A multi-mode excitation hardness prediction method based on controlled laser air-force detection (CLAFD) technique

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Abstract

A novel material hardness testing method was proposed based on controlled laser air-force detection (CLAFD) technique. Polyurethane was chosen as the study object. Multi-mode excitation was adopted. Partial least square as the modeling method was used to build the hardness prediction model on the data of laser displacement. Different preprocessing methods were carried out for eliminating the noise of the original data. The results showed the multiplicative scattering correction (MSC) had the best performance. Among four modes, the relationship coefficients of the prediction set (Rp) was above 0.90, and the residual prediction deviation (RPD) was more than 2. This result demonstrated that all four modes could be carried out to test the hardness of polyurethane. Furthermore, the Rp of the transient was 0.93, the RPD was 2.51, the excitation time was 1 s, showing that the transient mode performed with precision in high-speed hardness detection. The highest precision was based on the stress relaxation mode, so we did further study on the interval modeling analysis for the data of stress relaxation mode. The results showed that the hardness could not be predicted if only one single interval was used. However, the performance improved with the increase in the number of intervals. The Rp was up to 0.96, the coefficient of calibration set (Rc) was up to 0.99, and the RPD was 3.54 when the time of the stress relaxation mode lasted 60 s. Based on the results above, the prediction ability would improve further when the relaxation time is increased. The study will provide a new real-time, non-destruction and cross-contamination free hardness detection method for material science, especially for those materials such as artificial biological tissue, function food products, etc.

1. Introduction

Hardness is the measure of resistance to localized deformations, induced by either mechanical indentation or abrasion. It is one of the most important physical properties of materials [1, 2]. The evaluation of hardness is closely linked with the production affecting human life. The evaluation of hardness is not only relevant to the material property evaluation in the area of chemical engineering [3–6], the taste or the texture of food in the area of food engineering [7–9], or the metal property evaluation in the area of mechanical engineering [10, 11]. The indentation test as the main hardness evaluation method is widely applied to the detection of material hardness. The instruments of this common test include the universal testing machine for the metal properties test, the shore durometer for the polymer properties test, the texture analyzer for the food texture test, etc. According to different kinds of hardness detection standards, such as international standard, industry standard, national standard, stable detection is carried out by the indentation test. However, this method can be inadequate to detect in a manner that is efficient, in real-time, non-destructive, and cross-contamination free. With the development of the material science and the sensor technology, researchers developed the tactile hardness test method by imitating the tactile of human fingers [12–16]. However, as a tactile test method, the sensor should touch the surface of the sample. Therefore, this method is not free of cross-contamination, particularly in the
field of biomaterial hardness. Furthermore, this method was influenced by tactile sensor precision and structure; so, the test precision, speed and range of this method need to be improved [14].

In 1994, in order to realize the nondestructive detection of hardness or firmness of rough surface, Prussia et al proposed a method in which the impulsive jet was used to excite the surface of the rough sample and the laser beam was used to detect the compression deformation of the sample [17]. This method is cross-contamination free, high-speed, non-destructive, simple to operate and does not require extensive preparation of the sample. The laser impulsive jet, or laser air-puff (LAP) technique, was applied to the assessment of the mechanical properties of food materials. Some researchers found that there is a correlation between the parameters of the LAP technique and the penetration hardness of peaches, as the regression analysis was found to be $R^2 = 0.77$ [18]. In the study of kiwifruit hardness detection, McGlone et al pointed out that the technique was suitable for the detection of sorting, and it was difficult to detect the hard-textured fruits (fruits with a penetration hardness of more than 5 N) [19]. In their latter study, they determined that the LAP technique could be used for the coarse screening of fruit hardness. Nonetheless, they also suggested that if the higher pressure puff was to be adopted, the detection effect could be better [20]. The conclusion from McGlone et al nearly neglected the potential that the laser air-puff technique could be applied in the quantitative detection of material hardness.

In recent years, however, we have used the LAP technique to explore the evaluation of the tenderness [21] and freshness [22, 23] of chilled beef. In these studies, we found that the reason why the technique couldn’t be applied to the quantitative detection of the mechanical characteristics or qualities of a material was that the air-puff could not be controlled efficiently. Pressure is different depending on the distance from the bottom of the nozzle to the test material; the relationship between the distance and the pressure is nonlinear (proven by the results of the experimental study). Even though, the pressure was set at a specific value by the valve, the force on the surface of the objective materials changes continuously in the process of the compressed sample. There are other influencing factors on the air force of the sample’s surface. The pressure source of the LAP technique comes from the air compressor. The pressure in the air compressor has rapid attenuation high pressure air jet from the nozzle, which causes the fluctuation of the force load on the surface of samples. Moreover, evaluating previous studies, we found that the intracavity of most nozzles was designed as a conical structure to generate conversion. However, this structure lacked rectification for airflow. Therefore, the divergent phenomenon created by the LAP causes air force inconsistencies on the surface of an object to worsen. Those factors could explain why the laser air-puff technique could not carry out the precision detection or the quantitative detection of materials hardness.

Based on the challenges of the LAP technique, this study proposed the controlled laser air-force detection (CLAFD) technique, and a brand-new design and development was carried out. The specific development details of the CLAFD technique is described in section 2.1. However, there are limitations from sampling complexity and ethics to prepare the samples when studying the properties of biological organization. It is easier to build a biological polymer sample with similar mechanical properties, than it would be to build based on biological organization. Using polyurethane materials and adjusting their formulation can be used to modify the mechanical properties of a material. The use of polymer materials to simulate biological tissues is an important research method in life science [24, 25]. Therefore, polyurethane was chosen as the study object, and four excitations were designed to detect the hardness of polyurethane based on the CLAFD technique. Partial least-squares regression (PLSR) combined the different kinds of preprocess algorithms that were used to establish the hardness prediction model of polyurethane, and the specific studied content is as follows: (1) Transient, alternate loading, creep-recovery, and stress relaxation, four excitation modes were compared to predict the hardness of polyurethane; (2) three pre-processing methods were adopted to improve the ratio of signal-to-noise before the hardness prediction model, and the optimum preprocess method was identified. (3) The law of the participation operation of different interval data was specified by interval modelling on the stress-relaxation mode which had the highest prediction precision.

2. Materials and methods

2.1. Study procedure

The study procedure is shown in the figure 1. It mainly includes CLAFD system development, preparation of the sample, hardness test, laser response signal preprocessing, establishment of different hardness prediction model, and prediction performance comparison.

2.2. CLAFD system

CLAFD technique has essential differences in the system control and structure compared to the LAP technique. The traditional plastic and metal nozzles were substituted by a quartz nozzle. Precision machining and fire
polishing were used to manufacture the quartz nozzle. The length of the rectification segment of the nozzle was set to 9 mm, the diameter of the inner cavity was only 3 mm, and the light transmittance was more than 90%. The coaxial of the air path and laser path were established, and the problem of the high requirement of the inner cavity with the opaque materials in the triangulation of the laser displacement sensor was solved completely. Because the intracavity of the nozzle of the CLAFD system was a tenuous hole. The problem of divergence of airflow after jetting from the nozzle was solved as well, as shown in figure 2.

The diameter of the air inlet was 12 mm, much larger than the diameter of the inner hole of the quartz nozzle. This was fundamental to control the air force. The high precision measurement on the air force of the arbitrary height sample was performed by the electromagnetic force balance sensor (the resolution was 0.001 N). In order for the electromagnetic force balance sensor to work with the planer sensing way, it has to be in the range of a laser displacement sensor. The neural network algorithm was used to establish the air-force control model. The variables included the air-force on the surface of the sample, the distance acquired by the laser displacement sensor, and the output pressure of the electric proportional valve. The real-time air-force adjustment was performed by the embedded microprocessor with the air-force control model. Qt widgets for technical applications (QWT) and C++ programming languages were used in developing the software interface in this study. The CLAFD system structure is as follows:

According to the basic requirements of the multi-mode excitation prediction in the study, the CLAFD system was designed and manufactured, as shown in figure 3. The CLAFD system included the body, the lifting test-bed system, the air-force generating system, the deformation detection system, the stress sensing system, the control and information processing system. The lifting test-bed system includes the lifting platform, stepping motor and its driver. The height of the sample on the lifting test-bed system could be adjusted. The air-generating system was installed on the top of the body, including the air compressor, pressure maintaining valve, two-grade air filter, solenoid valve, electric proportional valve, air chamber, and quartz micro-nozzle. And the production and adjustment of excitation air-flow were performed. The air chamber included quartz window,
body and quartz micro-nozzle from top to bottom. The deformation detection system included the laser
displacement sensor, signal amplifier and A/D conversion module. The deformation information of samples
was obtained in real-time by the deformation detection system. The coaxial of the air path and laser path was
completed by the design of the quartz micro-nozzle; the coaxial is the key for precision detection of deformation
information. The stress sensing system included an electromagnetic force balance sensor and its D/A conversion
module, the electromagnetic force balance sensor was installed on the lifting platform. A tray on the
electromagnetic force balance sensor was used to hold the sample. The sample was placed just below the micro-
nozzle. The electromagnetic force balance sensor could acquire real-time air-force information of the sample
surface which is planar sensing. The resolution of the electromagnetic force balance sensor was 0.001 N and the
resolution matched the stress resolution requirement of creep-recovery and stress relaxation.

2.3. Sample preparation
In this study, a new hydrophilic elastic material, cast polyurethane elastomer, Hei-Cast 8400 (HandK Ltd,
Tokyo, Japan), was selected to prepare the patterned samples. The Hei-Cast 8400 is a typical casting material,
which includes the component A (polymer polyol), B (polysiocyanate) and C (heterocyclic diol, as chain
extender). The low viscosity of the A, B, and C mixture provides a fine flowing property and exact shape
replication. Once the mixture is cured, the sample is highly elastic and tear resistant. By controlling the content
of C, the Shore hardness can be adjusted from 10 HA to 90 HA. In this study, the mass ratios of A, B, and C were
set to 100:100:600, 100:100:500, 100:100:400, 100:100:300, 100:100:200, 100:100:150 and the mixture was cured
under 60 °C for 90 min. The Shore hardness of the samples is about 10, 20, 30, 40, and 50 HA. The exact
hardness was tested by a Shore A durometer (Elecall Electric Co., Ltd China). There were 14 samples for every
single shore hardness, therefore, there were 80 samples prepared for the study (figure 4). The geometrical sizes of
samples are Ø23.89 ± 0.60 mm (diameter) × 13.91 ± 0.10 mm (height).

2.4. Multi-mode excitation signals
As shown in figure 5, the excitation signals of the multimode rheological characteristics detection system
included the transient signal, alternate load signal, creep-recovery signal, and stress relaxation signal. The
parameters setting of different modes excitation test is as follows: for the transient test, the air-force was 0.2 MPa,
the time of loading was 1 s, and the repeat times was 5. For the alternate load test, the maximum of air-force was
0.2 MPa, the minimum of air-force was 0.02 MPa, and the time of period was 6 s. For the test of creep-recovery,
the air-force was 40 g, and loading was 30 s. For the stress relaxation test, the deformation of compression was
0.15 mm (the strain was 0.02), the loading time was 60 s.

2.5. Parameters of measurement uncertainty
The CLAFD technique is a pure novel detection technique, the measurement uncertainty is the most important
index to illustrate the performance of this technique. Therefore we proposed the parameters of measurement
uncertainty of four excitation modes as follows: (1) For the transient signal, the replicability parameters were
adopted. By statistical analysis on the 5 excitations of all the samples, the peak error was 0.0021 ± 0.0006 mm,
the peak time error was 0.0149 ± 0.0042 s. (2) For the alternate load signal, we tested the lagging phase angle
between the excitation signals and the response signals. The angle was 0° < δ = (2.95 ± 0.45)° < 90°. The phase
angle of the hysteresis was small, which indicated that the material exhibited a highly elastic state. (3) For the
creep-recovery and stress relaxation signals, the following parameters were proposed for evaluating the control
performance: the rise time $\tau_r$: the time required to rise from zero to the first time that the force reaches the steady-state value of loading after the system triggered (s). The peak time $\tau_p$: the time required to reach the first peak after the force exceeds the steady-state value under loading (s). The settling time $\tau_s$: the shortest time required for the force to reach and maintain within the error band ($\pm 2\%$) of the steady loading value. The delay time $\tau_d$: the time required to reach 50% of the loading stability for the first time. The peak overshoot $\sigma_p$: the percentage of air-force peak value exceeding the steady value of loading. The error band $\pm \Delta$: fluctuation of air-force after the system reaches a steady state. The steady-state overshoot $\sigma%$: the percentage of steady-state loading value exceeding the set value. The value of these parameters is shown in the table 1.

**Figure 4.** Sample of cast polyurethane elastomer.

**Figure 5.** Typical excitation signals of CLAFD system, (a) Transient, (b) Creep-recovery, (c) Alternate load, (d) Stress relaxation.
Table 1. Parameters of measurement uncertainty of the creep-recovery and stress relaxation modes.

| Modes          | $\tau_d$ (s) | $\tau_f$ (s) | $\tau_p$ (s) | $\tau_s$ (s) | $\sigma_p$% | $\sigma$% | $\pm \Delta (\sigma)$ |
|----------------|--------------|--------------|--------------|--------------|-------------|-----------|------------------------|
| Creep-recovery | 0.323 ± 0.005 | 0.508 ± 0.090 | 0.557 ± 0.023 | 0.343 ± 0.031 | 1.980 ± 0.086 | 0.340 ± 0.021 | 0.068 ± 0.013 |
| Stress relaxation | 0.320 ± 0.091 | 0.599 ± 0.104 | 0.768 ± 0.161 | 0.826 ± 0.125 | 1.849 ± 0.090 | 0.315 ± 0.292 | 0.099 ± 0.008 |
2.6. Hardness test
After the sample of polyurethane was prepared, the shore durometer (ELECALL Electric Co., Ltd China) was used to test the hardness of every single sample. Five unreplicated points were selected randomly to be tested, and the average value of the hardness of five points was regarded as the hardness of the polyurethane sample.

2.7. Data processing
The signals could be influenced by the system vibration, airflow field, and the surface shape of the sample during the hardness test. This approach is different from the traditional hardness detection method of measuring the depth of an indentation in the material created by a given force. The entire signal data from different modes of CLAFD system was used as modeling data to predict the hardness quantitatively. Therefore, the data processing included two parts, one of them was the preprocessing for the original data, the other part was the establishment of the prediction model of polyurethane hardness. Matlab R2016a (Mathworks Inc., Natick, MA, USA) was used to process data and establish the hardness quantitative prediction model in the study.

2.7.1. Preprocess of original data
Three preprocess methods, Savozky-Golay (S-G) convolution smoothing, standard normal variate transformation (SNV), and multiplicative scattering correction (MSC) were used to preprocess the various types of modes data.

2.7.2. Prediction model establishment
As a good alternative to the classical multi-linear regression, the principal component regression method is more robust. Partial least-squares regression was used in the study to build the quantitative prediction for the hardness of polyurethane. PLSR is a multivariate calibration with factors analysis based on principal component analysis (PCA). Only the independent variable matrix is decomposed in the PCA, and the redundant information is eliminated. However, both the dependent variable and independent variable matrix are analyzed in the application of PLSR. In this study, the data of hardness was the dependent variable and the data of laser displacement was the independent variable, the former was introduced into the progress of the decomposition of the latter. Therefore, the principal components of laser displacement was associated with the hardness of polyurethane. The basic progress of modeling is as follows:

Assume there were \( n \) polyurethane samples, and the matrix of laser placement with the sampling time was \( X = (x_{ij})_{n \times p} \), the \( p \) was the sampling number. The corresponding matrix of hardness was \( X = (x_{ij})_{n \times p} \). The principal component decomposition was carried out on the matrices of laser displacement and hardness of polyurethane as shown in the formula (1) and (2).

\[
X = \sum_{i=1}^{r} t_i q_i^T + E = TP^T + E \tag{1}
\]
\[
Y = \sum_{i=1}^{r} u_i q_i^T + F = UQ^T + F \tag{2}
\]

Where \( T \) was the score matrix of \( X \), and \( P \) was the loading matrix of \( X \), \( U \) and \( Q \) were the score matrix and the loading matrix respectively of \( Y \). \( E \) and \( F \) were the error matrixes of \( X \) and \( Y \). \( r \) was the number of principal components. The linear regression relation between \( U \) and \( T \) was as follows:

\[
u_i = b_i t_i \tag{3}\]

Where \( u_i \) and \( t_i \) were the \( i \) column of \( U \) and \( T \). And then the \( T \) was rotation transformed. The effect of the rotation transformation was that information of \( U \) was introduced when the \( T \) was decomposed from the \( X \), and information of \( T \) was introduced when the \( U \) was decomposed from the \( Y \). The specific method was that the variables were transformed during the iteration. \( P^T \) was calculated by \( T \) replacing with \( U \), and \( Q^T \) was calculated by replacing the \( U \) with \( T \). After the relation between \( T \) and \( U \) was solved, the hardness could be predicted by the following equation:

\[
Y_{\text{new}} = T_{\text{new}}BQ^T = X_{\text{new}}PBQ^T \tag{4}
\]

Where \( Y_{\text{new}} \) was the matrix of a new group polyurethane hardness, \( X_{\text{new}} \) and \( T_{\text{new}} \) were the measured value of hardness and its score matrix respectively. The hardness value of a single one sample was calculated by equation (5):

\[
Y_{\text{new}}^T = T_{\text{new}}^TBQ_{\text{new}}^T = X_{\text{new}}^TPBQ_{\text{new}}^T \tag{5}
\]

The most of determination of principal components during the modeling prediction progress is using PLSR algorithm. The fitting error will be decreased and the prediction precision will be improved when the number of principal components increases. However, if the number of principal components are too much, usually no
Table 2. Reference measurement of polymer samples in calibration and prediction sets for the transient excitation mode.

|                | Minimum value | Maximum value | Average value | Standard deviation | Sample number | Coefficient of variation |
|----------------|---------------|---------------|---------------|--------------------|---------------|--------------------------|
| Calibration set| 13.67         | 66.00         | 42.53         | 12.55              | 60            | 0.30                     |
| Prediction set | 15.00         | 60.00         | 42.49         | 11.98              | 20            | 0.28                     |

more than 10, the over-fitting of prediction model will happen, and the prediction precision will decrease. The leave-one-out was used to determine the number of principal components.

2.7.3. Model evaluation
All 120 samples were divided into the calibration and the prediction sets. The calibration model was built by the calibration set, and the prediction set was used to test the performance of the model. The specific evaluation indexes include the relationship coefficient R (equation (5)) between the prediction value and the measured value of hardness, standard error (equation (6)), and residual prediction deviation (RPD).

\[
R = \frac{\sum_{i=1}^{N} (y_i - \bar{z}) (\bar{y} - \bar{z})}{\sqrt{\sum_{i=1}^{N} (y_i - \bar{z})^2} \sqrt{\sum_{i=1}^{N} (\bar{y} - \bar{z})^2}}, \quad \bar{z} = \frac{1}{N} \sum_{i=1}^{N} z_i, \quad \bar{y} = \frac{1}{N} \sum_{i=1}^{N} y_i
\]

Where N was the number of samples, \( y \) was the measured hardness of polyurethane, and \( z \) was the prediction hardness. The absolute value of R was closer to 1, which meant that the prediction value had a high correlation to the measured value. The correlation coefficient of calibration set was expressed by Rc; Rp expressed the correlation coefficient of the prediction set. While the relationship coefficient of prediction model is the same after different preprocessing, the root mean square error (RMSE) needs to be calculated to further evaluate the model precision. There were two RMSEs in the study: one was the root mean square error for calibration set (RMSEC), the other one was the root mean square error for prediction set (RMSEP), and the calculation equations as follows:

\[
RMSEC = \sqrt{\frac{1}{m-k-1} \sum_{i=1}^{m} (y_i - \bar{z})^2}
\]

\[
RMSEP = \sqrt{\frac{1}{n-1} \sum_{i=1}^{n} (y_i - \bar{z})^2}
\]

Where \( y_i \) was the measured hardness value of polyurethane, \( z_i \) was the predicted hardness value, \( m \) was the number of calibration set samples, \( n \) was the number of prediction set samples, and \( k \) was the number of principal components. The RPD is the ratio of standard error and root mean square error of calibration set, which is an important index to evaluate the stable and dynamic adaptability of the prediction model. According to the study of Khaledian et al (2017), the prediction of model was not stable and the precision was low when the RPD is less than 1.4. The model could be used when the RPD match 1.4 \( \leq \) RPD \( \leq \) 2 [26]. The model had good stability and high precision when the RPD \( > \) 2. The prediction effect will be further improved with the increase of RPD.

3. Results and discussion

3.1. Hardness prediction by transient modes
The deformation and hardness data of polyurethane was divided into 4 random groups. The models of the calibration set and the prediction set were established by the ratio of 3:1 of groups. Then, 4 groups of modeling results were obtained, according to the value of R and the RMSE to determine the optimum grouping method. For the modeling prediction by the transient mode, table 2 shows the optimum grouping of the hardness of polyurethane. In this way, the range of the hardness of calibration set was 13.67–66.00 Shore A, and the range of prediction set was 15.00–60.00 Shore A, which matched that the requirement of the hardness range of calibration set should be greater than that of the prediction set. There were little differences between the two sets on the parameters of average value, standard error, and coefficient variation (ratio of the standard deviation and average value). These results demonstrated that the data of grouping was reasonable and stable.

After the grouping above, the PLSR algorithm was used to establish models of calibration and prediction sets, the results of modeling with different preprocessing methods were as shown in table 3. The Rc value of prediction set modeling was 0.62 by original laser displacement data. This demonstrated that the correlation existed between the predicted hardness values and the related measured hardness values. However, the RPD was
1.07 < 1.4, so the prediction model was not stable. Therefore, the original data was unsuitable for modeling to predict the hardness of polyurethane, and the preprocesses was needed to process the original signals. As shown in table 3, the relationship coefficient was more than 0.8 and the RPD was more than 1.4 using the preprocessed data modeling by three different preprocessing methods: S-G, SNV, and MSC. These results demonstrated that the original data of transient mode needed to be preprocessed before being used to build the prediction model.

Furthermore, a comparison of the three prepossessing methods was conducted in order to assess which was the most efficient. For MSC, the Rp of prediction set modeling was 0.93. This was more than that of using S-G, however it was the same Rp value to that of SNV. The MSC had the minimum RMSEP. The RPD of MSC was 2.51, which was more than the other two preprocessing methods. Moreover, the Rp and the RMSEP were close to Rc and RMSEC. According to the analysis above, The MSC was the best preprocessing method to process the original data for transient modeling predicting the hardness of polyurethane.

### 3.2. Hardness prediction by creep-recovery mode

Creep-recovery mode was used to predict the hardness of polyurethane with PLSR modeling. According to the optimum modeling effects, the best grouping method of the calibration and prediction sets was determined as shown in table 4. The range of the hardness of calibration set was 13.67–66.00 Shore A, and the range of prediction set was 15.00–60.33 Shore A, which matched that the requirement of the hardness range of calibration set should be greater than that of the prediction set. The differences between the two sets surrounded the parameters of average value, standard error, and coefficient variation which were more than those of transient mode.

As shown in table 5, when the number of principal components was 5, the Rp was 0.38, and the RPD was 0.6, much less than 1.4, which demonstrated that the original data of creep-recovery could not be used to establish the prediction model for evaluating the hardness of polyurethane. After the preprocessing of the S-G algorithm, the Rp raised to 0.61, and the RPD was 1.22, which was still less than 1.4. Therefore, the S-G could not be used as the preprocessing method for modeling. When SNV and MSC were used as the preprocessing methods, the Rc and Rp of both of them were raised to 0.93 and 0.95. Although the RMSEC of SNV was slightly more than that of MSC, the RMSEP of MSC was less than that of SNV. While the RPD of MSC was the most 2.54, and greater than 2, this indicated the prediction model with MSC as the preprocessing method was most stable. Therefore, MSC

### Table 3. Hardness prediction of polymer with different preprocessing methods using transient excitation mode.

| Preprocessing method | Principal components | Calibration set | Prediction set |
|----------------------|----------------------|----------------|----------------|
|                      | Rc                   | RMSEC          | Rp             | RMSEP | RPD |
| Original data        | 6                    | 0.78           | 8.14           | 0.62   | 11.19 | 1.07 |
| Savitzky-galay       | 8                    | 0.93           | 4.48           | 0.82   | 7.13  | 1.68 |
| SNV                  | 5                    | 0.97           | 3.07           | 0.93   | 5.15  | 2.33 |
| MSC                  | 5                    | 0.95           | 3.72           | 0.93   | 4.77  | 2.51 |

The number of smoothing points of S-G was 5.

### Table 4. Reference measurement of polymer samples in calibration and prediction sets for the creep-recovery excitation mode.

| Sample number | Minimum value | Maximum value | Average value | Standard deviation | Coefficient of variation |
|---------------|---------------|---------------|---------------|--------------------|-------------------------|
| Calibration set | 13.67         | 66.00         | 43.31         | 11.89              | 0.27                    |
| Prediction set  | 15.00         | 60.33         | 40.17         | 13.64              | 0.34                    |

### Table 5. Hardness prediction of polymer with different preprocessing methods using the creep-recovery excitation mode.

| Preprocessing method | Principal components | Calibration set | Prediction set |
|----------------------|----------------------|----------------|----------------|
|                      | Rc                   | RMSEC          | Rp             | RMSEP | RPD |
| Original data        | 5                    | 0.41           | 14.81          | 0.38   | 22.89 | 0.60 |
| Savitzky-galay       | 7                    | 0.89           | 5.71           | 0.61   | 11.19 | 1.22 |
| SNV                  | 5                    | 0.95           | 3.54           | 0.93   | 5.42  | 2.52 |
| MSC                  | 5                    | 0.95           | 3.62           | 0.93   | 5.37  | 2.54 |

The number of smoothing points of S-G was 5.
was the optimum preprocessing method when the creep-recovery mode was used to test the hardness of polyurethane. Comparing the performances of transient mode modeling, the Rp of both of them was 0.93, but the RPD of transient mode was 2.51. This was slightly less than the RPD of creep-recovery mode, so the model of the latter was more stable. Therefore, the creep-recovery mode had more advantages in the aspect of model performance. It is important to point out that the transient mode has the characteristic of high-speed detection, and the performance of its prediction model was stable and with high-precision.

### 3.3. Hardness prediction by alternate load mode

The best of grouping was determined comparing different modeling performances of different groupings by alternate load mode. The grouping result of the calibration set and the prediction set is displayed in table 6. It was the same as the grouping result of that of transient mode.

As shown in table 7, when the polyurethane deformation original data of load was used to build the hardness prediction model, the best Rp was 0.25 with 4 principal components. The RMSEC was too great (24.64), the RPD was 0.29, and far less than 1.4. Therefore, the original data could not be used to build the hardness prediction model directly. The Rp raised to 0.74 after the smoothing of S-G, and the RPD was 1.39, slightly less than 1.4. These results indicated that S-G smoothing improved the modeling effects and the hardness prediction model was still unstable. The hardness prediction effect was improved significantly using the data with SNV and MSC preprocessing. Their Rp values increased up to 0.90, the Rc was up to 0.97, and the RPD was greater than 2. This result indicated that both of the two preprocessing methods could be used to preprocess deformation data of alternate load mode to establish the prediction model. Comparing the results of these two preprocessing methods, the RMSE of MSC was less than that of SNV, and the RPD of the former was greater than that of the latter. MSC could be the best preprocessing algorithm for the alternate mode modeling prediction. For this method, the Rp was 0.90, and the RPD was 2.30, which indicated that the model had good stability. Compared with the transient mode and creep-recovery mode, the modeling prediction effects of the alternate mode were inferior.

### 3.4. Hardness prediction by stress-relaxation mode

The grouping parameters of the hardness prediction by stress-relaxation mode is shown in table 8. The range of hardness of calibration set was 13.67–66.00 Shore A, and the range of prediction set was 15.83–60.33 Shore A. There were minimal differences between the two sets on the parameters of average value, standard error, and coefficient variation. These results demonstrated that the data of grouping was reasonable and stable.

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**Table 6.** Reference measurement of polymer samples in calibration and prediction set for the alternate load mode.

|             | Minimum value | Maximum value | Average value | Standard deviation | Sample number | Coefficient of variation |
|-------------|---------------|---------------|---------------|--------------------|---------------|--------------------------|
| Calibration | 13.67         | 66            | 42.54         | 12.55              | 60            | 0.30                     |
| Prediction  | 15            | 60            | 42.49         | 11.98              | 20            | 0.28                     |

**Table 7.** Hardness prediction of polymer with different preprocessing methods using the alternate load mode.

| Preprocessing method | Principal components | Calibration set | Prediction set |
|----------------------|----------------------|-----------------|----------------|
|                      |                      | Rp  | RMSEC   | Rp   | RMSEP | RPD |
| Original data        | 4                    | 0.25 | 16.55   | 0.14 | 24.64 | 0.29 |
| Savitzky-galay       | 8                    | 0.90 | 5.56    | 0.74 | 8.61  | 1.39 |
| SNV                  | 5                    | 0.97 | 2.91    | 0.90 | 5.92  | 2.02 |
| MSC                  | 6                    | 0.97 | 2.83    | 0.90 | 5.21  | 2.30 |

The number of smoothing points of S-G was 5.

**Table 8.** Reference measurement of polymer samples in calibration and prediction set for the stress-relaxation mode.

|             | Minimum value | Maximum value | Average value | Standard deviation | Sample number | Coefficient of variation |
|-------------|---------------|---------------|---------------|--------------------|---------------|--------------------------|
| Calibration | 13.67         | 66.00         | 41.99         | 12.37              | 60            | 0.29                     |
| Calibration | 15.83         | 60.33         | 44.12         | 12.40              | 20            | 0.28                     |
The modeling prediction of the hardness of polyurethane by stress-relaxation is shown in Table 9. Both the original hardness data and the data after preprocessing were used to establish the prediction model with a PLSR algorithm by stress-relaxation mode. The Rp of both of them were over 0.8, and the RPD was greater than 2. The Rp of the original data was 0.88, and the RPD was 2.06 when the principal components were 8. These results demonstrated that the original data of stress-relaxation mode could be used to establish the hardness prediction model directly. The optimum prediction results were obtained by using the preprocessing methods: S-G, SNV, and MSC, with the principal components 4, 3, and 5, respectively. The Rc values were 0.89, 0.94, and 0.95, and the RPD was 2.06, 2.78, and 3.53, respectively. The modeling effects were further validated by comparing those models to the original data. The Rp, Rc, and RPD of MSC values were the highest, and the values of RMSEC and RMSEP were among the lowest, therefore indicating that the model was the most stable.

Compared to the modeling prediction effects on the hardness of polyurethane by the transient mode, the creep-recovery mode, and the alternate load mode with different kinds of preprocessing methods, the stress-recovery was the best mode to build the model to evaluate the hardness of polyurethane. Preprocessing was helpful to improve the signal-to-noise ratio of original deformation data, and the MSC was the best preprocessing method for modeling in the study. Although the stress-relaxation mode with the MSC as the preprocessing method had a good modeling prediction performance (Rc = 0.99, Rp = 0.96, and RPD = 3.54), we still hope to find out the law of participation operation of its different intervals of data.

The regression coefficient of the model was the parameter to evaluate the degree of influence from the independent variable on the dependent variable. For the stress-relaxation mode, 3,001 sampling points of deformation were applied to the regression analysis by PLSR algorithm, and 3,001 regression coefficients were obtained. As shown in Figure 6, three obvious peaks of the regression coefficient curve were found. This indicated that there was supposed to be a better prediction in the time range of these three peaks. Therefore, the whole time of stress-relaxation mode, from 0–60 s and the 3,001 sampling points were divided into four independent subintervals, $\alpha$ (0–2.7 s), $\beta$ (2.7–6.7 s), $\gamma$ (6.7–11.5 s), and $\delta$ (11.5–60 s), and three combined subintervals, $\alpha + \beta$ (0–6.7 s), $\alpha + \beta + \gamma$ (0–11.5 s), and $\alpha + \beta + \gamma + \delta$ (0–60 s). The subintervals were used to establish the polyurethane hardness prediction model using stress-relaxation mode with MSC. The results of the modeling are shown in Table 10.

| Preprocessing method | Principal Components | Cal. set Rc | Cal. set RMSEC | Pred. set Rp | Pred. set RMSEP | Pred. set RPD |
|----------------------|----------------------|-------------|----------------|--------------|----------------|-------------|
| Original data        | 3                    | 0.92        | 4.84           | 0.88         | 6.09           | 2.04        |
| Savitzky-galay       | 4                    | 0.92        | 4.62           | 0.89         | 6.01           | 2.06        |
| SNV                  | 3                    | 0.95        | 3.91           | 0.94         | 4.46           | 2.78        |
| MSC                  | 9                    | 0.99        | 1.79           | 0.96         | 3.50           | 3.54        |

The number of smoothing points of S-G was 5.

![Figure 6. Regression coefficients of stress relaxation mode prediction.](image)
When the independent subintervals were used to build the prediction model to evaluate the hardness of polyurethane, the Rp of the four subintervals were 0.47, 0.46, 0.45, 0.55, respectively. The RPD of them were 0.97, 0.89, 0.89, and 1.20, and all of them less than 1.4. These results demonstrated that the independent subintervals could be used for modeling evaluation on the hardness of polyurethane by the stress-relaxation mode. For the combined subintervals, the Rp of $\alpha + \beta$ was raised to 0.91 and its RPD was raised to 1.98 $> 1.4$.

The results were much better than the independent subintervals, and these results indicated that good modeling effects could be obtained when the processing of the stress-relaxation mode lasts to 6.7 s. When the processing was executed to 11.5 s, the Rp of the combined subinterval $\alpha + \beta + \gamma$ was 0.94, and the RPD was 2.79. Compared with the $\alpha + \beta$, the hardness prediction effects were improved by $\alpha + \beta + \gamma$ modeling. As the time of relaxation continued to last to 60 s, the results of modeling continued to get better. The Rp was 0.96 and the RPD was 3.53. The correlation between hardness measured values and predicted values in the calibration set and prediction set is shown in figure 7.

According to the modeling prediction results of independent subintervals and combined subintervals, it can be inferred that the prediction precision of the model would be further enhanced with increased increments of relaxation time, and the stability of the model would be improved as well. Therefore, the stress-relaxation mode could be regarded as the high-precision prediction mode to analyze the hardness of polyurethane in the study.

### 4. Conclusion

A hardness prediction method of materials was proposed based on the multi-mode excitation by the CLAFD system. The excitations of the transient mode, creep-recovery mode, alternate load mode, and stress-relaxation mode were carried out by well-controlled air-force, and the corresponding responses were obtained by a laser displacement sensor. Different preprocessing methods were used to enhance the signal-to-noise-ratio on the original deformation data of polyurethane in the study to improve the effects of the modeling prediction by PLSR. The main conclusions are as follows: (1) Comparing the original data to the data with different kinds of preprocessing methods that were used to establish the hardness prediction model, it could improve the precision

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**Table 10.** Hardness prediction of polymer with different preprocessing methods using the stress excitation signal.

| Stage | Principal Components | Calibration Subset | Prediction Subset |
|-------|----------------------|--------------------|-------------------|
|       |                      | Rc  | RMSEC | Rp   | RMSEP | RPD |
| $\alpha$ | 5-1G                | 0.54 | 10.67 | 0.47 | 12.82 | 0.97 |
| $\beta$  | 5-2G                | 0.71 | 8.54  | 0.46 | 13.96 | 0.89 |
| $\gamma$ | 8-2G                | 0.57 | 9.96  | 0.45 | 13.90 | 0.89 |
| $\delta$ | 5-3G                | 0.81 | 7.30  | 0.55 | 10.35 | 1.20 |
| $\alpha + \beta$ | 5-4G            | 0.93 | 4.39  | 0.91 | 6.27  | 1.98 |
| $\alpha + \beta + \gamma$ | 6-4G     | 0.96 | 3.27  | 0.94 | 4.46  | 2.79 |
| $\alpha + \beta + \gamma + \delta$ | 9-1G    | 0.99 | 1.79  | 0.96 | 3.50  | 3.54 |

**Figure 7.** Correlation between hardness measured values and predicted values in the calibration set and prediction set. (a) was prediction results of calibration set, (b) was prediction results of prediction set.
and stability of modeling predictions. (2) Of the three preprocessing methods, MSC was the best among them. (3) The Rc was over 0.90, and the RPD was greater than 2 of the modeling predictions using the data with the MSC preprocess for all four excitation modes. Therefore, all of them could be adopted for the prediction of hardness. (4) The excitation time of the transient mode was only 1 s. Meanwhile, the Rc of transient mode was 0.93 and the RPD was 2.51. Therefore, the mode could be considered a high-speed prediction. (5) Independent subintervals of the stress-relaxation mode could not be used to establish a model for the hardness prediction of polyurethane, although there were some independent intervals with high regression coefficients. The combined subintervals were much better than the independent subintervals in the hardness prediction. As the time of relaxation continued to last to 60 s, the results of modeling continued to improve; the Rc was up to 0.99, Rp was up to 0.96, and the RPD was up to 3.53. It can be deduced that the precision of the model would be further enhanced with increased increments of relaxation time, and the stability of the model would be improved as well. Therefore, the stress-relaxation mode could be regarded as the high-precision prediction mode on the hardness evaluation of polyurethane in the study.

The precision and speed of hardness prediction would be enhanced with the sustainable development of the CLAFD system and the optimization of preprocessing and modeling algorithms. Although the polyurethane was chosen as the research subject, the multi-mode excitation prediction method would be suitable for numerous material hardness detections, such as chemical materials, biomaterials, etc. Although, as an important property, the hardness was predicted in the study, the prediction of the other properties of polyurethane could be explored, such as viscosity, elasticity, etc.

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Conflicts of interest

The research team has no conflicts to declare.

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