Design of polyester and fiberglass/polyester wet-laid mats

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ABSTRACT *The increased availability of recycled polyester provided the incentive for this study into the design of roofing mats containing polyester and glass fibers. Fiber diameter, fiber length, mat basis weight, binder composition, and binder level for various blends of polyester/fiberglass were evaluated on laboratory handsheets and on wet-laid mats from a pilot former. Results obtained on these wet-laid webs showed good correlation with subsequent trials conducted on pilot and commercial wet and dry lines, indicating that laboratory handsheets can be used to evaluate and optimize fiber and binder parameters in both wet- and dry-laid webs. The added value of using wet-laid webs is that they eliminate the bias introduced by the various methods of dry-laid construction. The effects of changes in fiber or binder parameters are readily apparent on wet-laid webs, which have negligible structural integrity.*

Techniques for quantifying formaldehyde emissions from binders during drying/curing cycles and subsequent storage were also examined. Laboratory results showed good correlation with the workplace, indicating that the techniques can be adapted to other volatile materials.

KEYWORDS
Binders
Fiber mats
Formation
Glass fibers
Nonwovens
Polyesters
Product design
Roofing papers
Test methods

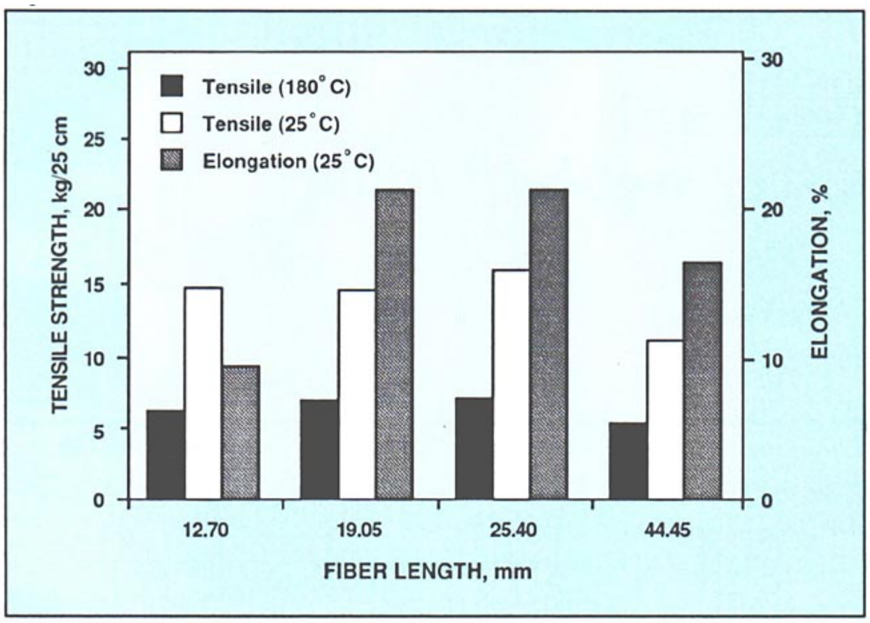
Interest in the manufacture of wet-laid polyester and polyester/fiberglass composites has been sparked by two factors: (a) an increase in the supply of recycled polyester fiber and (b) available machine time on wet-forming fiberglass-mat production lines. Polyester fibers offer advantages over fiberglass in the areas of elongation and flexibility. These advantages, however, come at the expense of tensile strength and high-temperature stability. The challenge is to achieve a favorable compromise for a given application through innovative mat design. This requires an efficient method of evaluating various mat designs.

The properties of dry-laid mats are largely determined by the method of construction. Operations such as spunbonding, carding, needling, and calendering exert a significant influence on mat characteristics. The variety of dry-laid constructions can complicate the task of evaluating the specific effects of changes in fiber and binder parameters. An alternative approach is to evaluate fiber and binder properties using wet-laid mats. This approach factors out the large effects associated with variations in web construction for dry-laid webs. With a wet-laid mat of uniform formation, it is possible to optimize fiber parameters and then evaluate binders without the confounding influences of web construction. Because wet-laid mats have no appreciable structural integrity, the relative strength or weakness of a binder system is readily apparent.

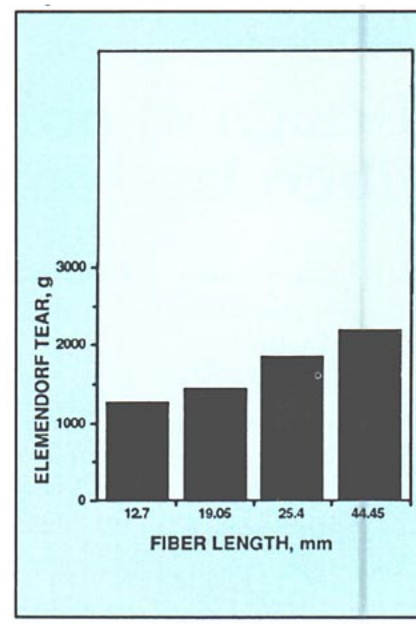
This is the approach we used to evaluate and optimize the design of polyester and fiberglass/polyester wet-laid mats for use in roofing materials. The latex binders used in wet-laid roofing mats are usually modified with co-binders such as urea-formaldehyde (UF) or melamine-formaldehyde (MF). These co-binders provide the dimensional stability needed to withstand high-temperature application of modified bitumen. Formaldehyde emissions for typical binder systems were also evaluated.

Preliminary work was carried out on laboratory handsheets. Trends and conclusions of significance were confirmed during subsequent pilot trials on an inclined-wire former (Sandy Hill Delta former) (*1*). The results obtained from laboratory handsheets and from trials on the inclined-wire pilot former are presented here.

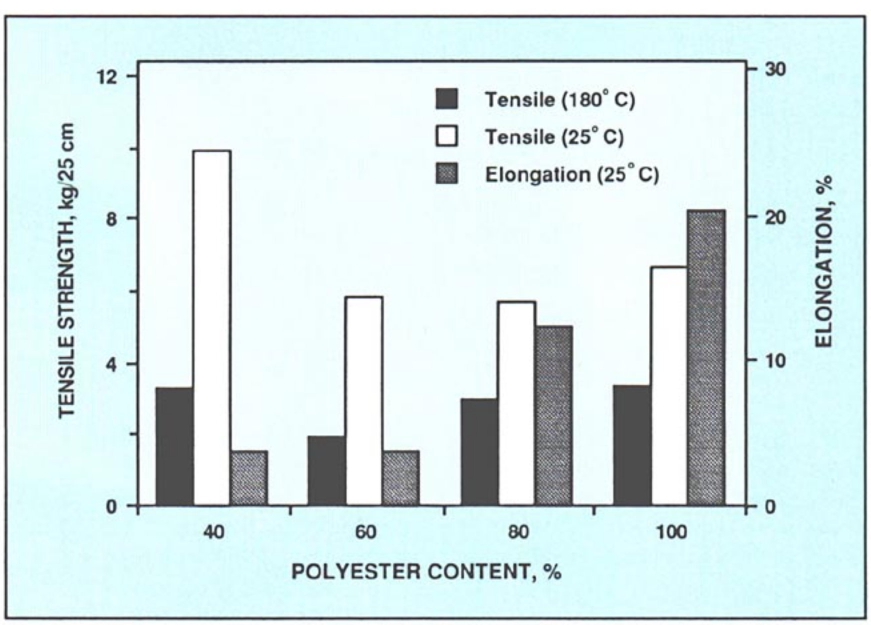
1. Laboratory data for mat properties (tensile strength and elongation) as a function of fiber length



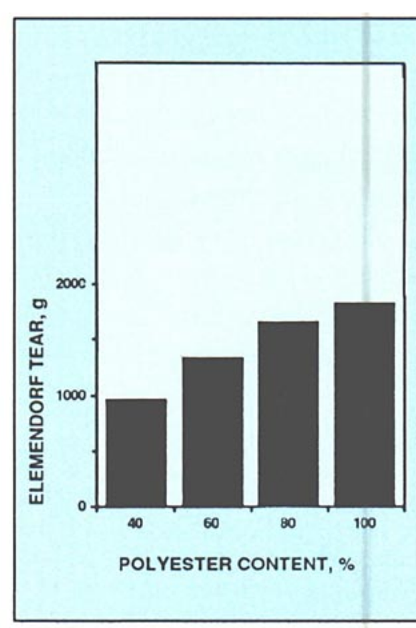
2. Mat tear strength as a function of fiber length



5. Pilot-trial data for mat properties (tensile strength and elongation) as a function of polyester content in polyester/fiberglass blend



6. Mat tear strength as a function of polyester content in polyester/fiberglass blend



Subsequent work on other pilot and commercial wet and dry lines showed good correlation with these results, indicating that the wet-laid technique can also be used to evaluate binders intended for use in dry-laid mats.

General observations

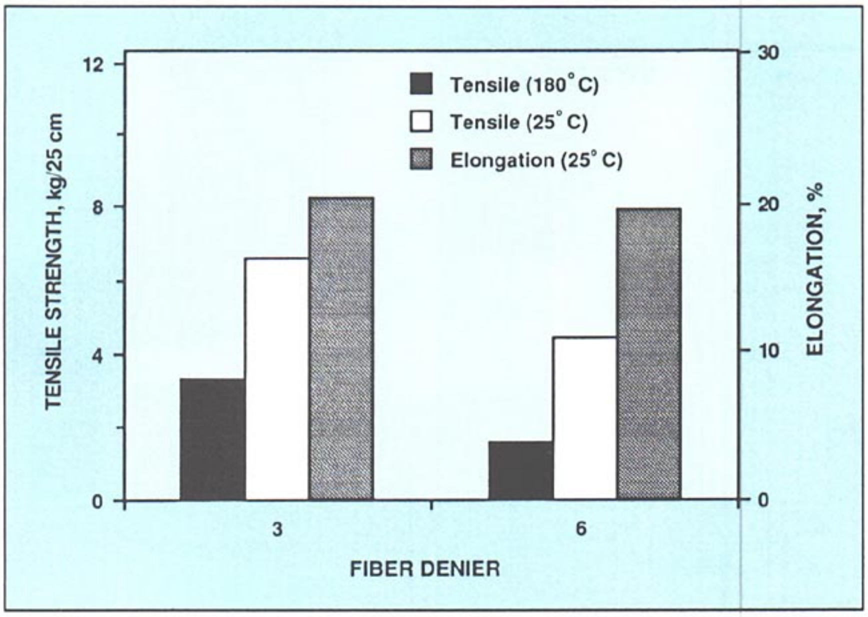
Wet-laid polyester mats are not a dominant force in the market, but their performance characteristics demonstrate significant potential. Sheet performance is determined by three major factors:

1. Fiber parameters
2. Mat definition
3. Binder considerations.

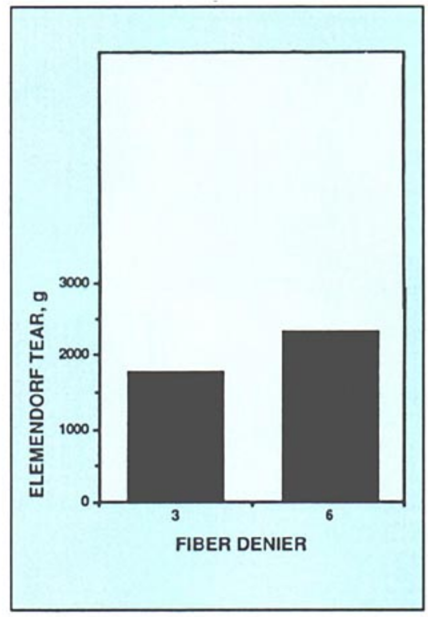
Although all three interact, each must be addressed individually in a logical fashion to maximize performance of the final sheet. In the context of roofing materials, the properties of greatest importance are tensile strength at ambient and high temperature, elongation at ambient and high temperature, and tear resistance. Elongation and tear are dominated by fiber parameters, are characteristically high, and vary less significantly than tensile strength. In the wet-laid context, we chose load-to-break as the measure of high-temperature performance rather than elongation.

In this study, unless otherwise noted, all mats were constructed of 19-mm, 3-denier polyester, had a basis

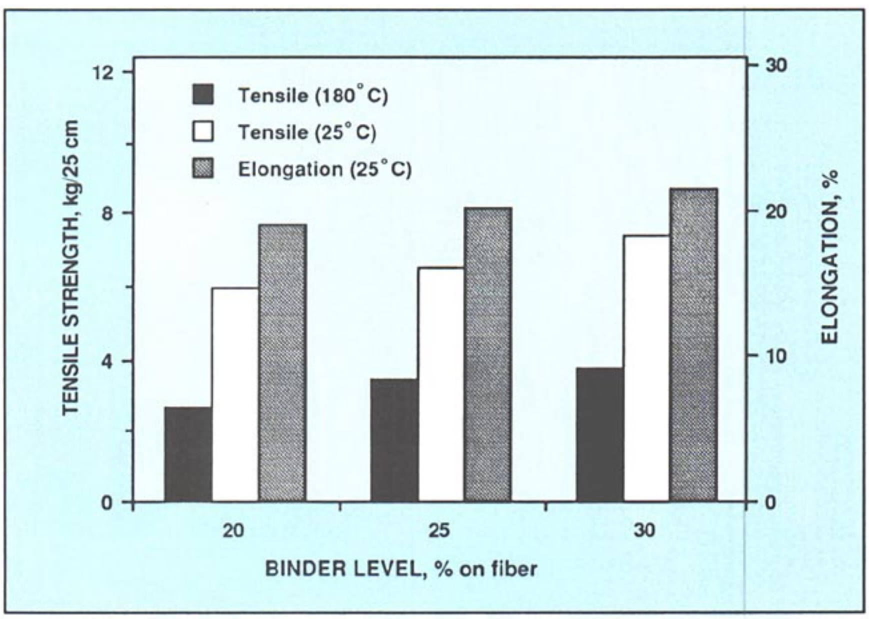
3. Pilot-trial data for mat properties (tensile strength and elongation) as a function of fiber denier at constant length-to-diameter ratio



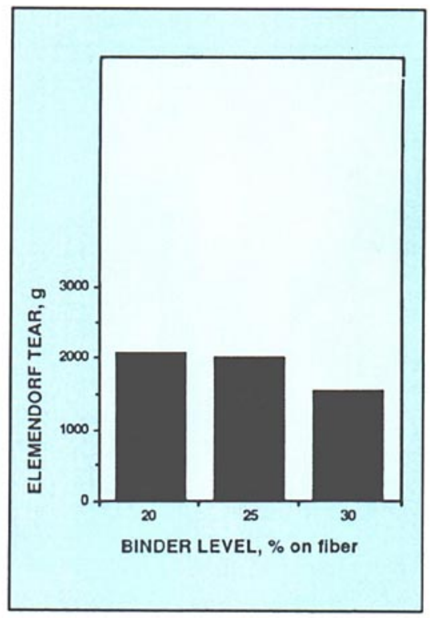
4. Mat tear strength as a function of fiber denier at constant length-to-diameter ratio



7. Laboratory data for mat properties (tensile strength and elongation) as a function of binder level at constant basis weight



8. Mat tear strength as a function of binder level at constant basis weight



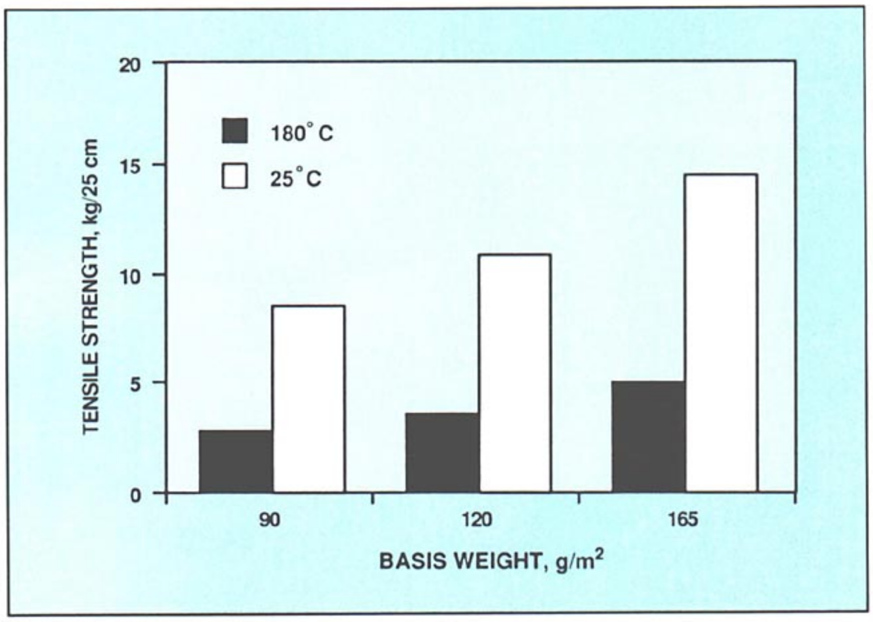
weight of 170 g/m^2, a binder level of 20% based on total weight (25% based on fiber weight), and a constant binder system.

Fiber parameters

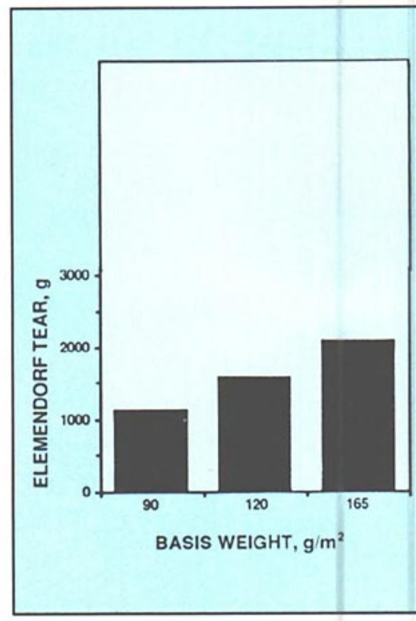
In general, sheet tensile and tear strengths are maximized by using the longest fibers that can be handled without compromising web formation. **Figure 1** shows that tensile strength for the polyester mats increased gradually up to a fiber length of 25.4 mm and then dropped sharply for the longest fiber length. **Figure 2** shows that tear increased steadily with fiber length. Subsequent pilot trials indicate that a good compromise in terms of handleability and mat properties is obtained with 19-mm, 3-denier fibers with a length-to-diameter ratio of 1000. We have also done considerable laboratory work using 25.4-mm (1 in.), 3-denier fibers and 19-mm (0.75 in.), 1.5-denier fibers.

The effect of denier was quite dramatic when length-to-diameter ratio was maintained relatively constant. **Figure 3** shows a 40–50% reduction in tensile strength when fiber denier was increased from 3 to 6. **Figure 4** shows that tear strength increased with the higher-denier fibers. For a roofing material, this is probably a poor trade-off, since polyester mats usually are short on tensile strength and long on tear strength relative to fiberglass mats.

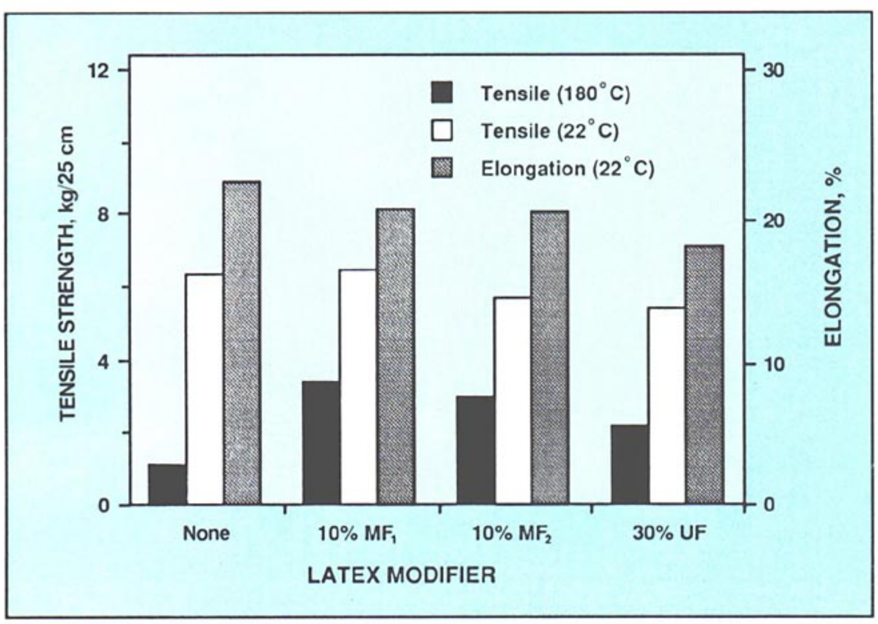
9. Pilot-trial data for mat tensile strength as a function of basis weight at constant binder level



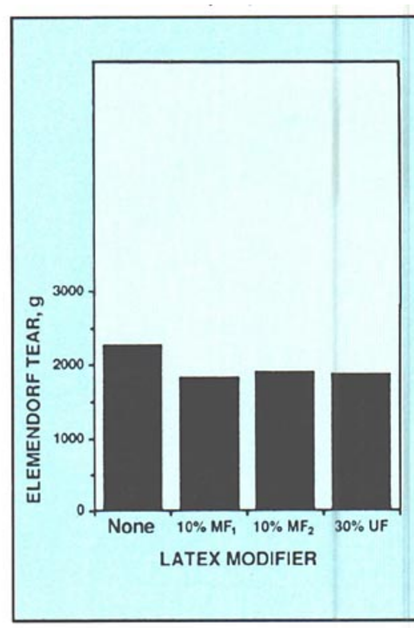
10. Mat tear strength as a function of basis weight at constant binder level



11. Pilot-trial data for mat properties (tensile strength and elongation) as a function of latex modifier. MF_1 and MF_2 are commercial melamine-formaldehyde products.



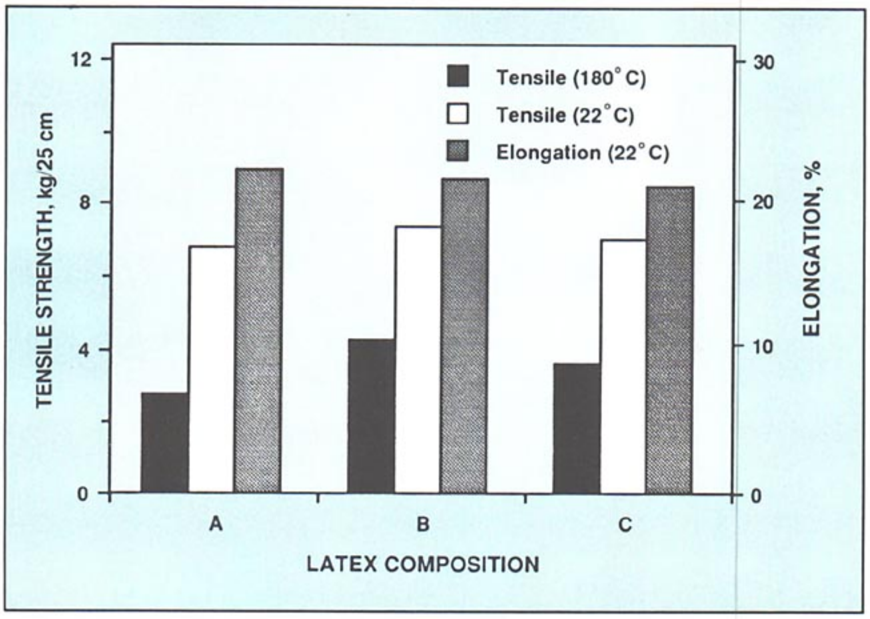
12. Mat tear strength as a function of latex modifier. MF_1 and MF_2 are commercial melamine-formaldehyde products.



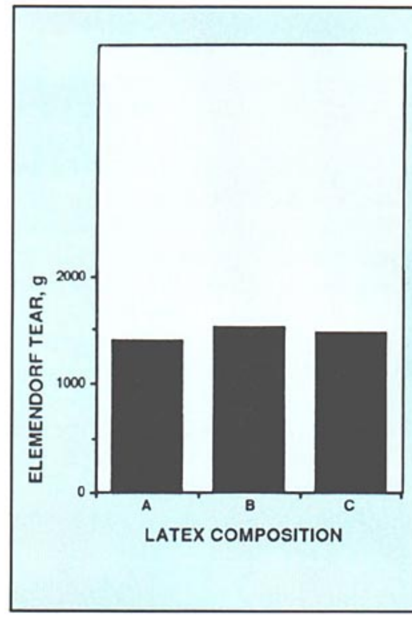
When fibers are blended, the effects are somewhat confounded by subtle changes in sheet formation. Better results are frequently achieved when the secondary fiber is shorter than the primary fiber. Thus, the relative lengths of the two fibers should be selected on the basis of which properties are most desirable in the chosen application.

When considering blends of polyester and fiberglass, care must be taken to achieve uniform fiber laydown and overall sheet formation without fiber "wrap around," which leads to disproportionately lower tensile and higher tear strengths. Addition of modest levels of glass fiber to a polyester sheet can increase tensile strength and high-temperature dimensional stability, with some penalty in tear strength and elongation. Elongation tends to fall off very rapidly, as seen in **Fig. 5**. In our experience, 15-20% fiberglass provides optimum tensile strength and elongation. On a more qualitative level, inclusion of 10-15% fiberglass in a polyester mat raises the aesthetic quality of the web, improves general handling characteristics, and reduces production-line shrinkage significantly without a major sacrifice in tear strength or elongation. On the other end of the spectrum, introduction of small amounts of polyester into a fiberglass mat likewise can enhance

13. Pilot-trial data for mat properties (tensile strength and elongation) as a function of latex compositions modified with the same MF



14. Mat tear strength as a function of latex compositions modified with the same MF



I. Formaldehyde content in typical binder systems[a]

	Free formaldehyde (wet mix),[b] ppm	*Formaldehyde emissions at 175°C,[c] kg*	*Residual formaldehyde in mat,[d] ppm*	*Formaldehyde emissions during storage,[e] ppm*
Standard latex	550	0.75	<20	<100
Ultralow latex	10	0.2	<20	<100
Latex (90%)/MF (10%)	1,000	>3	500	1,000
Latex (80%)/UF (20%)	12,500	>2.5	4,500	15,000

[a]These binder systems are made of commonly used commercial materials. Other common materials could provide different results.
[b]Nash reagent on serum.
[c]Estimate based on consumption of binder solids at 1000 kg/h.
[d]AATCC 112-1965; 20 h @ 49°C (120°F).
[e]AATCC 112-1965; 4 weeks @ 49°C (120°F).

tear strength and flexibility without disastrous reductions in tensile strength (*2*). **Figure 6** shows tear strength for several blends of polyester and glass fibers. Both of these areas require more extensive study with specific product goals, since they offer unique balances of properties.

Mat definition

The effects of binder level and basis weight are predictable, but the magnitude of their influence needs clarification. **Figure 7** shows that at constant basis weight, tensile strength increases with increasing binder level. Elongation has a relatively flat response. This is not surprising, since this is a fiber-dominated property. **Figure 8** shows a decrease in tear strength when binder level was increased from 20% to 30%.

When binder is held constant and basis weight is varied, tensile strength and tear strength increase directly with mat weight, as seen in **Figs. 9 and 10**, respectively. Elongation did not vary with basis weight.

Binder considerations

Binder composition plays an important role in determining mat properties. In applications requiring good high-temperature performance, wet-laid polyester mats usually require binder systems containing an external crosslinker. Accordingly, at the outset of this study, a latex optimized for polyester substrates and maximum reactivity with thermosetting coresins was defined. The binder used throughout these studies was a high-molecular-weight self-crosslinking acrylic latex with a glass-transition temperature (T_g) of 25°C (determined by differential scanning calorimetry). This T_g is typical of binders used in commercial fiberglass roofing mats. Of course, changes in binder T_g can affect property balance.

Preliminary work indicated that the best overall balance of properties was achieved with latex modified with approximately 10% MF. The UFs commonly used in fiberglass mats were not quite as effective in the polyester system. **Figures 11 and 12** illustrate the influence of latex modifier in a composition designed for maximum reaction with MF. **Figures 13 and 14**, on the other hand, demonstrate the effect of three latex compositions, each modified with the same MF.

Mats without high-temperature constraints require no co-binder. Tensile strength and elongation at ambient temperature are not appreciably affected by MF modification (Fig. 11), while tear for the 100% latex system is about 20% higher (Fig. 12).

Dry-laid webs for high-temperature roofing applications frequently use a self-crosslinking latex as a sole binder. Wet-laid laboratory handsheets are particularly useful in evaluating binders, since these uniform webs eliminate the effects of different dry-laid web constructions and normal variability (*3*). The quality of wet sheet formation can be controlled by mastering the appropriate techniques for good fiber laydown. The end result is a reproducible means of evaluating binder and general mat properties in the laboratory with a high correlation with machine performance. Polymer strength and elongation at ambient and elevated temperatures are readily assessed, since fiber/binder interactions, wet-out, and adhesion phenomena can be easily observed. One of the side benefits in using the wet-laid approach is the ability to predict mat loft or caliper in dry-formed webs with subsequent good correlation with performance on a commercial machine.

Formaldehyde issues

Formaldehyde emissions are a legitimate concern in this era of heightened environmental awareness. Three areas of major concern are: (a) the formulation and application areas, (b) the drying/curing oven, and (c) residual formaldehyde in the mat, which can be released downstream, particularly when exposed to high temperature and humidity.

Formaldehyde exposure in the applicator or formulation area is presumably a function of free formaldehyde in the system. The origin of the formaldehyde can be traced to commonly used polymers containing methylolated amide as well as related crosslinking functionalities and additives, including preservatives and coresins, such as MF and UF.

What happens in the ovens and postcure is highly dependent on cure conditions. Our laboratory technique involved sampling the air while drying and curing mats under the controlled conditions typically used to evaluate standard physical properties. The observed formaldehyde concentrations were related to actual amounts in the original mix by carefully running calibration curves on formaldehyde-free materials spiked with appropriate levels of formaldehyde. In our experience, essentially all of the free formaldehyde in a latex is released in the oven, plus an additional amount from the reactive crosslinker in the polymer backbone. In all instances, the total amount of emissions varies directly with temperature. However, the situation in the final web is reversed. Lower cure temperatures increase the potential for higher levels downstream (storage, shipping, converting operations) or during end use. Heat and humidity during storage trigger release of additional quantities of formaldehyde.

Table I lists formaldehyde content in typical binder systems. Materials fall into two groups: latex sole binder and systems modified with UF or MF. Emissions from typical self-crosslinking latexes generally are orders of magnitude below the formaldehyde-modified systems.

Monitoring of commercial nonwovens equipment by the NIOSH-52 method indicated good correlation of trends with laboratory measurements. Several latex binder systems had substantially lower formaldehyde emissions than conventional state-of-the-art latexes without sacrificing important properties.

Conclusion

With recycled polyester becoming available in large quantities, composite mats of polyester/fiberglass appear to be a promising product-design opportunity. Webs with an attractive balance of properties can be created from these materials at high rates of speed. The challenge is to achieve favorable compromises through effective mat design.

Laboratory evaluation of wet-laid webs shows promise as a screening tool for binder development. Because the wet-laid web has negligible structural integrity, the effects of different binders are readily apparent. This is not the case in dry-laid webs, where structural integrity varies with the manufacturing process used to construct the web. Good correlation was observed for physical properties measured on laboratory handsheets and those measured on webs from commercial dry-laid machines.

Formaldehyde emissions depend on levels in the binder system and on cure history. Cure at higher temperatures results in higher emissions on the production line and lower residual formaldehyde in the end product. Typical self-crosslinking polymers are considerably lower in formaldehyde content than systems modified with commonly used urea-formaldehyde and melamine-formaldehyde external crosslinkers.

Experimental

Formaldehyde measurement

Serum analysis. Nash reagent—acetylacetone (pentanedione-2,4)—is typically used to analyze wet mixes (serum or very dilute, microfiltered latex system) to avoid error from even slight turbidity. The lutidine derivative that is formed is measured colorimetrically (*4*).

Production-line emissions. NIOSH-52 is the method of choice for monitoring production-line emissions. The technique involves collecting 24 L of air at 0.1 L/min. Formaldehyde is derivatized in collection tubes filled with absorbent and subsequently eluted and analyzed by an approved analytical facility (*5*).

Residual formaldehyde. Method AATCC No. 112-1982 was used to assess residual formaldehyde in the mats. The standard method involves exposing the mat to water vapor at 49°C (120°F) for 20 h. To generate an ultimate or limiting value, we extended the basic test to four-week cycles, since the amount of formaldehyde observed leveled off after about two weeks.

Emission during drying/curing cycles. Formaldehyde release during drying/curing cycles was determined in the laboratory by sampling (trapping in a bubbler) the air in a Mathis oven under conditions typical of commercial operation. The flow of effluent air into the bubbler was controlled with a vacuum pump attached to a rotameter at a flow rate of 1.5 L/min. The observed concentrations were related to amounts on the original mix by carefully running calibration curves on formaldehyde-free materials spiked with appropriate levels of formaldehyde. This permits calculation of ballpark estimates of the total amount of formaldehyde emitted in the operation and allows valid comparisons between materials.

Mat preparation

Handsheets. Handsheets were prepared with a Williams sheet mold using standard Separan®/Katapol white water chemistry. Polyester was dispersed in a British disintegrator. In polyester/fiberglass composites, the two fibers were dispersed separately and blended just before dropping to the headbox to avoid "wrap around."

Physical tests. Pilot machine data are the average of cross and machine directions. Tensile strength was tested at 100% rate of extension at ambient temperature and at 200% rate of extension at 180°C. Elmendorf tear strength was determined using the radial tear test (ASTM D 1922-78) (*6*).□

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