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Acoustic emission studies on the lubrication of a fibrous collagen material-leather

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For thousands of years, collagen materials, such as leather, have been among the most dominant natural fibrous materials used by humans. Fatliquoring is one of the critical steps in the leather-making process, wherein oil or a lubricant is added to the leather to prevent the leather fibers from sticking together, thereby providing sufficient pliability to the leather. We have examined the feasibility of using the acoustic emission (AE) technique to characterize the degree of lubrication of leather produced with various fatliquor concentrations. In a tensile test, an acoustic transducer was contacted with the leather samples to collect their AE quantities and properties. The samples lubricated with a fatliquor concentration less than 10% showed twin peaks on the plot of hits rate versus time. This implied that a non-uniform fracture occurred in a leather structure that was not sufficiently lubricated. In contrast, a sufficiently lubricated leather structure showed a steady increase in hits rate with time until it fractured. Traditional stress-strain tests did not reflect these behaviors. Observations also showed a direct correlation between the cumulative hits and fatliquor concentration. The results of this work may provide a route to identify an adequate degree of lubrication in the leather.

1. Introduction

For thousands of years, collagen materials, such as leather, have been among the most dominant natural fibrous materials used by humans, especially for clothing, upholstery, and shoes. Leather is economically significant because it is the major by-product derived from the meat industry. Fatliquoring is an oil-addition process by which the leather fibers are lubricated so that after drying they will be capable of slipping over one another. The fatliquored leather therefore attains a greater softness and flexibility than is imparted by the tannages. We have previously reported its pronounced effect on drying rate and leather properties [1]. Our measurements showed that the drying rate decreases as the fatliquor concentration increases by a factor of $(1 - f)^{1/2}$, where f is the fatliquor concentration fraction. Observations also showed that the tensile strength, elongation and toughness all increase steadily with increased fatliquor concentration, whereas the Young's modulus decreased. We also demonstrated a correlation between the elongation and Young's modulus for fatliquored leather.

The fatliquoring process must be done properly to ensure that the leather's fibrous structure is adequately lubricated in order to prevent its fibers from sticking together during the leather drying process. Currently the method described in this report is the only method to measure the degree of lubrication. Lubrication affects the resistance of fiber movements and deformations, and from previous studies we learned that acoustic emission measurements are very sensitive to these changes in resistance [2]. Therefore, we recently investigated the feasibility of using acoustic emission technology to measure the degree of lubrication associated with the fatliquoring process.

Sound waves are longitudinal mechanical waves. Transient sound waves can be generated in the region of a material that experiences abrupt changes in strain. This phenomenon is known in materials science as acoustic emission (AE) and is generally detected by means of ultrasonic transducers coupled to the material. Typical examples of events, which produce acoustic emission, are fiber movements and breakages, interfacial bond failure in fiber composite materials, growth of microcracks, and delamination of thin films. The operating frequencies of transducers used in AE measurements are usually in the range of 50–1000 kHz, which is well above the audible sound frequency. This permits all ambient noise to be filtered out, leaving only the frequencies of interest. For some years now in our research center, we have recognized acoustic emission (AE) as a useful method for characterizing leather properties [3]. We have studied the sounds emitted by leather when it is stretched (in a tensile test) and examined the

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relationship between tensile strength and AE quantities [4]. The most important AE data that we studied included the amplitude distribution of hits, rate of hits and acoustic energy emitted from leather during stress-strain tests. Observations showed an excellent correlation between the tensile strength of leather and the corresponding acoustic cumulative energy at break, read from an acoustic-emission analyzer [4]. Moreover, a linear relationship was discovered between the acoustic cumulative energy at break and the initial acoustic cumulative energy, when the leather was elongated by ten percent of its original length. More importantly, a correlation was observed between the initial acoustic cumulative energy and the tensile strength of leather [4]. The implication of these phenomena is that the tensile strength of leather may be predicted without breaking the leather by measuring the initial cumulative acoustic energy. The long-range goal of this ongoing research project is the production of an AE tester, providing the leather industry with a nondestructive way in which to monitor the quality of leather at each intermediate leather-making stage. As a result, tanners would be able to adjust their leather-making processes accordingly to yield high quality leather.

Besides tensile strength, one of the other important mechanical properties required for leather products, particularly those used for upholstery, is the ability to withstand tearing. Recently, an effort was made to use the acoustic-emission method to gain insight into the reason for tear failure [5]. In a tongue-tear test, chrome-tanned leather samples were contacted with an acoustic sensor to collect various acoustic quantities. Measurements showed that the samples stronger in tear strength gave a significantly lower acoustic count. In contrast, the samples with poor tear strength generated more sound pulses, i.e., more acoustic counts [5]. This is contrary to results from tensile failure tests, where the higher strength leather always produces more total acoustic counts. Observations also showed that harsh drying conditions or a thin corium could lead to a brittle structure, which consequently yields poor tear resistance [6]. In an AE test, this can be reflected in a high AE hit-rate because of frequent fiber breaking and friction associated with the brittle structure. More recently, we applied AE technology to measure the degree of opening-up of the leather structure [2]. This research project was in response to the urgent need for an effective means to identify the proper liming conditions that produced a sufficient degree of opening-up. We have demonstrated that a history plot of acoustic emission counts could detect a change in the degree of opening up of the fiber structure associated with an increase in liming time. AE tests revealed that leather made with a shorter liming time (i.e., less than the standard 24 hours) showed a relatively smooth history plot, whereas leather with a liming time greater than the standard 24 hours produced a pronounced saw-shaped pattern, and the longer the liming time, the more erratic the pattern becomes. This behavior mirrors the change in the extent of opening-up of the fibrous structure engendered by lime action. The results of this work have provided a route to monitor the degree of opening up of leather.

We are now reporting another use for AE technology in associating it with the lubrication of leather. As a natural fibrous material, leather emits sound waves engendered by a sudden stress accompanied with any significant fiber movement or fiber deformation including breakage. From analysis of the hits, frequency and energy associated with emitted sound waves during the tensile tests of leather treated with various amounts of fatliquor, one may gain a correlation between acoustic emission quantities and the degree of lubrication. All the AE data reported above were measured with transducers that resonate within the ultrasonic range (50–200 kHz), thereby avoiding the problems of noise produced by the testing machine or environment. As mentioned before, in materials science, the term "AE" refers only to the ultrasonic range. Nevertheless, others also reported sound emissions in the audible range (from 1–20 kHz) to evaluate various materials properties, particularly in evaluating the "handle" of a material [7–9].

2. AE principles

Leather is a fibrous collagen material. The deformation of leather (as leather is squeezed, torn or stretched) caused by an external force is accompanied by a rapid movement, relocation, or breaking of structural elements such as fibrils, fibers and/or fiber bundles. As a result, sound waves are produced that can be detected by an acoustic transducer and converted into electronic signals. This basic phenomenon may be defined as an acoustic emission event, which is translated by an AE analyzer as a "hit" [10]. Fig. 1 shows a waveform (a burst AE signal detected by the transducer) and the commonly used parameters for AE. When the AE transducer senses a signal over a certain level (i.e., the threshold), an AE event is captured. The amplitude of the event is defined at the signal peak. A single acoustic emission event (hit) may consist of many emission counts, which are the number of times a signal from the transducer crosses a preset amplitude threshold, as illustrated in Fig. 1. The number of times the signal rises and crosses the threshold is the count of an AE event. The time period between the rising edge of the first count and the falling edge of the last count is the duration of the AE event. The energy of the hits is commonly reported as the average area under envelope of the waveform as shown in Fig. 1, so the hit amplitudes and hit durations determine the energy [11].

Fig. 2 presents our AE system setup with a tensile tester, in which the leather sample is gripped between

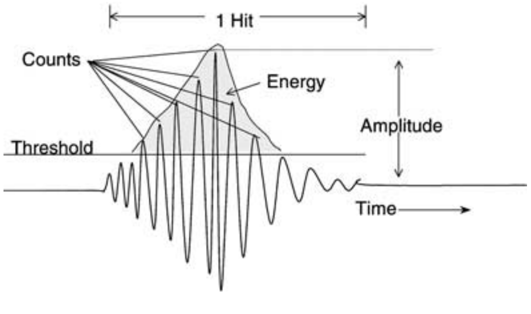

Figure 1 Acoustic waveform.

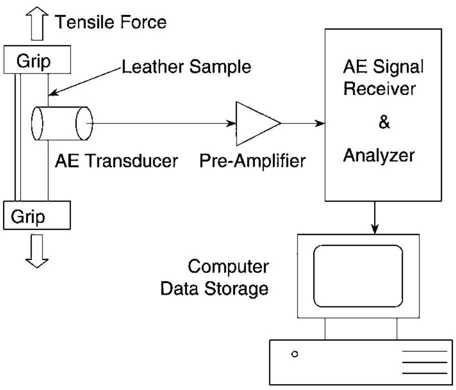

Figure 2 Acoustic emission instrumentation.

two jaws. The sample is subjected to a tensile force and slowly stretched at a constant strain rate. Due to this tensile stretching, the leather sample emits sound waves, which are detected by an acoustic transducer. The AE transducers are generally very sensitive piezoelectric sensors. Since the AE signals are often weak, a preamplifier is connected right after the AE transducer to minimize noise interference and prevent signal loss. The main amplifier amplifies the signals before being sent to the signal analyzer. After that, the AE quantities are stored in a computer for further analysis. Each acoustic hit generates a wavetrain from the transducer consisting of a number of oscillations (waves), so-called "acoustic counts." The hits with high amplitudes always produce high numbers of counts. For practical purposes, a simple equation can be used to relate signal amplitudes, hits and counts [11].

$$\sum N(t) = \left(\sum H(t)\right)\nu\tau/\beta \tag{1}$$

Where $\Sigma N(t) =$ cumulative counts at time t, $\Sigma H(t) =$ cumulative hits at time t, $\nu =$ resonant frequency of sensor, $\tau =$ duration of hits, and, $\beta =$ the amplitude distribution slope parameter.

3. Experimental

3.1. Materials

Bovine leather, chromed-tanned from steer hides with a thickness between 2.4–2.6 mm, were obtained from Prime Tanning Co. (St. Joseph, MO). Each leather was split into two sides along the backbone line. The sides were treated with various concentrations (w/w%) of fatliquor solutions composed of sulfated and sulfonated oil and petroleum lubricating oil (trade name: ATLASOL CAM) from Atlas Refinery, Inc. (Newark, NJ). This type of fatliquor has been widely used for lubricating chrome-tanned leather. The sides were left to drain overnight and were set out before vacuum drying. A Cartigliano vacuum drying machine was used to dry the leather at 60°C for 10 minutes. The vacuum pressure was maintained at 0.8 bar for the drying experiments, which equals 20 kPa absolute pressure. It is a typical pressure used in a vacuum drying operation. Vacuum-dried sides were subsequently toggled on a metal screen and air-dried at a lower temperature (35°C) for 30 minutes. The sides were conditioned by rewetting the corium side, rolling them up and placing them in plastic bags to equilibrate overnight. The sides were then pressed through a Molissa staking machine at a rate of 1.6 meters/min. Finally the sides were equilibrated in a conditioning room at 23°C with 50 percent RH for 1 week before physical property testing.

3.2. AE instruments

We simultaneously performed tensile tests and AE data collections to study their relationships. To obtain acoustic-emission data, a small piezoelectric transducer resonating at 150 kHz (Model R15, Physical Acoustics Corp., Princeton, NJ), 10 mm in diameter and weighing 20 g, coated with a film of petroleum grease for more efficient acoustic coupling, was clipped against the leather sample. Electrical signals emanating from this transducer when the Instron stretched the leather samples were processed with a Model 1220A preamplifier and a LOCAN-AT Model 3140 acoustic emission analyzer (Physical Acoustics Corp.). As illustrated in Fig. 1, each acoustic hit from an acoustic event in the sample causes a damped oscillation to be emitted by the transducer. The analyzer records the duration of each oscillation hit, its amplitude, and its energy. Only hits giving maximum amplitudes greater than 35 dB (threshold) from the transducer were counted.

3.3. Methods

Dumbbell-shaped leather samples were cut from the standard test area as described in ASTM D2813-91 with the long dimension parallel to the backbone. Test samples were stored in a conditioned room at 23°C and 50% RH before testing according to ASTM standard method D1610-96. The moisture content of samples was determined to be 15 ± 1 percent moisture by a leather moisture meter (Delmhorst Instrument Co., Towaco, NJ). Tensile properties were measured with a gauge length (distance between two jaws) of 67 mm. An upgraded Instron mechanical property tester, model 1122, and Testworks 3.1 data acquisition software (MTS Systems Corp., Minneapolis, MN) were used throughout this work. The cross-head speed was reduced from the standard 254 mm/min (10″/min) to 50 mm/min in order to provide sufficient time for AE data collection.

The fatliquor content in the leather after fatliquoring was estimated by measuring the amount of hexane-soluble matter existing in the leather samples. It was determined on a moisture free basis according to ASTM method D3495-00, "Standard Test Method for Hexane Extraction of Leather."

Scanning electron microscopic examinations were conducted on the cross section of leather samples to examine the degree of opening up in the fibrous structure. Samples were mounted on aluminum specimen stubs using colloidal silver adhesive (Electron Microscopy Sciences, Ft. Washington, PA) and sputter-coated with a thin layer of gold. Images were collected using a Model JSM 840A scanning electron microscope (JEOL USA, Peabody, MA), integrated with a model Imix 1 digital image workstation (Princeton Gamma-Tech, Princeton, NJ), and operated in the secondary-electron imaging mode.

4. Results and discussion

4.1. Acoustic hits rate

Plotting the rate of hits as a function of time is a basic way to graph AE activities. Fig. 3 displays the chronological course of the test, demonstrating the hits rate vs. time profiles during the tensile testing of leather made with two significantly different fatliquor concentrations. Fig. 3a shows that the 2% fatliquored sample produces twin peaks in the hits rate vs. time curve. This behavior was also observed in the 5% fatliquored sample. The first peak signifies a partially fractured leather sample; with some unbroken fiber bundles still holding the leather together. The second peak shown on the figure is probably due to the subsequent breakage of those remaining fiber bundles. In contrast, the 10% fatliquored sample as shown in Fig. 3b yields only one major peak. This sample emits very little sound for the first thirty-seconds stretched, followed by a steep increase in the hits rate to a peak point at around 60 seconds, and then a sudden decrease as the leather is totally fractured. The same behavior was also observed in the 15% and 18% fatliquored samples. Multiple-peaks as shown in Fig. 3a imply the existence of non-uniform stress sharing and a premature fracture in the leather structure. In contrast, a single peak as shown in Fig. 3b indicates that the leather structure uniformly shares the tensile stress without premature breakage or fracture as shown in the 10% fatliquored sample. This may be attributed to a better lubricated fiber structure, in that the fibers are more free to move and to slip over each other, thereby lining up in the direction of stretching and consequently sustaining higher tensile stress. Fig. 4 presents the stress-strain curves from traditional tensile tests. Although the 10% fatliquored sample (Fig. 4b) showed a better tensile strength and breaking elongation than the 2% fatliquored sample (Fig. 4a), the tensile test is not sensitive enough to reflect the non-even fracture that occurred in the 2% fatliquored sample as mentioned before. Lubrication also affects tear strength. In a tongue-tear test, the difference in tear strength between the 10% and 2% fatliquored samples was even more significant (80 N/mm vs. 55 N/mm, respectively).

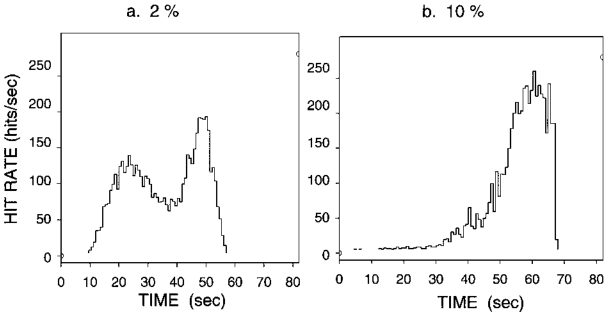

Figure 3 Acoustic hits history for 2% and 10% fatliquored leather.

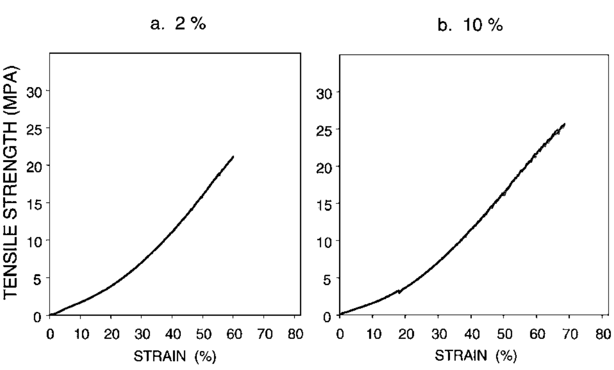

Figure 4 Stress-strain curves for 2% and 10% fatliquored leather.

A fibrous structure is a necessity for the energetic acoustic activities demonstrated in Fig 3. In other words, the acoustic hits profiles demonstrated previously are a unique phenomenon for a fibrous structure. Leather consists of collagen fibers with a very complex structure, a network of interwoven fiber bundles with large spaces unevenly distributed among them. This may be elucidated better by SEM photos. Fig. 5 shows a typical cross-sectional view of leather treated with 10% fatliquor, which has a distinctively fibrous structure. It has been reported that fiber bundles (20–200 μm) of leather are comprised of very fine element fibers (10 μm), which can further be divided into even finer fibrils (0.01–0.5 μm) [12, 13]. Fiber movements engender the mechanical waves when a fibrous material such as leather is stressed.

In contrast, a non-fibrous structure such as a plastic film (for example, polyester) will not be capable of emitting sound when it is stretched until it is totally fractured, as shown in Fig. 6a. For the same reason, a raw hide (Fig. 6b) does not produce sound either, until the final moment of fracture.

4.2. Acoustic frequency patterns

We also examined the acoustic frequency response to the change of fatliquor concentration. Fig. 7 illustrates the change of frequency as the tensile test proceeds. Fig. 7a demonstrates that the transducer emits a high pitch sound twice from the 2% fatliquored sample, whereas the 10% fatliquored sample, as shown in Fig. 7b, transducer only produces a high pitch sound once throughout the tensile test. The high pitch sound is ascribable to the fracture of leather. The same arguments discussed in the previous section can also be applied here. The twin-peaks in the sound frequency reflect poorly lubricated leather that fractures non-uniformly. In contrast, well-lubricated leather will emit a single peak because almost all the fiber bundles fracture simultaneously.

4.3. Acoustic count rate

One acoustic hit can consist of many counts, depending on the intensity of the wave train. The stronger or more energetic the wave train, the higher the number of counts will be produced. Therefore, the number of counts is a function of the magnitude of acoustic energy. As shown in Fig. 8, there is a linear relationship observed between the acoustic counts and the energy. The correlation coefficient (r) between these two AE quantities is 0.97.

Fig. 9 illustrates the AE count rate-history produced during tensile stretch. It clearly demonstrates that the sample with a lower offer of fatliquor (2%, Fig. 9a) shows an earlier set-off in count history than the 10% sample (Fig. 9b). The reason behind this behavior is

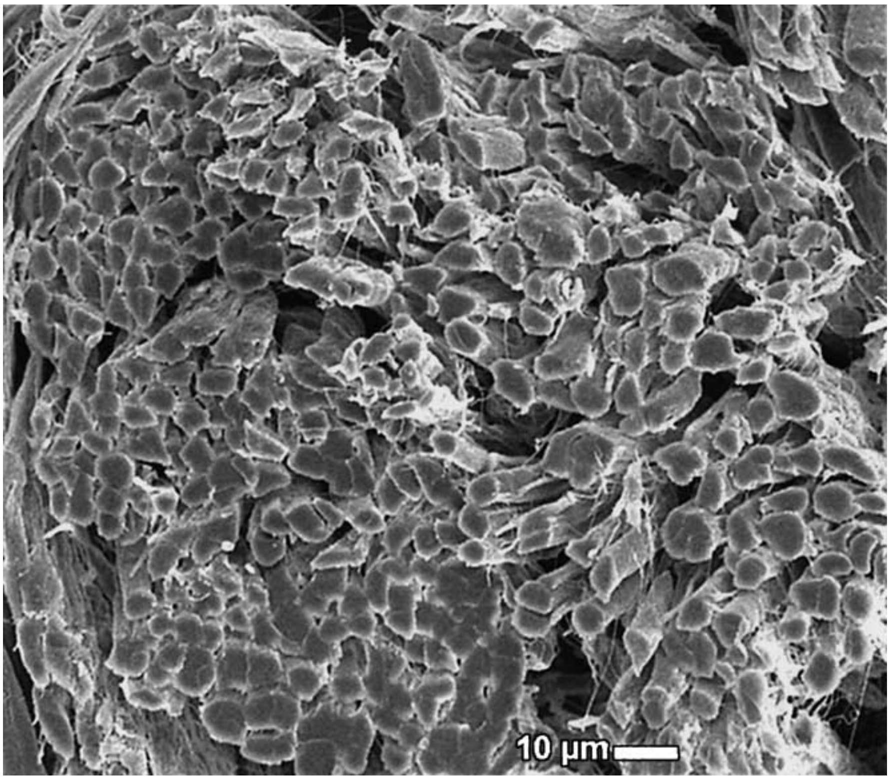

Figure 5 A cross-sectional view of a typical leather sample.

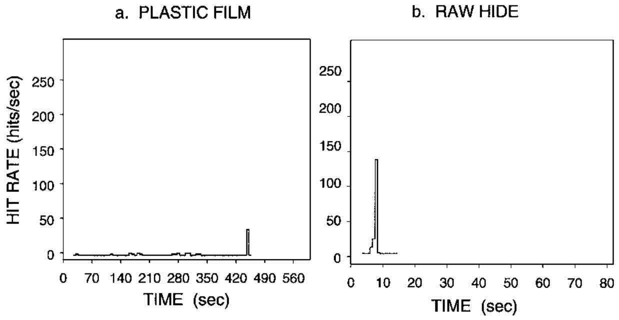

Figure 6 Acoustic hits history for plastic film and raw hide.

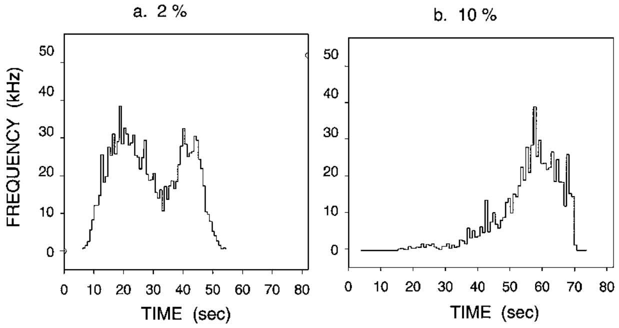

Figure 7 History plot of frequency.

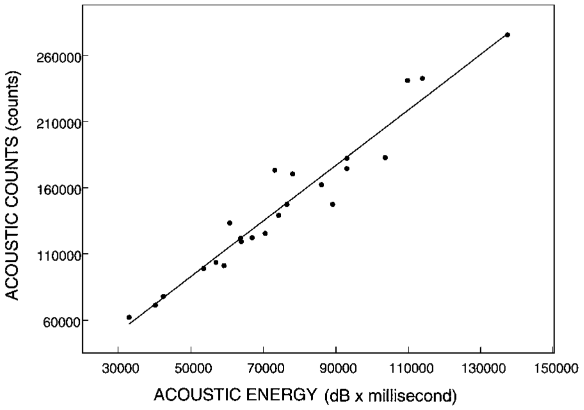

Figure 8 The linear relationship between the acoustic counts and the acoustic energy.

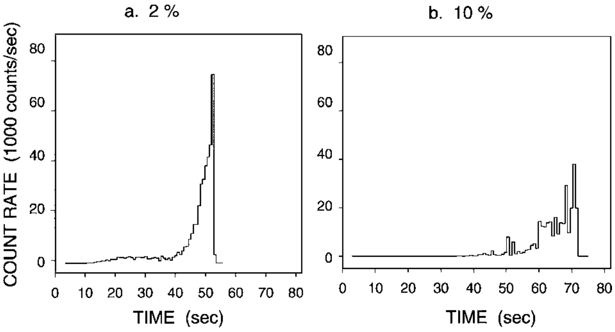

Figure 9 Acoustic counts history for 2% and 10% fatliquored leather.

that the fibers in the poorly lubricated leather possess a higher friction coefficient, and consequently produce a higher initial stress. As a result, the fibers start to emit sound very early as the leather is stretched. Fig. 10 presents a “blown-up” picture of Fig. 9, showing the count rate history for the first 10 seconds. It clearly demonstrates the 2% fatliquored sample has an earlier AE activity than that of the 10% fatliquored sample.

4.4. Cumulative hits

As illustrated in the previous figures, a rate plot of AE hits or counts during the tensile test can highlight the changes in AE activity because of the variation in

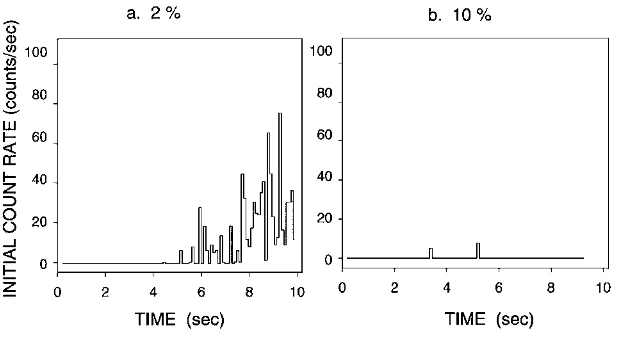

Figure 10 Initial acoustic count rate.

fatliquor concentration. Observations so far have shown that the hit-rate history patterns correlate well with the degree of lubrication of the fibrous structure. However, a quantitative correlation between an acoustic quantity and the degree of lubrication would be more desirable. We then investigated the cumulative hits as a function of fatliquor concentration. Fig. 11 shows the typical plots of the cumulative AE hits, i.e., the summation of hits measured since the start of the tensile test up to the fracture of the test samples, as a function of time. These curves propagate very similarly to those of stress-strain curves. The cumulative hits slowly increase with stretching time until the leather is fractured. Fig. 11 provides a convenient format for reading off a total emission quantity. The total hits of a leather sample are strongly associated with the flexibility of leather. The more flexible the leather, the less amount of acoustic hits are emitted in a tensile test. Fig. 12 plots the total hits (the cumulative hits read off as the sample fractured) as a function of fatliquor concentration. The total hits decrease drastically as the fatliquor concentration increases.

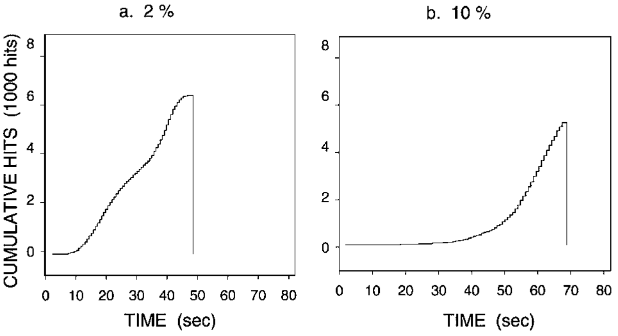

Figure 11 Cumulative plot of acoustic hits to obtain the total acoustic hits.

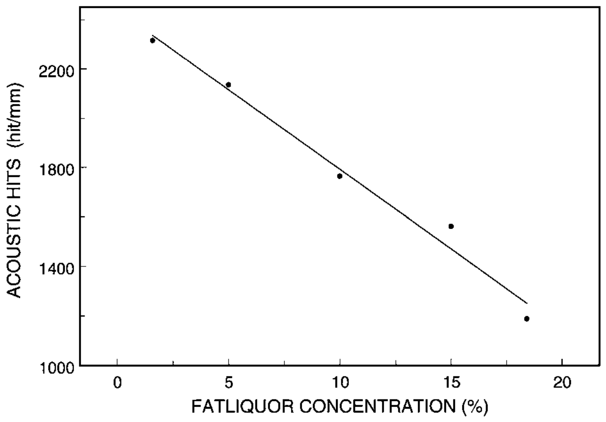

Figure 12 The linear relationship between the total acoustic hits and fatliquor concentration.

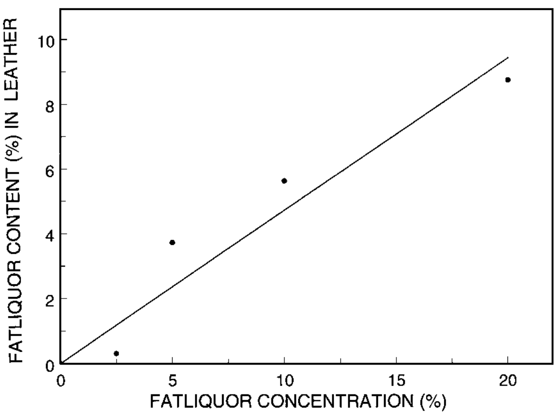

Figure 13 The linear relationship between fatliquor concentration and fatliquor content.

The action of fatliquoring as mentioned previously is to lubricate the fibrous structure. The higher the concentration of fatliquor, the more the amount of fatliquor or lubricant is absorbed by the leather, as demonstrated in Fig. 13. Therefore, the total acoustic hits are decreased with an increase in the fatliquor concentration applied in the fatliquoring process as demonstrated in Fig. 12.

5. Conclusions

The proper lubrication of a fibrous structure such as leather is essential to its mechanical performance, and consequently to its quality. This investigation has demonstrated the usefulness of AE technology to determine the adequate lubrication used in the fatliquoring process. The cumulative hits showed the most direct correlation to the degree of lubrication, whereas the plot of hits rate vs. time pictures the direct link between the un-even fracture of leather and its poorly lubricated structure. This provides an insight into the reason for the increased strength of fatliquored leather. The results of this work may provide a route to monitor the degree of lubrication of leather, which was previously difficult to measure.

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