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## Bubbles in Wet, Gummed Wine Labels\*

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Abstract. It is shown that bubbling on wine bottle labels is due to absorption of water from the glue, with subsequent hygroscopic expansion. Contrary to popular belief, most of the glue's water must be lost to the atmosphere rather than to the paper. A simple lubrication model is developed for spreading glue piles in the pressure chamber of the labelling machine. This model predicts a maximum rate for application of labels. Buckling theory shows that the current arrangement of periodic glue strips can indeed accommodate paper expansion. This project provides interesting applications of various areas of undergraduate mathematics, such as trigonometry, Maclaurin series, dimensional analysis, and fluid mechanics. It illustrates that simple mathematical modelling may provide insight into a complicated real-world problem.

Key words. paper expansion, water absorption, buckling, glue spreading

AMS subject classifications. 76D, 76S, 73K

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1. Introduction. Immediately after they emerge from the labelling machine, wine labels normally appear to be adhering smoothly to the glass bottles, with a good, even coverage of glue. However, in a significant number of cases, bubbles appear after 10 to 15 minutes. Although there is much variation, bubbles are typically 5 mm wide, a few millimeters in height and 15 mm in length. Usually they are at least 2 cm from a label edge, but otherwise have no obvious pattern of distribution. This disfigurement is at odds with the desire of wine producers to present a high-quality product (see Figure 1.1).

Some shipments of wine have been returned by customers simply because of the imperfect labels. Since label bubbling is prominent in around 1 in 20 labelled batches,

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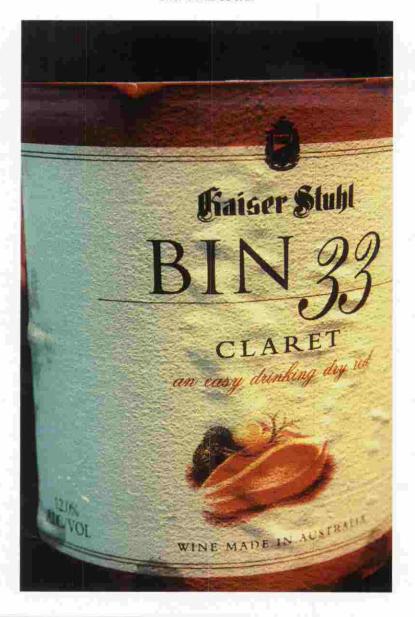


Fig. 1.1

there are important economic implications for the industry. Hence, Herbert Hruby, of Southcorp Wines Pty. Ltd., presented this problem at the introductory plenary session of the 1996 Australian Mathematics-in-Industry Study Group (MISG) [1], attended by approximately 160 mathematicians and industrial representatives. A diverse collection of eight problems was presented by representatives of various industries. Afterwards, each delegate was free to attend groupwork sessions for a small number of problems, with each session assigned to a separate room. These sessions met regularly over the next five days. The problem of bubbles in wine labels attracted about a dozen devoted participants and almost the same number of part-timers, who chose to work on more than one problem.

As will become evident from the following analysis, this project provides a useful source of education in mathematical modelling. First, it shows that complicated real-world problems may be profitably analyzed using simple mathematical models. The mathematical tools need not be sophisticated in order to yield insight. This project provides interesting applications of various areas of undergraduate mathematics, such as trigonometry, Maclaurin series, dimensional analysis, and fluid mechanics. However, the range of mathematical tools used here is broader than that used for most laundered textbook problems. Experience in this kind of problem will give students some confidence in confronting real-world problems and will also illustrate the interconnectedness, through common applications, of various branches of mathematics.

2. Possible Contributing Factors. What is wrong with the 5% of label batches that misbehave? No one knows for certain. The group had to piece together existing empirical and anecdotal evidence in order to postulate possible mechanisms of label crinkling. It was hoped that these mechanisms could be described initially by very simple, physically based mathematical models that are capable of making testable predictions. Simple investigative mathematical modelling is an efficient, inexpensive procedure that can rule out dead ends before costly experiments are set up.

It is widely believed that water uptake and hygroexpansion of the paper is involved. Although bubbles may form 15 minutes after application, it is observed that bubbles may expand or contract over periods of several days in response to changes in atmospheric humidity. Second, glue temperature and glue wetness at the time of application are known, by experience, to be important factors. Anecdotal evidence suggests that label bubbling tends to be worse on white wines, which are bottled at lower temperatures than red wines. The project group surmised that the most relevant temperature-dependent property would be glue viscosity, which might influence the spread of glue strips on the bottle.

Adjustable parameters include labelling rate, pressure applied to labels, water content of glue, and paper thickness and density. A rational mathematical model is desirable if the relative importance of these parameters is to be understood. In this case, various members of the group agreed, after days of discussion, to make calculations on glue spreading, drying, hygroexpansion, and paper buckling.

3. Spreading of Glue Piles. The labelling glues are alkaline caseinate solutions, formed from casein protein that occurs naturally in milk. According to [5], wet casein glues are near-Newtonian, having little variation in viscosity, at low concentrations, but they become non-Newtonian at high concentrations. In Southcorp's Nuriootpa bottling plant, and at most other plants, a pallet applies glue to a label in regularly spaced strips, approximately 0.6 mm in width and 1.5 mm apart. The label is then drawn away by mechanical finger grips, leaving mounds of glue on the paper, as in Figure 3.1.

The label is then placed on the bottle and pressure is applied to its face. Sponges and brushes are used to do this, and finally, as an added measure, the bottles are placed in a pressure chamber in which a curved rigid wall is pressed against the label. Wrinkle-free, labelled bottles do emerge; bubbles appear later.

As we shall verify in subsequent calculations, the glue mounds are rapidly flattened during the initial stages of compression. Therefore, we assume that almost immediately after the glue mounds on the label make contact with the glass bottle, they are flattened into an almost rectangular shape of width  $2L_0$  and height  $h_0$ , determined by the initial cross-sectional area  $A = 2h_0L_0$  of the mound. Subsequently, each glue strip is subjected to a force F (assumed to be fixed) by the pressure devices,

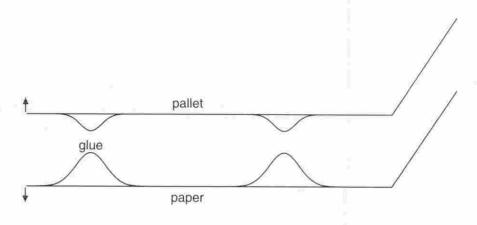


Fig. 3.1 Application of glue.

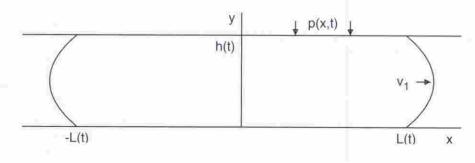


Fig. 3.2 Glue pile in pressure chamber.

causing it to spread, as in Figure 3.2. We are interested in the effectiveness of these devices during the squeezing stage of the process.

From standard lubrication theory [8], the Navier-Stokes equations reduce to

(3.1) 
$$\frac{\partial p(x,t)}{\partial x} = \mu \frac{\partial^2 v_1(x,y,t)}{\partial y^2}$$

with pressure p, dynamic viscosity  $\mu$ , time t, Cartesian coordinates (x, y), and x-component of fluid velocity  $v_1$ .

Consider the material flowing into and out of a thin strip of width  $\Delta x$ , depth Z, and height h(t). The rate of change of glue volume in the region

(3.2) 
$$(x, x + \Delta x) \times (0, h(t)) \times (0, Z)$$

is  $Z\Delta x \ dh/dt$ , which must equal the rate of material flowing in minus the rate of material flowing out;

(3.3) 
$$Z\Delta x \frac{dh}{dt} = Z \int_{0}^{h(t)} v_1(x, y, t) dy - Z \int_{0}^{h(t)} v_1(x + \Delta x, y, t) dy.$$

Dividing through by  $Z\Delta x$  and taking the limit as  $\Delta x \to 0$ , we obtain a simple form of the equation of continuity [8], a local conservation law

$$\frac{dh}{dt} + \frac{\partial Q}{\partial x} = 0,$$

where

$$Q(x,t) = \int_{0}^{h(t)} v_1 dy.$$

Now, by twice integrating throughout (3.1) and respecting the conditions  $v_1 = 0$  at y = 0, h we obtain

$$(3.6) \quad v_1(x, y, t) = \frac{+1}{2\mu} \frac{\partial p}{\partial x} y(y - h(t)).$$

Consequently,

(3.7) 
$$Q(x,t) = \frac{-1}{12\mu} \frac{\partial p}{\partial x} h^3(t).$$

Then (3.4) implies

$$\frac{\partial^2 p}{\partial x^2} = \frac{12\mu}{h^3} \frac{dh}{dt}.$$

Integrating throughout (3.8) and noting p=0 when  $x=_{-}^{+}L(t)$ , we arrive at

$$p = \frac{-6\mu}{h^3} \frac{dh}{dt} (L^2 - x^2).$$

Now the total force applied to a glue strip is

(3.10) 
$$F = +Z \int_{-L}^{L} p \, dx = \frac{-8\mu Z}{h^3} \frac{dh}{dt} L^3,$$

where Z is the length of a label. Assuming  $h(t)L(t) \approx A/2$ , where A is the constant cross-sectional area of the glue pile, we have the ordinary differential equation

(3.11) 
$$\frac{1}{h^6} \frac{dh}{dt} = \frac{-F}{\mu Z A^3}$$

with solution

$$(3.12) H = (1+5T)^{-1/5},$$

where  $H = h/h_o$ , with  $h_0$  the initial height and  $T = t/t_s$ , and with  $t_s$  a natural time scale for glue spreading,

(3.13) 
$$t_s = \frac{\mu Z A^3}{F h_0^5}.$$

Thus, although the initial spreading rate is very rapid, justifying our earlier assumption of an initially flattened profile, the spread rate becomes very slow after a time of order  $t_s$ . Explicitly, at time  $t_s$  the width of the glue pile increases by a factor  $(1+5)^{1/5}=1.4$  (a 40% increase), whereas an additional 40% increase would not be achieved until six times this period,  $t_s$ . The period  $t_s$  (or close to it) thus represents the optimal effective application time for F, there being little point in adopting a smaller application time (thereby underutilizing the device) or in uselessly increasing the processing time for labels. Note that there are no conceivable alternatives to

(3.13) for algebraically constructing a glue-spreading time scale from the dimensional parameters of interest. Therefore, more sophisticated fluid models would be likely to reach very similar conclusions. Since the optimal processing time for a label is close to  $t_s$ , approximately  $t_s^{-1}$  labels per unit time should be processed under optimal operating conditions. In terms of the mean pressure P = NF/XZ applied to the label, the maximum labelling rate in labels per unit time is

$$R_{max} = \frac{h_0^5 P X}{N \mu A^3},$$
(3.14)

where X is the label width in the x-direction and N is the number of glue strips on each label. The glue strips are clearly visible through the back of an empty bottle. Typically, N is 65. We were able to measure all of the quantities appearing in (3.14) except the pressure P. At the time of writing, this has still not been done. If we assume that P is of the order of the pressure applied by a human finger (this is the true rule of thumb!), then a quick calculation limits the labelling rate to around 180 per minute, which is a rate commonly used in practice. However, since the labelling rate limit is directly proportional to pressure, measurement of the latter is important. Note also that the labelling rate limit is inversely proportional to glue viscosity, which is strongly temperature dependent. For example, measurements taken at the NB Love Adhesives company show that the viscosity of the glue GRIPIT 1340 reduces from  $2.84 \times 10^3$  poise at  $10^{\circ}$ C to  $3.6 \times 10^2$  poise at  $30^{\circ}$ C.

4. Water Transport from Glue to Paper and Atmosphere. Since the glue strips are around 0.6 mm wide and the paper is 0.1 mm thick, it is valid to assume a one-dimensional model for absorption of liquid into the paper normal to the surface. As a first approximation, we neglect the effect of paper expansion on water transport. Absorption by capillary action of a liquid by a porous medium at volumetric water content (volumetric proportion of local region occupied by water)  $\theta(y,t)$  is governed by a nonlinear diffusion equation [4, 9]

$$\frac{\partial \theta}{\partial t} = \frac{\partial}{\partial y} \left[ D(\theta) \frac{\partial \theta}{\partial y} \right]$$

subject to  $\theta = \theta_0$  (constant saturated value) at y = 0 and  $\theta = \theta_i$  (initial water content) at t = 0. Here,  $D(\theta)$  is the nonlinear diffusivity of liquid in the medium.

At early times, before any significant amount of water reaches the outer boundary, the solution must agree with the similarity solution appropriate for the semi-infinite domain. Hence, with  $\theta$  a function of  $y/\sqrt{t}$  at early times, the total volume of water absorbed per unit cross-sectional area, must take the form

$$(4.2) i = St^{1/2} + O(t),$$

where S is the experimentally measurable sorptivity [3] that depends only on  $\theta_i$  and  $\theta_0$  and on the shape of  $D(\theta)$ . We guess that the average capillary properties of paper may be approximated by those of a silty clay loam, for which  $S = 10^{-4} \text{ms}^{-1/2}$  [7]. At this rate, water would proceed to saturate the thin label in about 1 second. This was borne out by our own experiments, in which labels wetted with excess water curled in 2 seconds. However, D scales like inverse viscosity  $\mu^{-1}$  [4], from which it follows that S scales like  $\mu^{-\frac{1}{2}}$  and absorption time scales in proportion to  $\mu$ . Casein glue, with a viscosity of  $10^5$  cp, compared to 1 cp for water, would take a day to saturate the paper. What is observed is that hygroscopic deformation of labels first occurs after 15

minutes. This corresponds to absorption of a liquid of intermediate viscosity  $10^3$  cp. We suggest that the paper is acting as an imperfect filter, allowing a little case to enter with the water, but leaving behind a more concentrated glue.

After the glue has been applied and the label has been pressed to the bottle, the glue must dry so that it can form a rigid gelatinous bond. It is commonly believed that most of the glue water, lost during the period of gel formation, is taken up by the paper label. Eurokete, a commercially available labelling paper, has a Cobb value of 25 g m<sup>-2</sup> (or 0.025 mm equivalent water depth). This value is defined to be the level of water uptake in paper in contact with free water for 60 seconds, enough time for saturation to occur. On the evening of Tuesday, January 30, 1996, a number of glued. 10 cm by 10 cm labels emerging from the glue pallet in the Nuriootpa plant, in the Barossa Valley of South Australia, were measured to have a glue mass of 2 g <sup>+</sup> 0.5 g. Since casein glue is approximately 65% water by volume, this corresponds to a water level of 130 g m<sup>-2</sup>, five times the Cobb value of the paper. Only about 20% of the glue water will be absorbed by the paper in the important early stages of glue drying. This may increase viscosity by two orders of magnitude [5]. However, most of the water must eventually be removed by atmospheric drying. Hence it is important to leave channels between the glue strips and to provide a drying environment after packing bottles. Since the upper surface of a label is varnished, drying can occur only at the edges. If the solar radiation constant is converted to latent heat, the equivalent evaporation rate is 0.4 cm day<sup>-1</sup>. Rates of 1 cm day<sup>-1</sup> are observed over desert soils. With extra drying assistance, drying could be expected to proceed to the center of the labels in about 5 days. This agrees with the observed bubble response times as atmospheric conditions change.

5. Deformation of Labels. Considerations here are very similar to those of the "Piping in Newsprint Rolls" problem discussed at the 1988 MISG [2]. By analogy with that problem, it is already clear that enough water is available to cause visible hygroexpansion in the paper labels. There are some significant differences between that problem and the current one. Unlike in newsprint rolls, there is glue present and this will help to prevent sliding of the paper sheet. The glue is applied in periodic strips and the question arises whether this is a good design for preventing paper buckling as it absorbs water and expands.

In fact, the expansion is highly anisotropic. Due to the stresses applied in the paper production process, the wood fibers align preferentially in one direction. The paper is more rigid in the fiber direction and expansion occurs predominantly in the transverse direction. With the bottle axis vertical, labels are always printed so that the fiber direction is also vertical. Otherwise there would be extra stresses when the paper is made to conform to the curvature of the bottle.

It is recommended by the bottling machine manufacturer that glue strips should be applied transverse to the wood fibers, as this provides reinforcement against hygroexpansion. Applying glue strips parallel to the wood fiber direction, as in current practice, could weaken the cross-fiber hydrogen bonds, thereby enhancing paper deformation.

In our own test, after 60 seconds of immersion in water, a label expanded by 0.7% in the direction parallel to one edge, but with no noticeable expansion in the orthogonal direction. A representative from AC Printworks, a label supplier, identified the direction of expansion as the recorded cross-fiber direction. This expansion would be enough to accommodate a semicircular cylinder of diameter 1.2 mm on an initially flat  $10~{\rm cm} \times 10~{\rm cm}$  label. Elongations of 3% during immersion had been reported.

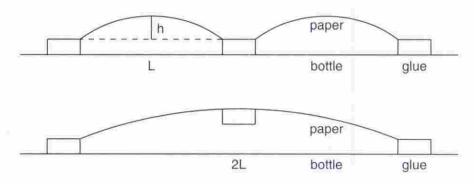


Fig. 5.1 Buckling of sheet between glue strips. Top: secure strips; bottom: central strip detached.

When a sheet of paper absorbs water, it expands in the direction transverse to the fibers, with resulting strain  $\varepsilon = \Delta L/L$ . If the paper is not free to expand indefinitely, it must be experiencing a compressive stress P. An unattached length L of paper will eventually buckle when the load reaches a critical value. Linear elasticity theory has previously been applied to paper buckling at the 1988 MISG [2]. For a beam of length L and thickness d, the critical stress for buckling is [10]

(5.1) 
$$P_c = \pi^2 E I / L^2$$
,

where E is Young's modulus (approximately  $2 \times 10^9 \text{Nm}^{-2}$  for paper), and  $I = d^3/12$ . In linear elasticity theory,

$$(5.2) P = \varepsilon E d.$$

Hence, for a strain  $\varepsilon$ , the critical length of unattached sheet at the onset of buckling is

$$L_c = \frac{\pi d}{2\sqrt{3\varepsilon}},$$
(5.3)

independent of Young's modulus. If the glue strips remain attached and expansion takes place transverse to the strips, then buckling will first occur when  $\varepsilon$  is large enough so that  $L_c$  is the length of unattached sheet (approximately 1.5 mm) between the glue strips. For paper of thickness  $d=100~\mu\mathrm{m}$ , this occurs when the strain is  $3.7\times10^{-3}$ , about half the expansion that we observed in fully wetted labels. For simplicity, we take the shape of a buckle, viewed cross-sectionally, to be a circular segment of half-angle  $\theta$ .

It can be verified using elementary geometry that  $\theta$  is equal to the angle between a tangent to the circular segment at either of its endpoints and the chord (the flat, unbuckled label viewed cross-sectionally) that defines the circular segment. For a segment of small half-angle  $\theta$ , we have  $\varepsilon \approx \theta^2/6$  and  $\theta \approx 4h/L$ , implying

$$(5.4) h^2 \approx 3\varepsilon L^2/8.$$

For fully wetted paper with  $\varepsilon \doteq 7 \times 10^{-3}$  and with glue strips attached, the buckle height will be approximately 80  $\mu$ m, slightly less than the paper thickness (typically 80–100  $\mu$ m). Hence, with an unattached sheet of this length between glue strips, swelling may be accommodated without noticeable visual effects. When the critical

buckling length decreases to less than 2L (Figure 5.1 bottom), a detached central glue strip may be lifted. By combining (5.3) and (5.4), at the onset of buckling, the paper will lift a distance

$$(5.5) h = \frac{\pi d}{4\sqrt{2}},$$

which depends only on the paper thickness. From (5.4), when the strain increases to the value  $7 \times 10^{-3}$  for fully wetted paper, h will increase to 160  $\mu$ m. We suggest that this is the mechanism by which glue strips are first threatened.

However, a full analysis would require more information on the strength of the glue–bottle bond at various water contents. Furthermore, the glue is expected to be viscoelastic, and the glue strips may be under longitudinal tension after drying.

6. Conclusions. This is a multifaceted problem that benefitted from the teamwork of a number of mathematical scientists. Simple mathematical models have provided some insight. The deformations observed in labels are entirely consistent with water absorption and subsequent hygroscopic expansion of the label paper transverse to the direction of the paper fibers. There is more than enough water in the glue layer to fully saturate the paper labels. Most of the excess water must eventually be removed via the edge of the label by atmospheric drying. Therefore, labelled bottles need to be stored in a good drying environment.

Our simple model predicts a near-optimum labelling rate given in (3.14). With the exception of pressure applied to the label, which could be measured, most of the parameters appearing in this equation are known. Before this optimum labelling rate can become an industry standard, it will need to be verified experimentally.

Beyond the simplistic models presented here, there are several more sophisticated modelling tasks worthy of future study. These include the filtering of mixtures of fluids with different viscosities, a three-dimensional model of glue spreading, and buckling of sheets with time-dependent reinforcements due to drying glue. It is envisaged that government and industry funding agencies will be approached to support further experimental and theoretical investigations along the lines suggested here.

For educational purposes, there are several points that can be reinforced by this example. Back-of-envelope calculations can often be very instructive. Following some simple measurements and calculations, it became clear that, contrary to popular opinion, the glue contains more than enough water to totally saturate the paper. This caused us to shift our attention to atmospheric drying. The relationship between strain and buckle height, developed in the previous section, is a good application of elementary geometry. As it combines circle geometry with Maclaurin expansions for the elementary trigonometric functions, the geometric part of section 4 may be used as an example for freshman mathematics.

As in most practical modelling tasks, ideas of dimensional analysis and scaling were useful here. A time scale for glue spreading was a natural and useful outcome of lubrication theory. The form of this time scale could have been indicated by basic dimensional analysis, without any reference to dynamics beyond the choice of significant influential parameters (viscosity, pressure, and depth). In retrospect, we should have found the time scale in this way. The scaling of hydraulic conductivity with viscosity explains the widely differing time scales for water uptake and glue absorption in paper.

When they are possible, simple similarity reductions of boundary value problems lead to useful functional dependence. The Boltzmann similarity reduction of a general nonlinear diffusion equation, with Dirichlet boundary conditions, leads to a concentration profile expanding at early times as the square root of time. This allowed us to make a very simple approximate calculation of the time-dependent water uptake in paper.

The paper buckling model is an example of answering a practical "when will it happen?" question using a critical bifurcation parameter.

Finally, there are some general educational lessons. First, there is a place for mathematicians working in a multidisciplinary team. Successful teamwork involves good communication with other team members whose perspectives may be different. Second, mathematical modelling may be very useful even in challenging multifaceted practical problems that a mathematician might normally regard as "messy." This activity is unlike solving the familiar laundered problems that one normally encounters in undergraduate courses. Participating students were surprised and amused to see their professors proffering confused ideas as they struggled with this problem. This in turn gave the students confidence to offer their own ideas without any fear of embarrassment. This lesson, on how initial inroads are made on a complicated practical problem, is one of the most valuable.

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