental temperature.

adsorption tests, dry superabsorbent polymer (SAP) fabric was weighed on a Sartorius A200S scale (±0.0001 g). The SAP fabric was put on an aluminium tripod stand with a perforated plastic top and then placed inside the jar (Figure 2). The jar was then hermetically sealed and set in a thermostatically controlled climate chamber (Vötsch Industrietechnik model VCL 0003) (Figure 3) at the selected temperatures (20, 25, 30, 35, 40, 45, 50, and 55°C) for the experimental period [6]. The weights were taken continuously every 12 hours to the equilibrium weight (variation over three consecutive readings was less than 0.001 g). In order to maintain uniform salt solution

Table 4: Empirical model coefficients and statistical results of adsorption isotherm equations.

Empirical model coefficients and statistical indices	Sorption isotherm models									
	Modified Freundlich	Modified Henderson	Henderson-Thomson	Sabbah	Copace	Modified Oswin	Modified Halsey	Modified GAB	Modified Chung-Pfost	Chen-Clayton
A	1.107138	-0.01203724	-0.0120372	0.5236092	-4.0363668	0.1405324	-2.0926071	0.17611	20,847,583.31	1.1749166
B	0.01113	0.67460275	0.6746028	2.7169972	-0.0093462	0.002567	0.0105528	0.99364	12,814,009.33	0.0892532
C	2.8788971	-190.57813466	-190.57813	-0.297895	4.3751963	1.119306	0.8994385	97.0335	2.6449135	5.8675057
D	-0.0042136									-0.2248577
MRE	0.0398348	0.0399003	0.03990026	0.04095892	0.043923818	0.054972	0.0715976	0.10515	0.1185924	0.1189009
RSS	0.0742023	0.0681407	0.06814069	0.07787578	0.074204606	0.126072	0.205966	0.48703	0.5756977	0.4730814
RMSE	0.0340501	0.0326297	0.03262972	0.03488279	0.034050653	0.044383	0.0567293	0.08723	0.0948434	0.0859761
R^2	0.9887788	0.9886147	0.98861474	0.98822565	0.987099615	0.977223	0.9608152	0.90135	0.9067649	0.9233966
Adj.R^2	0.988218	0.988241	0.98824145	0.98783960	0.986676652	0.976477	0.95953	0.89812	0.903708	0.919566
AIC	-240.328	-248.06	-248.059951	-239.513364	-242.6038444	-208.682	-177.267	-122.187	-111.483	-121.77

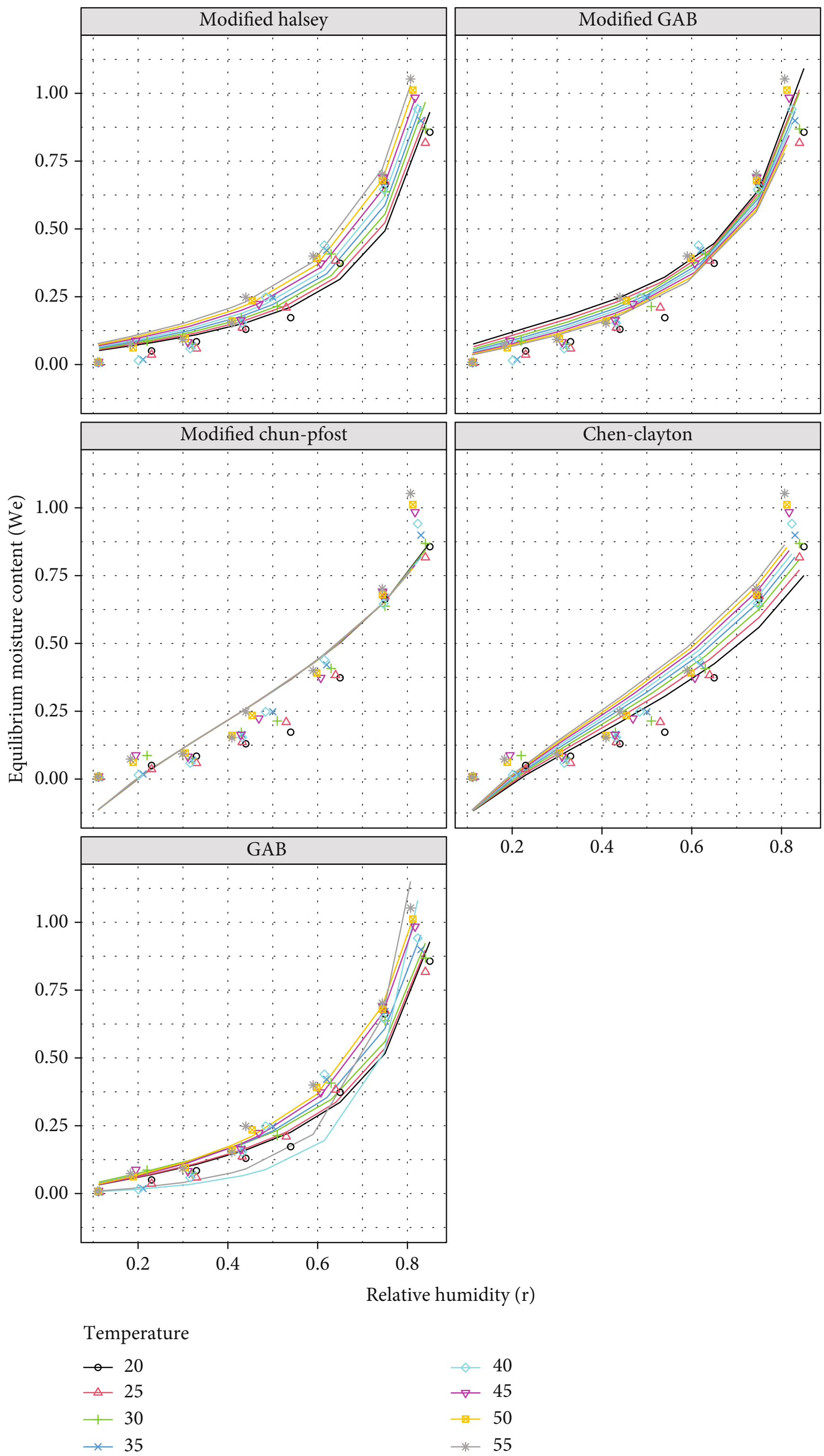


FIGURE 5: Graphs of EMC of model prediction (lines) and measured values (points) vs. relative humidity at the different experimental temperature.

concentration, the jars were swirled intermittently. The oven method was applied to measure equilibrium moisture content (105°C for 24 h); the oven dried fabrics were regenerated and used in subsequent experiments [7]. Three replications were made with each sequence of relative humidity and temperature.

2.3. Model Fitting. The experimental data was fitted in selected models (Table 2) that considered temperature factor, using R software and applying the nonlinear regression function [9–11]. The coefficients of the equations were determined. The fitness of the models was appraised based on the following indices:

Table 5: Empirical model coefficients and statistical results of GAB sorption isotherm equation.

Parameter	20°C	25°C	30°C	35°C	40°C	45°C	50°C	55°C
A	0.1528	0.1591	0.1595	0.1810	1.0000	0.2159	0.2250	1.0000
B	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000
C	1.8141	1.7822	2.4126	1.7549	0.0509	1.2502	1.2704	0.0683
SEE	0.0812	0.0818	0.0579	0.0612	0.1612	0.0267	0.0231	0.1280
MRE	0.6307	0.6603	0.4818	0.5580	1.2325	0.2503	0.1946	1.0566
RSS	0.0330	0.0335	0.0168	0.0187	0.1299	0.0036	0.0027	0.0819
RMSE	0.0642	0.0647	0.0458	0.0484	0.1274	0.0211	0.0183	0.1012
R^2	0.9484	0.9442	0.9730	0.9722	0.9050	0.9955	0.9967	0.9463
Adj.R^2	0.9278	0.9219	0.9622	0.9611	0.8671	0.9938	0.9954	0.9248
AIC	-7.2276	-7.1033	-12.6311	-11.7614	3.7404	-25.0496	-27.3176	0.0517

(a) Residual plot: plots of residuals against the measured values

(b) Mean relative error (MRE)

$$\mathrm{MRE} = \frac{100}{N}\sum_{i=1}^{N}\left|\frac{W_e - W_p}{W_e}\right|. \tag{1}$$

MRE is a measure of the accuracy of a model's predictions. A lower MRE value indicates a more accurate model. Generally, it is deemed that MRE values less than 10% indicate an adequacy for practical purpose application [12].

(c) Akaike information criterion (AIC) ([13–15]

$$\mathrm{AIC} = -2l + 2n\left(\frac{N}{N-n-1}\right), \tag{2}$$

$$2l = -N(\log 2\pi + \log(\mathrm{error}) - \log N + 1). \tag{3}$$

AIC is a model selection method that offsets the characteristic fit of a model with the number of parameters used to achieve that fit. Lower AIC values indicate a better model.

(d) Residual sum of squares (RSS)

$$\mathrm{RSS} = \sum\left(W_e - W_p\right)^2. \tag{4}$$

(e) Root-mean-square deviation (RMSD)

$$\mathrm{RMSD} = \sqrt{\frac{\sum_{i=1}^{N}\left(W_e - W_p\right)^2}{N}}. \tag{5}$$

This function is for error evaluation; if the difference between the experimental and predicted values is large, the RMSD will be large. The error analysis follows normal distribution which is the basis for fitting ordinary least square regression models [14].

Table 6: Developed sorption isotherm equations.

Model	Equation
Model 1	$W_e = \dfrac{Ar^2 \ln(1/\ln(T))}{(\mathrm{r}\ \ln(1/\ln(T)) + B)}$
Model 2	$W_e = Ar^2 \exp(r + TB)$
Model 3	$W_e = Ar^2 \exp(r) \ln(T)$
Model 4	$W_e = A r \ln(B - r) \ln(T)$

(f) Coefficient of determination (R^2)

$$R^2 = 1 - \frac{\mathrm{RES}}{\mathrm{TEE}}. \tag{6}$$

(g) Adjusted coefficient of determination (Adj.R^2)

$$\mathrm{Adj}.R^2 = 1 - \left(1 - R^2\right)\left[\frac{N-1}{N-(n+1)}\right], \tag{7}$$

where W_e is the empirical value of EMC, W_p is the value predicted by model, N is the number of data points, and N is the number of coefficients in each model.

3. Results and Discussion

3.1. Sorption Isotherm Behavior. The results of moisture sorption of the SAP fabric investigated under varying temperatures and relative humidity are presented in Table 3. The sorption isotherms are seen to vary with temperature: the EMC value rose with the rise in water activity at constant temperature states, which would be due to the hydrophilic nature of the SAP. Also, there is a drop in EMC with rising temperature at near-equal water activity conditions. At

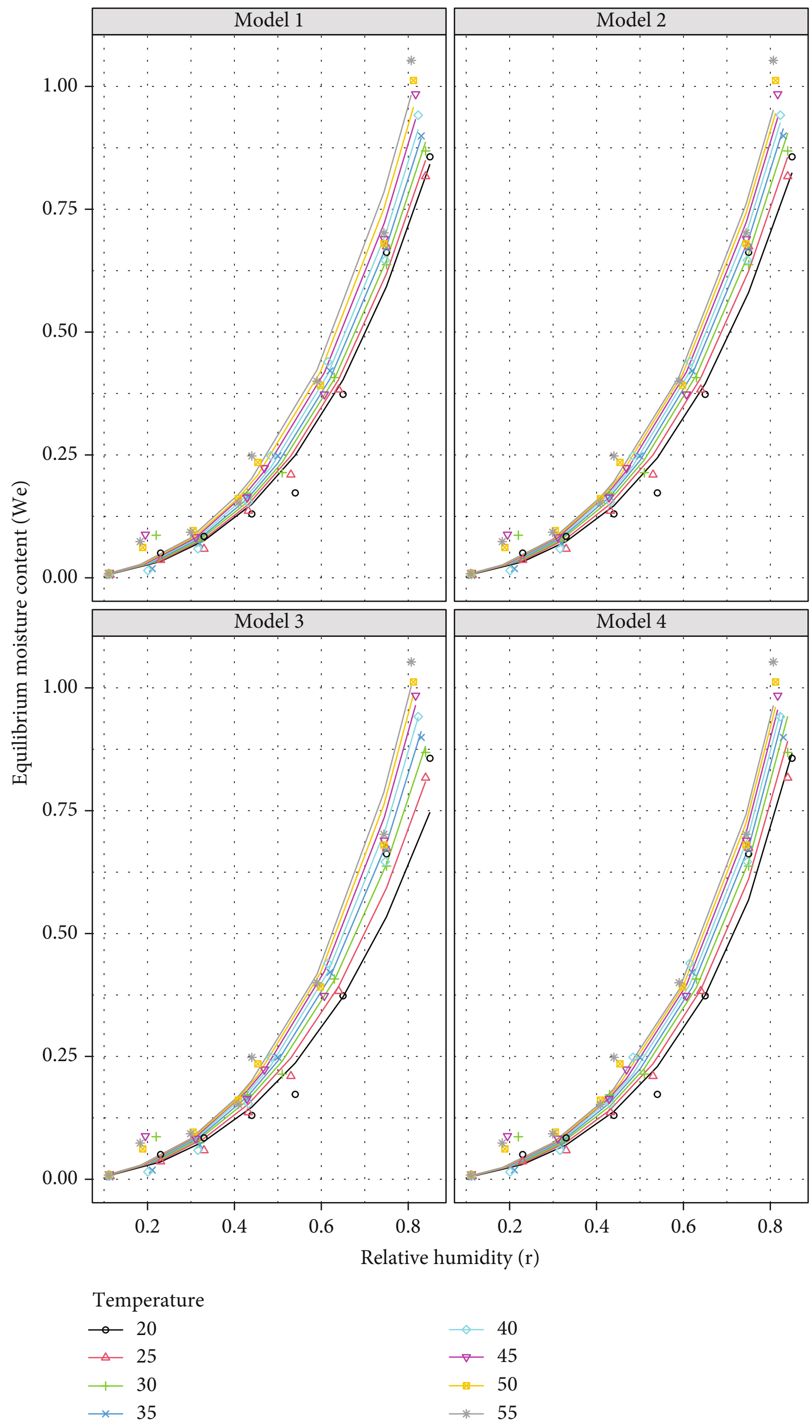


Figure 6: Graphs of predicted moisture content for developed models (lines) and measured values (points) vs. relative humidity for different experimental temperature.

higher temperatures, there is a drop in the dynamic polar sites for water molecules' attraction.

The trends of the water sorption isotherms for the SAP fabric exhibit an upward concave (Figure 4). In comparison to the general profile of the water sorption isotherms for food materials is that the shape of food material is sigmoidal [5]. From the adsorption theory, the isotherm for a substance would concave up at a low water content and if the monolayer adsorption heat is less than that of the condensation heat of the adsorbate [29]. From the findings of this study, it would imply that the value of monolayer adsorption heat for a superabsorbent polymer is less than that for most foods. It could then be postulated that the superabsorbent polymer fabric under this study would be suitable in controlling moisture for food materials given the differences in water adsorption patterns. Dried food materials would easily

Table 7: Empirical model coefficients and statistical results of developed sorption isotherm equations.

Model coefficients and statistical indices	Models			
	Model 1	Model 2	Model 3	Model 4
A	0.4191325	0.1627866	-1.1680986	-0.2007884
B	0.0086178		2.1723147	1.0335341
MRE	0.0373573	0.0388401	0.038297	0.0374914
RSS	0.066497	0.0723338	0.086168	0.0725119
RMSE	0.0322338	0.0336187	0.036693	0.0336601
R^2	0.9893002	0.9883554	0.986084	0.9890596
Adj.R^2	0.9891276	0.9883554	0.9858595	0.9888832
AIC	-251.82597	-248.57357	-235.24091	-246.28392

absorb moisture when exposed to high humidity conditions, and this is a precursor to microbial infestation on the food. From the findings, the SAP fabric could be applied in the storage of dried materials, especially in facilities that are at a high risk of exposure to highly humid conditions.

Prior studies [3] have reported on the application of granular and power SAP; the use of SAP fabric presents a replacement with better performance characteristics with regard to reuse, adsorption capacity, and ease of handling.

3.2. Moisture Sorption Modelling. The SAP moisture sorption data was used to fit the models in Table 2. R software codes were assembled and used in the fitting of the data to models and the calculation of the empirical model coefficients and statistical results of adsorption isotherm equations. The statistical results and residual plots were used in selecting the best-fitting model [5]. The results are as shown in Table 4 and Figures 4 and 5.

The GAB model equation has no temperature variable in it, and thus, parameters are fitted for each temperature; the results are provided in Table 5.

The analysis showed that modified Freundlich model was the most appropriate one in comparison with other existing models (Table 4). This model returned the lowest values of MRE. Compared to other models, it also returned low values of RSS, RMSE, and AIC with a high R^2 value. The mean relative error (MRE) is a suitable metric for evaluating the performance of nonlinear models. The model can be used to estimate the adsorption capacity of the SAP fabric with appropriate input of constants, temperature, and relative humidity. Thus, the size or weight of the fabric for a particular application can be estimated which contributes to the effective use of the material.

From the model equation, both temperature and relative humidity have a direct effect on EMC and that was also observed in the findings (Figure 4 and Figure 5). A moisture gradient is created with both increase in temperature and relative humidity, resulting in loss of moisture for temperature increase while moisture is gained with increase in RH. There is a corresponding change in the EMC as a result of a change in temperature and/or RH. This is illustrated in Figure 4.

This study also investigated the trend of the SAP fabric moisture sorption with temperature and relative humidity variation; this was achieved by modelling the sorption data. The shape of the curve had it that the model could not be linear or polynomial. Using symbolic regression in R software [30], the equations in Table 6 were obtained.

Using nonlinear regression functions in R software, the experimental data was fitted to the models. The coefficients and statistical results of the individual equations were determined. The developed models had a very close prediction of the EMC based on the respective input value of relative humidity and temperature.

The developed models gave a more realistic association between the temperature, EMC, and RH (Figure 6). It is shown that there is a direct correlation between relative humidity and EMC.

Table 7 shows the empirical model coefficients and statistical results of developed sorption isotherm equations.

The developed models are temperature-dependent; thus, temperature variation has a resultant effect on moisture sorption, and the temperature effect is prominent at higher relative humidity values. Model 1 is the best for estimating the adsorption capacity of the SAP fabric compared to the existing models. Other developed models provide very good estimation trends.

From the graphs of moisture sorption, it can be deduced that air of relative humidity below 10% can be used to dry the SAP fabric to below 5% moisture content. When ambient air at room temperature is heated to achieve high temperatures (>60°C) and low relative humidities (<10%), it can be passed through the fabric to dry and regenerate it.

4. Conclusions

The sorption isotherm for SAP fabric after oven drying determined using a gravimetric technique at 20, 25, 30, 35, 40, 45, 50, and 55°C temperatures was established. The value of EMC increased at a constant temperature with the increase in relative humidity. The sorption isotherm curves indicate that SAP fabric can be effectively used in crop drying; wet fabric can be regenerated by passing dry air at relative humidity below 20% or/and temperature above 55°C. The fabric can be used to control the humidity levels inside a grain storage container to prevent seeds from becoming too moist, which can lead to decay. By absorbing moisture, it can prevent condensation on the walls of silos. The modified Freundlich, modified Henderson, and modified Oswin models were found to better represent the experimental data for the study temperature range. Using symbolic regression and nonlinear regression in R software, four mathematical models were developed. Compared to the existing models, the new models give a more realistic association between the temperature, EMC, and relative humidity.

Abbreviations

AIC: Akaike information criterion
EMC: Equilibrium moisture content
ERH: Equilibrium relative humidity
MRE: Mean relative error
RMSD: Root-mean-square deviation
SAP: Superabsorbent polymer.

Data Availability

All data generated during this study are included in this published article.

Conflicts of Interest

The authors declare that they have no competing interests.

Authors' Contributions

PKK was the main researcher and performed the experiments. ANG helped in designing the experiments, reviewing the results, and analysing the data. DOO was the main supervisor and project lead that oversaw the whole research and was responsible for the fund acquisition. JOA was the major contributor in writing and reviewing the manuscript. All authors have read and approved the manuscript.

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