PAPER

EFFECT OF TEMPERATURE, GUM CONCENTRATION AND GUM RATIO ON CREEP-RECOVERY BEHAVIOUR OF CARBOXYMETHYL CELLULOSE-GUAR GUM MIXTURES: MODELING WITH RSM AND ANN

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ABSTRACT

In the present study, the effects of temperature (15, 25, 35°C), gum concentration (1.5, 2.0 and 2.5 %) and gum ratio (25:75, 50:50, 75:25) on the creep-recovery rheological properties of carboxymethyl cellulose (CMC)-guar gum (GG) mixtures were investigated. Within the studied range of experimental design, recovery index (ΔJ) value of CMC-GG gum solution was analyzed based on the design factors. Experimental recovery index responses were modelled using RSM (R^2= 0.9711) and ANN (R^2= 0.9829).

Keywords: artificial intelligence, creep and recovery test, response surface methodologyyrheological properties

1. INTRODUCTION

Gums are widely used in food industry to stabilize emulsion, prevent ice recrystallization (KAYACIER and DOGAN, 2006). The use of two or more gums is a very widespread practice in the food industry due to synergistic interaction between two or more different gums (KAYACIER and DOGAN, 2006). Guar gum (GG) is used as an economical thickening and stabilizing agents in the food industry and is often combined with xanthan, locust bean gum (LBG), or carboxymethyl cellulose (CMC) to increase synergistic changes in viscosity or gelling behaviour (PRADO *et al*., 2005). In generally, gum or gum combination is used in coating, drug delivery and food additives such as dressings, sauces, cereal products, fruit filling, confectionary products and frozen and baked products (PAI *et al*., 2002).

Synergistic interactions of mixed gums have an attractive in the food industry due to reducing production costs and improving product quality and mouth feel (MAO and RWEI, 2006; NORZIAH *et al*., 2006). The determination of viscoelastic properties of product is very important for prediction of product stability in many processing, transportation and storage (STEFFE, 1996). To determine viscoelastic properties of the samples, creep-recovery test is a very crucial method for providing knowledge about the internal structure of material (DOLZ *et al*., 2008). Several studies have been conducted on creep of food materials for evaluating in emulsions (DOLZ et al., 2008; YıLMAZ *et al*., 2012), salad dressing (ZHANG *et al*., 2008), milk (BAYARRI *et al*., 2009), tomato juice (AUGUSTO *et al*., 2013), instant pudding (DOGAN *et al*., 2014). DOGAN *et al*. (2014) also evaluated the effect of four different gums (carrageenan, alginate, guar and xanthan gum) and their different combinations on creep-recovery properties of the pudding samples.

Foods are very complex systems due to possible interactive effects of factors such as temperature, gum type, gum concentration etc. To reduce the number of extensive experimentation and learn about complex interactive effects, modeling and optimization is an inevitable stage for a process (BAS and BOYACı, 2007; MINGZHI *et al*., 2009). Therefore, optimization of the mixed gum formulation is important for both economical and rheological characterization of product (DOGAN *et al*., 2014). For this purpose, response surface methodology (RSM) is the mostly used statistical methods in optimization of rheological properties parameters. However, RSM does not adequately explain nonlinear models (Ross, 1999). To overcome this problem, nonlinear models based artificial intelligence models such as artificial neural networks (ANN) can be considered. ANN has been successfully applied in modeling of creep-recovery behavior for used in formulation of instant pudding (DOGAN *et al*., 2014), deformation of the grape molasses (TOKER and DOGAN, 2013) and construction of predictive models in meat emulsion (YıLMAZ, 2012).

There are many studies have been conducted on the synergistic behavior of gum mixtures like carrageenan-locust bean gum (TAKO and NAKAMURA, 1986), Xanthan-locust bean gum (COPETTI *et al*., 1997), Xanthan-modified guar gum (PAI and KHAN, 2002), CMC-arabic gum (GARCIA-ABUIN *et al*., 2010). However, there is a limited number of studies in the literature using creep and recovery test in order to evaluate and optimize gum combinations. To the best of our knowledge, no published literature was found on the effect of gum concentration, temperature and gum ratio on mixed gum (CMC-GG) optimization by response surface methodology approach based on creep-recovery parameters of the gum combination.

The aim of the this study was to observe the effects of gum concentration, temperature and gum ratio and their interaction terms on the creep-recovery characteristics of CMC-GG mixture samples and then to perform desirability function to determine optimum percentage deformation of mixture by considering several factors simultaneously.

2. MATERIALS AND METHODS

2.1. Materials

Polymer powders of guar gum (GG) and sodium carboxylmethyl cellulose (CMC) (Sigma-Aldrich Corp, St Louis, MO), with nominal molecular weight of 700,000 g/mol were kindly provided by Dr. Kerim YAPICI, Nanotechnology Engineering, Sivas, Turkey.

2.2. Preparation of gum solutions

The CMC and GG powders were dissolved in distilled water seperately at 25°C for 6 h using a magnetic stirrer with gentle shaking in order to prepare 1.5, 2 and 2.5% (w/v) stock solutions of CMC and GG. The CMC stock solution was thoroughly mixed with GG solution at different volume ratios, CMC:GG, such as 75:25; 50:50 and 25:75 respectively.

2.3. Rheological measurements

Creep and recovery tests of the samples measurements were carried out by a stress controlled rheometer (Malvern Kinexus Pro, UK) fitted by a cone-and-plate system. Cone and plate geometries are well suited for studying viscoelastic (stress growth, creep and dynamic) properties of foods. Diameter and angle of the cone were chosen 50 mm and 1°, respectively. The gap between the cone-and-plate was fixed at 0.05 mm for all measurements. A peltier plate assembly was used for temperature controlling purpose during the measurements with ±0.1°C precision.
Firstly amplitude sweep over a stress was performed under at a constant frequency of 1 Hz frequency at 25°C to find linear viscoelastic region (LVR). Then, stress sweep tests were conducted between 0.1 and 10 Hz to determine LVR. Creep – recovery tests were carried out under the constant shear stress of 0.05 Pa (in LVR) at three different temperatures (15, 25, and 35°C) to measure the deformation of mixtures. As known, creep-recovery tests include two parts, one of which is creep phase in which 0.05 Pa. Constant stress was applied for 120 s and the compliance values were recorded as a function of time. In the second part, which is recovery part, the applied stress was removed and then compliance values were also obtained as a function of time for 120 s. In creep-recovery test, the stress response of samples under constant stress in a total time of 240 s was measured. Each measurement was replicated two times. The effect of gum concentration, temperature and gum ratio on the creep-recovery characteristics data of CMC-GG mixed gel was examined. Recovery index or recovery percentage (ΔJ) is used to quantify the viscoelastic response of a materials as given in Eq. (1). The following equation as proposed by DOLZ *et al*. (2008) was used to calculate the percentage deformation of each gum mixtures.

$$\Delta J = 100\frac{J(120) - J(240)}{J(120)} \qquad (1)$$

In this equation J(120) and J(240) refer to the values of the compliance at the final creep phase and at the final recovery phase, respectively.

2.4. Experimental design and response surface methodology (RSM)

One of the models to describe chemical processes in analytical methods is quadratic model in comparison with linear and cubic equation. The design consists of three replicated center points. The individual and combined effects of temperature, gum concentration and gum ratio on recovery percentage were studied using Box–Behnken response surface method (BOX and BEHNKEN, 1960) (BBD) using MINITAB 16.0 (Minitab Inc. State College, PA, USA) (Table 1).

Table 1. Factors and their coded levels of BBD.

Independent variable	Representation	Low (-1)	Center point (0)	High (+1)
Temperature (°C)	X1	15	25	35
Gum concentration (%)	X2	1.5	2.0	2.5
Mixed gum ratio	X3	25:75	50:50	75:25

All creep and recovery tests were conducted in duplicate.
The collected data were fit to a quadratic mathematical model expressed as:

$$Y = b_0 + b_1X_1 + b_2X_2 + b_3X_3 + b_{12}X_1X_2 + b_{13}X_1X_3 + b_{23}X_2X_3 + b_{11}X_1^2 + b_{22}X_2^2 + b_{33}X_3^2 + E$$

where Y is the response (ΔJ, %), b's are regression coefficients. The independent variables; temperature, gum concentration and gum ratio were coded as X_1, X_2, and X_3, respectively. The analysis of variance (ANOVA) and regression analysis were performed to define the coefficients of the predictive model and significant terms using MINITAB 16.0 (Minitab Inc. State College, PA, USA). The optimum conditions for maximizing the apparent viscosity was determined using Response Optimizer tool in MINITAB 16.0 (Minitab Inc. State College, PA, USA).

2.5. Model validation

The constructed model was verified by additional trials at the optimal rheological properties of CMC-GG gum mixtures carried out in triplicate at the design conditions.

2.6. Artificial neural network (ANN) modeling

The input variables used in the ANN models were temperature (X_1), gum concentration (X_2), and gum ratio (X_3). The ANN model was generated as output variable recovery index (ΔJ, Y_1). Several ANNs with different numbers of hidden layer neurons was developed for describing creep behavior of gum mixtures. Input data were randomized into three sets: learning, validation and testing. Usually 30% of data are used for testing and remaining 70% for training and validation (MEHDIZADEH and MOVAGHERNEJAD, 2011). The experimental data including 30 data points was divided so that 70% was used to train the model, 15% was used to validate the model and 15% was used to test the generalization ability of the model. Activation function of hidden layer was "logsig" and the one in output layer was "purelin" as shown in Equations (2-3).

$$\text{logsig}(x) = \frac{1}{1 + e^{-x}} \qquad (2)$$

$$\text{logsig}(x) = \frac{1}{1 + e^{-x}} \tag{3}$$

Training of the network was performed with the function of "trainlm", that updates weight and bias values according to Levenberg-Marquardt optimization. In learning of network "learngd" as adaption learning function was used. The maximum training epochs were 1000, and mean square error was 0.0001. The other parameters of neural network were taken as defaults of neural network toolbox, MATLAB R2011a. The performance of the ANN was statistically measured by the mean squared error (MSE), average relative deviation (ARD) and the coefficient of determination (R^2). These statistics were used to assess the performance of model predictions compared to the actual data. The R_2 represents how well the approximated function predicts the response versus just using the response mean. Values closest to 1 are the best. The MSE is a representation of the difference between the predicted and actual values and gives a sense of how close the predicted values are to the observed values in the units of those values. Lower values of MSE are best and it was calculated using the formula in Equation 4. The ARD represents the percent error. Lower values are best and it was calculated using the formula in Equation 5.

$$\text{MSE} = \frac{\sum_{i=1}^{n} \left(X_{\text{pre},i} - X_{\exp,i}\right)^2}{n} \tag{4}$$

$$ARD = \frac{100}{n} \sum_{i=1} \frac{\left(X_{pre,i} - X_{\exp,i}\right)}{X_{\exp,i}} \tag{5}$$

where $X_{pre,i}$ is the predicted output from observation i, $X_{exp,i}$ is the experimental (target) output from observation i, $\overline{X}$ is the average value of experimental output, and n is the total number of data.

2.7. Selection of the best model

To compare the constructed models and decide on the best one among them, performances of the models was measured using root mean square error (RMSE), mean absolute error (MAE), and coefficient of determination (R^2), which are common comparison tools in modeling. A statistical difference measure test was also carried out to evaluate the performance of the model by calculating root mean square error (RMSE) and mean absolute error (MAE) values as follows:

$$RMSE = \left(\frac{1}{N} \sum_{i=1}^{N} \left(X_{pred,i} - X_{\exp,i}\right)^2 \right)^{0.5} \tag{6}$$

$$MAE = \frac{1}{N} \sum_{i=1}^{N} \left| X_{pred,i} - X_{\exp,i} \right| \tag{7}$$

where X_{exp} is the experimental value and X_{pred} is the predicted value of recovery index (ΔJ, %) value, N is the number of data.

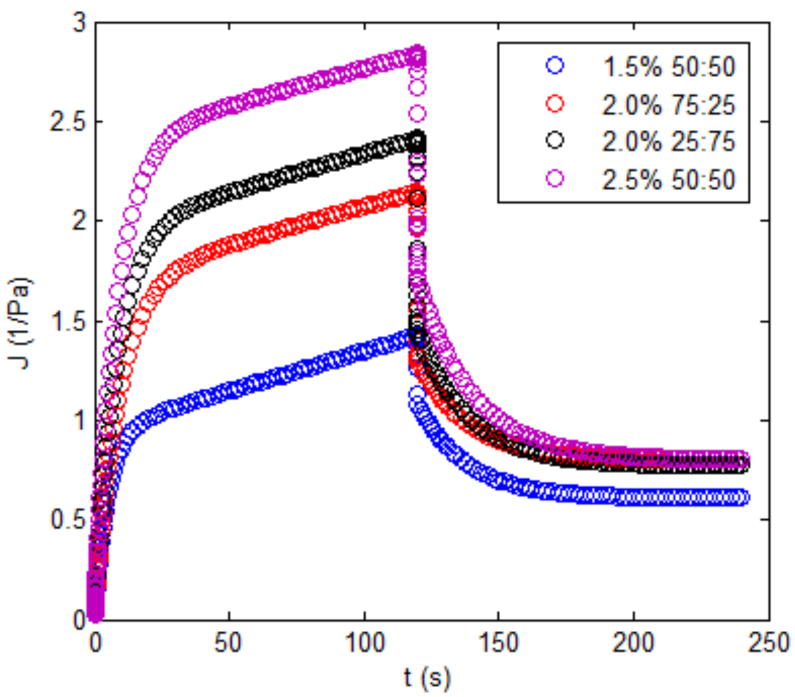
2.8. Statistical analysis

Statistical analyses were accomplished using MINITAB 16.0 to test the significance of different rheological properties of mixed gums. The pairwise comparisons were made by Tukey's test with a significance level of 0.05.

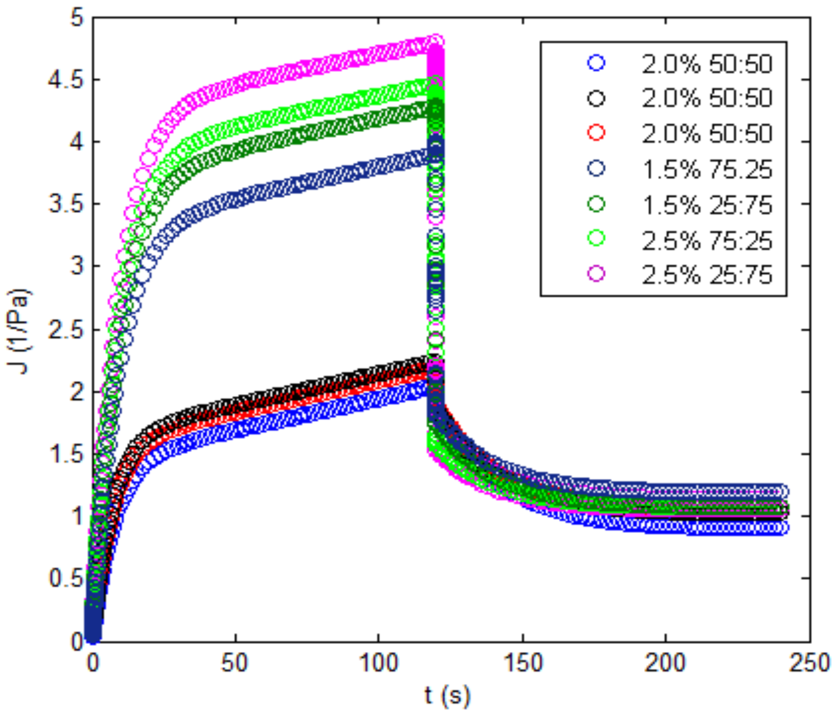
3. RESULTS AND DISCUSSION

3.1. Effect of concentration, ratio and temperature on the creep behaviour of gum mixtures

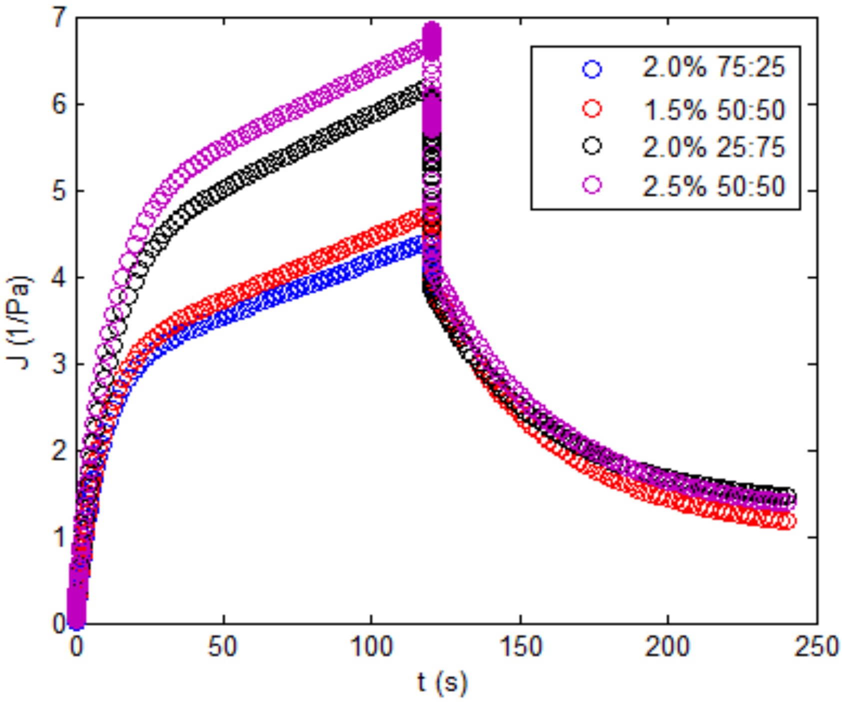
Fig. 1a-c shows graphical representation of creep and recovery experimental results for corresponding to all the mixed gum studied at 15, 25, 35°C respectively, in a time interval between 0 and 240 s. Following the application of stress, 0.05 Pa, deformation occured and deformation was recorded as function of time, J=f(t), during the rheological creep and recovery measurements. At time, t=120, J(120) corresponds maximum deformation value. In Fig. 1c, J(120) values are nearly 7 Pa^{-1} for 2.5% 50:50 at 35°C. On the other hand, it has reached to 2.8 Pa^{-1} at 15°C for the same level as seen in Fig. 1a. This implies that J(120) values gets higher value due to weaking internal structure of gum mixtures and thus higher deformability and more softening structure at 35°C. SOZER (2009) also reported that high compliance value shows weak structure of material.



A) T=15°C



B) T=25°C

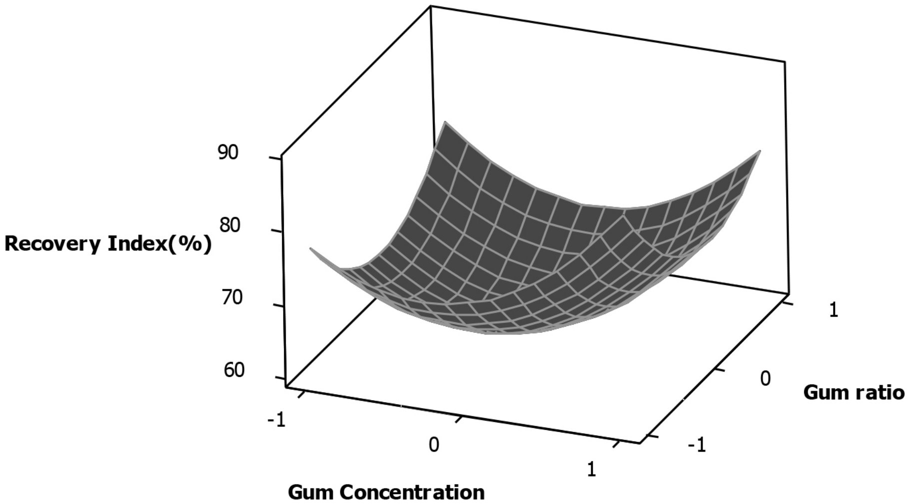


C)T=35°C

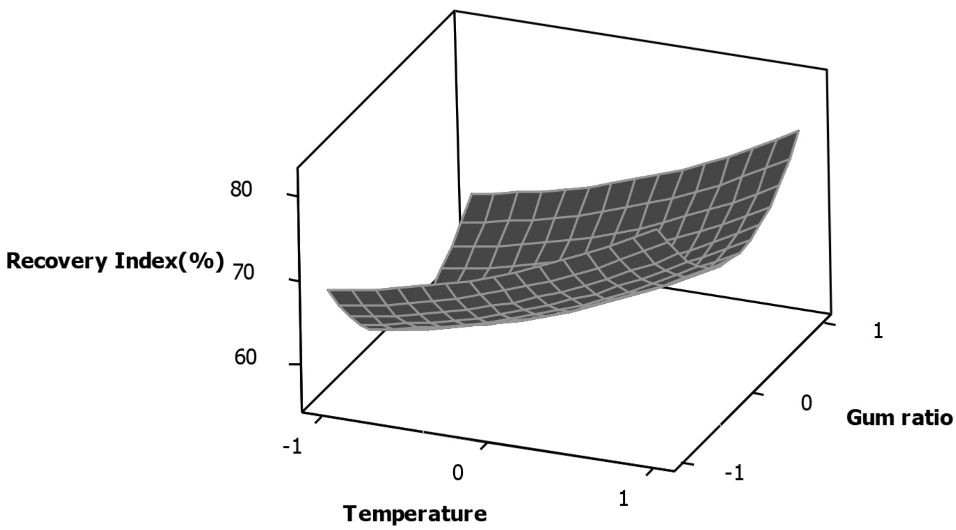
Fig. 1. Creep and recovery plot of gum mixtures for different temperature levels a) T=15°C b) T=25°C c)T=35°C.

CMC and GG is used as thickener and stabilizer in food processing. During food processing, temperature takes critical role due to affected rheological properties of food systems. Upon increasing temperature, it causes a change deformation structure of the gum mixtures. Gum mixture structure is weakened with increase of temperature. Creep compliance value, J(t), increases with temperature due to the related with the softness of the material structure (HAYTA and SCHOFIELD, 2005). These data also show that the recovery index in all gum mixtures is positively correlated with temperature at the fixed gum concentration and ratio (Fig. 2b-c). The drop in viscoelasticity at low temperature level comes from strong or stiffer structure of gum mixtures. On the other hand, there is no significant change for the effect of gum ratios on the recovery indexes for the fixed gum concentration at 15°C ($p>0.05$).

a)



b)



c)

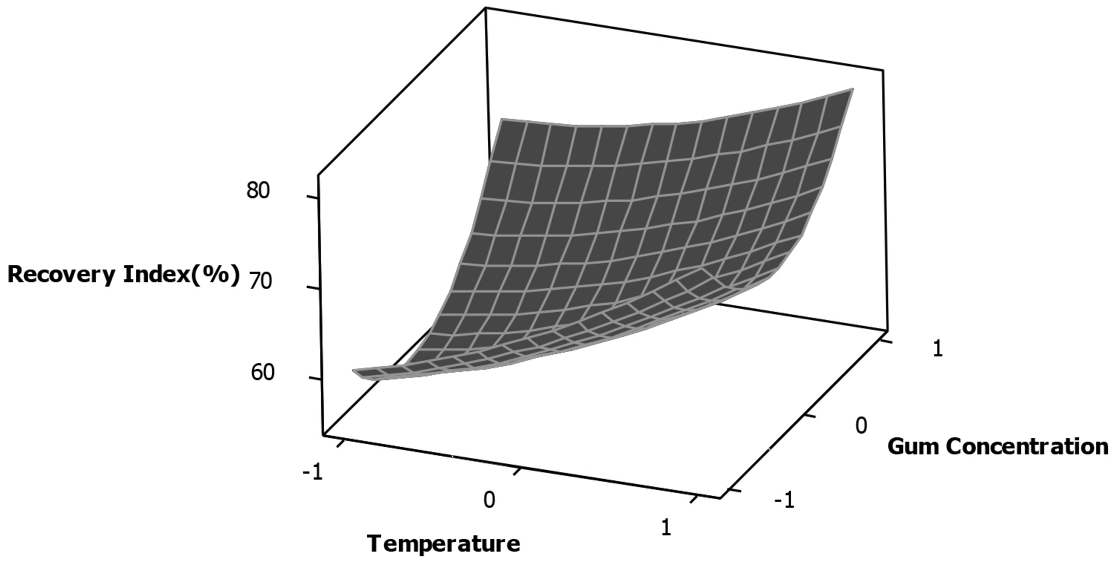


Fig. 2. Response surface showing the effect of a) gum concentration vs gum ratio, b) temperature vs gum ratio, c) temperature vs gum concentration on rheological properties of gum mixtures.

As CMC-GG mixtures concentration increased, recovery index values get higher and viscoelastic response of the mixtures is pronounced (Fig.2a). It is resulted from intermolecular interaction between CMC-GG mixture depended strongly concentration of the mixture. For 2.5% (w/v) gum mixture showed higher compliance, J(t), values indicating higher deformation than concentration of 1.5 and 2% (w/v) for all temperatures as depicted in Figs. 1a-c.

Recovery index, calculated as in Eqn. 1, has changed with gum ratio for 1.5% (w/v) and 2% (w/v) gum mixtures. When the mixed gum has 2% 50:50, designated as center model points, lower deformation is significantly achieved in Fig. 1a and b. Conversely, the strongest synergistic effect on recovery indexes has been observed when changed with mixed ratio (25:75 and 75:25) tabulated as in Fig. 1b. It is associated with increased elastic nature of gum mixture especially in 25:75 gum ratio in higher presence of GG at all temperature levels. Guar gum which highly extend in gum mixture entangles with network structure and binds with CMC. Furthermore, guar gum more dominates the structural arrangement in higher gum mixture indicating that is highly elastic nature and recovery indexes attains to 87.50% at 25°C shown in Table 2.

The results also suggested that hydrophilic intrinsinc interaction between CMC-GG (TIPVARAKARNKOON and SENGE, 2008) which leads to the increasing softening gum structure. Therefore, viscoelastic properties of the mixtures in the aqueous medium are enhanced. Even in lower concentration of gum mixture, significant synergetic effect was also observed for 1.5% 25:75 and 75:25 mixtures in Fig. 1b. It could be related that lower degree entanglement of gum mixture at lower concentration permits to the more elastic effect contribution on mixture nature at 25°C.

Table 2. Experimental measured values according to Box-Behnken response surface method (RSM) versus predicted values of RSM and ANN models for recovery index (%).

Temperature (°C) (X_1)	Gum conc. (%) (X_2)	Mixed gum ratio (X_3)	Recovery Index (%) (Y_1)		
			Experimental	RSM pred.	ANN pred.
35	1.5	50:50	77.24±3.04 ^{bc}	77.90	76.45
25	2.0	50:50	61.52±1.89 ^{f}	60.84	61.36
15	1.5	50:50	58.80±2.45 ^{f}	60.25	58.75
15	2.0	25:75	68.44±0.73 ^{de}	68.24	68.00
35	2.0	25:75	81.68±1.48 ^{ab}	81.86	80.71
25	2.5	75:25	77.91±0.30 ^{bc}	78.96	77.09
15	2.5	50:50	72.08±0.06 ^{cd}	71.42	71.49
25	1.5	25:75	78.11±0.60 ^{bc}	77.05	77.29
35	2.5	50:50	82.48±1.97 ^{ab}	81.01	81.48
35	2.0	75:25	76.00±0.86 ^{bc}	76.63	75.26
25	2.0	50:50	59.96±2.93 ^{f}	60.84	59.86
25	2.0	50:50	61.06±1.27 ^{f}	60.84	60.91
15	2.0	75:25	63.64±3.52 ^{ef}	63.01	63.39
25	2.5	25:75	87.50±0.16 ^{a}	88.56	86.30
25	1.5	75:25	77.27±3.05 ^{bc}	76.19	76.48

Data within the same column with different superscript letter are significantly different (p <0.05).

When higher presence of CMC (75:25) at the 25°C, recovery index value (77.91%) gets smaller value as compared value at 25:75 ratio revealing that more CMC addition led a smaller viscoleastic deformation of mixture and viscous behaviour of mixture increase at the 25°C. Hence, viscous flow deformation gains to importance for the higher CMC content of solution and viscous properties of mixing also increase at 35°C As seen in Fig. 1c, maximum compliance value of 1.5% 50:50 of solution is nearly same than 2.0% 75:25 of solution. It suggests also that strong viscoelastic effect of GG plays major role in determining the direction of synergetic effect of mixed gums at 35°C.
For comprehensive understanding of interactions between concentration, mixing ratio, temperature study on optimization of creep properties of CMC-GG mixtures was achieved to get most resistant and stable product during the efficient food product design.

3.2. Identification of optimized rheological characteristics of carboxymethyl cellulose-guar gum mixtures conditions through response surface modeling

RSM is a frequently used technique for modeling and determining optimal process conditions. Estimation of response surface analysis of experimental results for prediction of recovery index value (ΔJ) in gum mixtures based on temperature (°C) (X_1), gum concentration (%, w/v) (X_2) and gum ratio (v/v) (X_3) per BBD (Table 2), were further elucidated to identify the best process conditions in the ranges tested. The highest recovery index of about 87.50% was obtained at 25°C, 2.5% gum concentration, and 25:75 of gum ratio, whereas the poorest conditions were 15°C, 1.5% gum concentration, and 50:50 gum ratio, giving about 58.80% of recovery index (Table 2).
Second-order polynomial equations (quadratic model) were established to identify the relationship between recovery index and three dependent variables: temperature (°C) (X_1),

gum concentration (%, w/v) (X_2) and gum ratio (v/v) (X_3). Table 3 shows the effect of process variables and interactions on recovery index.

Table 3. ANOVA table for BBD model.

Term	Coefficient	T Value	P Value
Intercept	60.84	76.69	**0.000**
Block	-0.576	-1.623	0.121
Temperature (°C) (X_1)	6.805	14.01	**0.000**
Gum conc. (%) (X_2)	3.569	7.347	**0.000**
Mixed gum ratio (X_3)	-2.612	-5.376	**0.000**
X_1* X_1	2.024	2.830	**0.011**
X_2* X_2	9.780	13.68	**0.000**
X_3* X_3	9.570	13.38	**0.000**
X_1* X_2	-2.013	-2.929	**0.009**
X_1 *X_3	-0.220	-0.320	0.752
X_2* X_3	-2.186	-3.182	**0.005**

R-Sq = 0.9711%; R-Sq(adj) = 0.9581 %; R-Sq(pred) = 0.9299 %.

Another run with excluded insignificant terms according to Table 4 expressed by Eqn. 8:

$$Y=60.84+6.81X_1+3.57X_2-2.61X_3-2.01\,X_1X_2-2.19\,X_2X_3+2.02X_1^2+9.78\,X_2^2+9.57\,X_3^2 \quad (8)$$

where Y is predicted recovery index (ΔJ, %). Equation 8 was found fairly adequate to represent the data with R^2 of 0.97. The insignificant lack of fit for recovery percentage was (P = 0.46 >0.05), which also proved that the model fit the experimental data well.

Table 4. Significance of term coefficients for BBD; A: Temperature (°C); B: Gum concentration (%); C: Mixed gum ratio.

Term	Coefficient	Standard Error Coefficient	T Value	P Value
Constant	60.84	0.775	78.47	0.000
A	6.805	0.475	14.33	0.000
B	3.569	0.475	7.518	0.000
C	-2.612	0.475	-5.501	0.000
A*B	-2.013	0.672	-2.997	0.007
B*C	-2.186	0.672	-3.256	0.004
A^2	2.024	0.699	2.896	0.009
B^2	9.780	0.699	13.994	0.000
C^2	9.570	0.699	13.693	0.000

All factors showed significant effects (p<0.05) on recovery percentage and the interactions between temperature-temperature, concentration–concentration, ratio-ratio interactions, temperature–concentration and concentration-ratio showed significant effects (Table 4). The most important factors determining the recovery index were temperature with highest

coefficient (6.81), which indicates that it is the most dominant factor influencing the overall recovery index value followed by gum concentration (3.57) and gum ratio (2.61) (Table 4).
Estimation of recovery index at varying process variables (temperature, gum concentration, and gum ratio) are represented in response surfaces plots shown in Fig. 2a-c. Fig. 2a shows the effect of gum concentration and gum ratio on the recovery index at mid-point temperature of 25°C. The recovery index of the gum -mixtures was strongly influenced by both variables. A decrease in gum ratio from 75:25 to 25:75 gave the highest recovery indexes of 87.50 % at about 25°C and 2.5 % gum concentration was observed (Fig. 2a). Gum ratio had the most negative effect on the recovery index, meaning that the increase in gum ratio caused decrease in deformation when stress was applied to gum mixture. On the other hand, the effect of temperature and gum ratio on recovery indexes is shown in Fig. 2b, where gum concentration was set at 2.0 % (w/v) as the center point. A linear increase in recovery indexes was observed as temperature increased (Fig. 2b). Similarly, the positive effect of temperature was observed in Fig. 2c, showing the effect of temperature and gum concentration on recovery indexes.
Therefore, to determine the optimal treatment conditions, the response optimizer tool in MINITAB® 16.1 (Minitab Inc.) was used. Optimization was performed regarding maximum recovery index value since low recovery index shows that mixed gums had higher resistance or lower deformation when stress is applied. Higher resistance of the sample against stress is important for product quality during handling and storage (DOGAN *et al*., 2014). Conditions for obtaining optimum recovery percentage of 95.38%, as predicted by the model, were found to be 35°C and 2.5 (%, w/v) and 50:50 gum ratio. These conditions were experimentally tested in triplicate to validate the models predictive ability. A recovery percentage of 90.34±2.79% was obtained and there was no significant difference between the experimental and model predicted values (p>0.05).

3.3. PREDICTIVE MODELING OF CREEP BEHAVIOUR USING ANN

3.3.1. Effect of architecture and topology on neural network

Selection of network topology in ANN modeling is the key issue. Several parameters such as the number of hidden layers, the number of neurons, the transfer function, the epochs and learning rate affected the network topology. The number of hidden neurons is one of the most important parameters of ANN modeling. A high number of neurons performs satisfactorily for training data but may fail for testing data (over-fitting), while a few hidden neurons cause unsatisfactory convergence (under-fitting) (TOKATLI *et al*., 2009). In this study, the number of neurons in the hidden layer was chosen by a trial and error method, varying the neurons from 3 to 12.
Several ANN models with different network topologies were trained, tested, and their performance was evaluated to select the best network topology. The R^2, RMSE, and ARD statistics from training and testing data for different ANN topologies is summarized in Table 5.
Table 5 shows that the 3-9-1 topology was chosen as the best topology (Fig. 3) with the minimum MSE and ARD, and maximum R^2 values. In case of training data set, the coefficient of determination (R^2) and ARD values were 0.9889 and 0.003%, respectively, whereas in testing data set, R^2 was 0.9866 and ARD was 0.009% (Table 5). ANN with 3-9-1 topology was also performed (R^2 values of 0.9889, 0.9784, 0.9866 and MSE values of 1.8478, 6.3565, 4.5024 in training, testing and checking (validation) periods, respectively) (Table 6).

Table 5. Performance of different Artificial Neural Network (ANN) model in estimation of Recovery Index (%).

Network	R^2		MSE		ARD (%)	
	Training	Testing	Training	Testing	Training	Testing
3:3:1	0.9820	0.6355	2.49	145.79	0.005	0.080
3:4:1	0.9942	0.9364	0.88	11.50	0.0003	0.004
3:5:1	0.9859	0.9664	2.17	4.24	0.002	0.039
3:6:1	0.9321	0.9091	12.85	26.97	0.006	0.025
3:7:1	0.9889	0.8964	1.85	4.46	0.002	0.002
3:8:1	0.9806	0.9445	3.38	10.67	0.0005	0.016
3:9:1	**0.9889**	**0.9866**	**1.85**	**4.50**	**0.003**	**0.009**
3:10:1	0.9926	0.9692	1.27	6.21	0.005	0.043
3:11:1	0.9910	0.9888	1.30	11.69	0.002	0.037
3:12:1	0.9970	0.9372	4.47	12.07	0.004	0.031

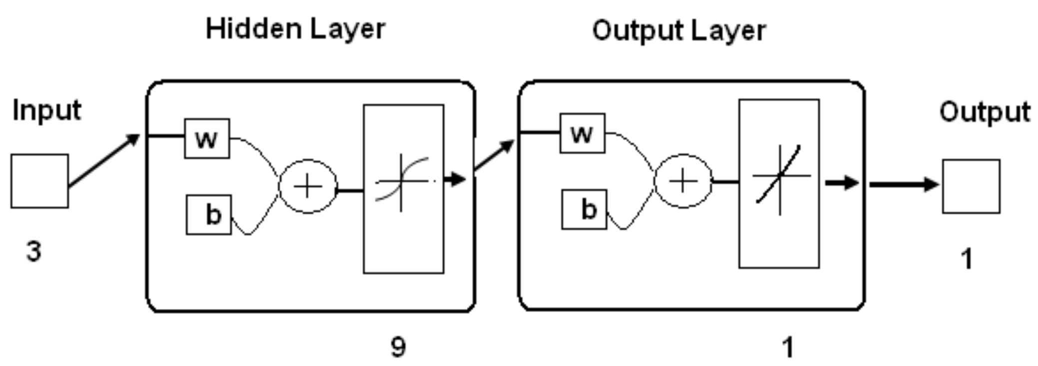


Fig. 3. Schematic representation of ANN to simulate the rheological properties of gum mixtures.

Table 6. Performance of Neural Network Model For Prediction of Recovery Index (ΔJ) Values of Gum Mixture

	Statistical Parameter	Training	Testing	Validating	Overall
Recovery Index (%)	R^2	0.9889	0.9784	0.9866	0.9829
	MSE	1.8478	6.3565	4.5024	3.0417

3.4 Comparison of RSM and ANN models

Fig. 4 shows the plot of predicted recovery index by RSM and ANN against the experimental values. The ANN model exhibited a better relationship between experimental and predicted recovery index values than the RSM model. The results showed that the ANN predictions were closer to experimental values than those of RSM.

Though both the models based on RSM and ANN performed well and offered stable response in prediction; the ANN based approach was better in predicting and fitting than the RSM model for testing the data. Thus, the accuracy of ANN model was higher and has better fitted data than the RSM model.

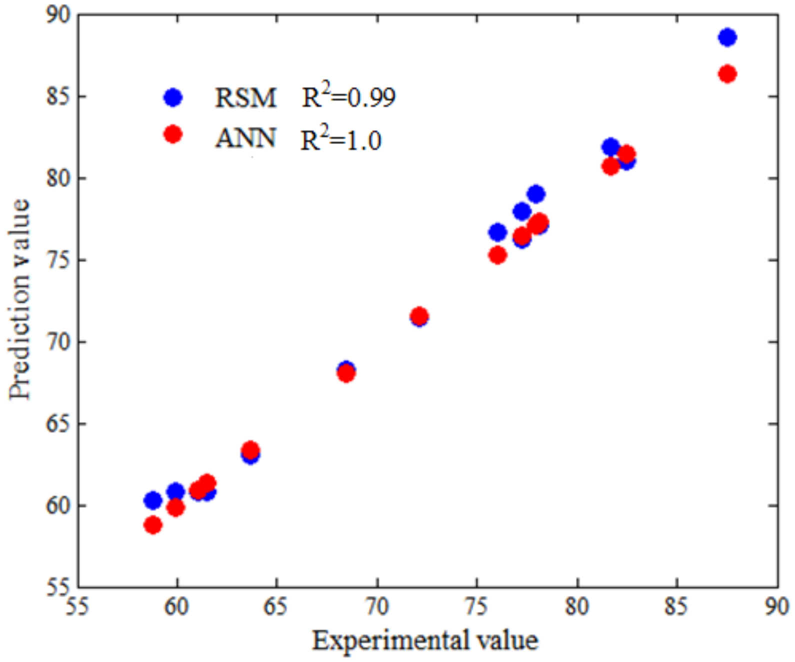


Fig. 4. Verification predicted values versus actual experimental values of RSM and ANN models for rheological properties of gum mixtures.

4. CONCLUSIONS

Detailed creep and recovery viscoelastic measurements at contant 0.05 Pa revealed that recovery index (ΔJ) remarkably changed according to temperature, gum concentration and gum ratio. Elastic or viscous effects contribution on the gum mixture structure affected the creep properties of CMC-GG mixtures. Within the studied range of experimental design, higher content of GG of mixed gum solution increased ΔJ value at all levels. In addition to mixed gum ratio, synergisim effect of gum mixtures was observed especially at 25°C. On the other hand, deformation properties of gum mixtures significantly changed with temperature.

Furthermore, optimization and modeling of gum mixtures are taken into consideration to obtain both best quality of product and economical point of the view. Experimental recovery index responses were optimized using prediction models such as response surface methodology (RSM) and artificial neural network (ANN) models. Several ANN topologies were evaluated in order to predict creep test for gum mixtures. These two methods were also compared for their predictive abilities. The best fit model was identified in ANN model with the highest coefficient of determination (R^2) and lowest MSE values. Therefore, ANN can be considered as a prediction tool for rheological properties of gums mixtures.

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