

Factors Affecting Initial Bond Strength in Solvent-Based Contact Adhesives

JANUSZ KOZAKIEWICZ, BIBIANNA KUJAWA-PENCZEK, and PIOTR PENCZEK, *Institute of Industrial Chemistry, 01-793 Warszawa, Poland*, and KRZYSZTOF PUTON, *Technical University, 26-600 Radom, Poland*

Synopsis

Novel interpretation is given for the development of initial bond strength maximum in solvent-based contact adhesives at particular elastomer/resin ratios in the adhesive films. Complex relations between initial bond strength, tack of adhesive films, adhesive open time, solvent retention, and resin content in adhesive films are discussed. These results are based on data using polychloroprene adhesives modified with butylphenolic resins and also on previous studies with other contact adhesives. It is suggested that solvent retention is the main factor affecting initial bond strength in solvent-based contact adhesives.

INTRODUCTION

Despite a considerable amount of data concerning properties/structure relationship for pressure-sensitive adhesives published, the effect of various factors on the initial bond strength for solvent-based contact adhesives modified with alkylphenolic resins has not yet been studied carefully. Since the maximum initial strength of adhesive bond is necessary when the bonded specimen has to be subjected to subsequent on-line operation, this problem is, however, very important from the industrial point of view.

The results of our earlier investigations on the adhesives obtained using polyurethane¹ and polychloroprene^{2,3} rubbers indicated that the initial bond strength reached a maximum value at a particular elastomer/resin ratio. A similar phenomenon observed when tack of adhesive film was plotted against elastomer/resin ratio for various pressure-sensitive adhesives has been explained taking into account a two-phase structure of an adhesive film.⁴⁻⁶

However, the results of our studies on polyepichlorohydrin adhesives demonstrated evidently that such interpretation was incorrect when initial bond strength and not tack maximum was concerned.⁷ In that study, solvent retention in an adhesive film was suggested as a factor responsible for this phenomenon. This suggestion was recently supported by new results obtained for polychloroprene adhesives prepared using two different solvent mixtures.⁸

In the present report, further results for polychloroprene adhesives are presented and an interpretation for the marked effect of elastomer/resin ratio on the initial bond strength is offered. It is believed that this interpretation can be applied to all contact solvent-based adhesives in general.

Experimental

Materials applied in this study were: polychloroprene rubber (Skyprene C-40T - Toyo Soda Co.); butylphenolic resins of the same compositions but of different melting points (mp = 60, 65, and 77°C); solvent mixture ethyl acetate/gasoline/toluene = 1/1/1.

Adhesives were prepared by mixing 18% solutions of rubber and resin at various elastomer/resin ratios. After 10 days storage, solvent retention of adhesive films, tack of the films, and initial strength of adhesive bonds were measured for all adhesives.

For solvent retention measurements, approximately 0.4-g portions of adhesives were applied onto glass shields to form 0.4-mm-thick layers, and weight changes were noted after 5, 10, 15, and 20 min open time.

Initial tensile "butt" strength of adhesive bonds was determined at ambient temperature using Instron dynamometer. Aluminum specimens applied for this test were etched in $K_2Cr_2O_7-H_2SO_4$ solution according to a standard procedure.

Tack of adhesive films was determined by the "rolling ball" method.⁹ The ball (4 g glass ball 14 mm in diameter) was rolled on the adhesive film and the distance at which it stopped was the measure of film tack.

RESULTS AND DISCUSSION

In Fig. 1, initial strength of Al/Al bonds is plotted against butylphenolic resin content in the adhesive expressed in phr (parts of resin per hundred parts of rubber).

It can be clearly noted from Fig. 1 that the plot of the initial bond strength vs. resin content passes through a maximum value for all of the three resins studied. The maximum for each resin occurs, however, at a different resin content in the adhesive film. Furthermore, the higher the melting point of resin, the higher is the optimum resin content in the adhesive film. This behavior suggests a significant effect of resin on the properties of adhesive film and can be considered as a result of complex interactions between resin, rubber, and solvents in the film. To examine more closely the assumed interactions, solvent retention in the ad-

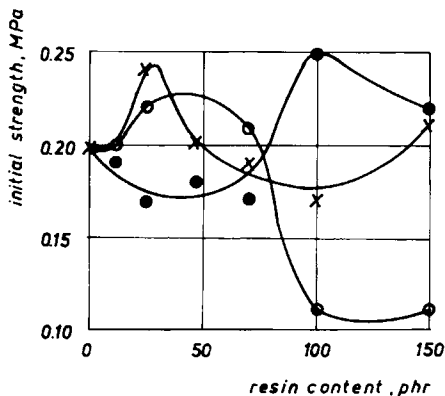


Fig. 1. Effect of resin content in adhesive film on the initial bond strength. Adhesive open time = 20 min: (x) butylphenolic resin of mp = 60°C; (o) butylphenolic resin of mp = 65°C; (●) butylphenolic resin of mp = 77°C.

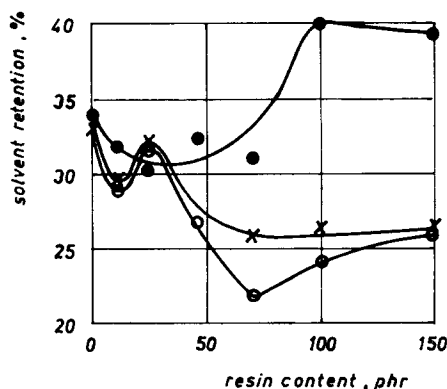


Fig. 2. Effect of resin content in the adhesive film on the solvent retention. Adhesive open time = 20 min: (x) butylphenolic resin of mp = 60°C; (O) butylphenolic resin of mp = 65°C; (●) butylphenolic resin of mp = 77°C.

hesive films and tack of the films were measured using the same adhesive compositions. The results are represented in Figs. 2 and 3.

When Figs. 1 and 2 are compared, one can notice that the maximum of initial bond strength corresponds to the maximum of solvent retention for all of the three resins. This effect confirms the interpretation applied earlier to the results obtained for polyepichlorohydrin adhesives and suggests that solvent retention is the main factor responsible for the occurrence of initial strength maximum at particular resin content in the adhesive film.⁷ This effect can be seen more clearly in Fig. 4, where initial bond strength is plotted against solvent retention for the adhesive containing the resin of mp = 60°C.

It can be also noted from Fig. 2 that the adhesives prepared using resins of different m.p. display different solvent retentions. Similar behavior reported recently for other groups of butylphenolic resins has been explained as being due to specific differences in molecular structure of the resins.¹⁰

In contrast with initial bond strength, the tack of adhesive films seems not to be related to solvent retention (compare Figs. 2 and 3), and its effect on the initial strength can be observed only when it becomes very low.* (See the curves cor-

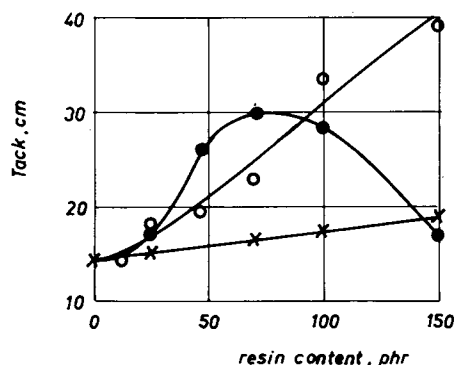


Fig. 3. Effect of resin content in the adhesive film on the film tack. Adhesive open time = 20 min: (x) butylphenolic resin of mp = 60°C; (O) butylphenolic resin of mp = 65°C; (●) butylphenolic resin of mp = 77°C.

* Low tack corresponds to a long distance the ball rolled, i.e., to high values in Fig. 3.

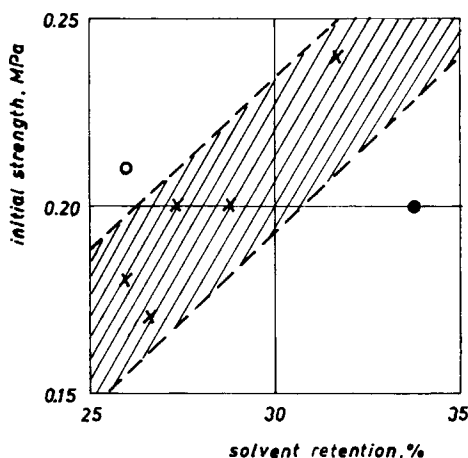


Fig. 4. Initial strength of adhesive bond vs. solvent retention in the adhesive film for adhesives containing butylphenolic resin of mp = 60°C: (O) point corresponding to the adhesive containing maximum amount of resin; (●) point corresponding to the adhesive without resin.

responding to resin of mp = 65°C in Figs. 1 and 3.) This relationship can be easily understood if one assumes that very low tack corresponds to a very dry surface of the adhesive film, which therefore is unable to wet another surface in the bonding process. Since the bonding pressure of 400 kPa is usually used with contact adhesives, it is obvious why the tack of an adhesive film is not as important a factor affecting the bond strength in this case as it undoubtedly is in the case of pressure-sensitive adhesives, where bonding pressures are much lower.

While all the results presented above were obtained using constant open time (20 min), it was interesting to examine as well the effect of adhesive open time on the initial bond strength and tack of adhesive films. In Table I, the resin contents in the adhesive film corresponding to a maximum initial bond strength (i.e., optimum resin amounts) are presented for various adhesive open times. The shifting of the optimum resin amount with increasing resin melting points which was observed in Fig. 1, can also be seen in Table I, and it is noteworthy that the same effect occurs for different open times. It can be noted as well from Table I that the maximum initial strength for shorter and longer open times is attained at different resin content. This phenomenon can be explained based on the conclusions presented above and concerning complex initial strength-solvent retention-resin content interrelationships.

TABLE I
Effect of Adhesive Open Time and Resin Melting Point on Optimum Resin Content in the Adhesive Film

Open time, min	Optimum resin content, phr		
	60°C	65°C	77°C
10	10	25	40
20	25	40	100
30	0	0	25

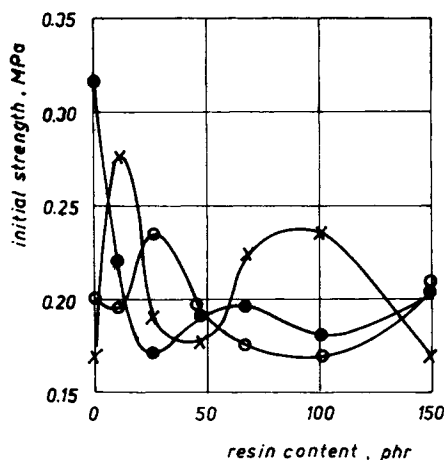


Fig. 5. Effect of adhesive open time on the initial bond strength for adhesives containing butylphenolic resin of $m_p = 60^\circ\text{C}$: (x) open time = 10 min; (O) open time = 20 min; (●) open time = 30 min.

The effect of adhesive open time on initial strength and tack is presented in Figs. 5 and 6. The initial strength (Fig. 5) for the adhesive containing no resin is higher when the open time is longer. It means that in this case film wettability is sufficient, irrespective of open time, and the only factor affecting bond strength is film cohesion, which increases with prolongation of open time. However, situation becomes entirely different when resin is added to the adhesive. Solvent retention is then affected not only by the adhesive open time, but also by resin content in the adhesive film (see Fig. 2). This two-sided effect results in complex changes of initial bond strength presented in Fig. 5.

The behavior of tack is less complicated because after the first period of solvent evaporation (ca. 10 min), i.e., when a partially dried layer has been already formed on the film surface, it does not significantly depend upon solvent retention and open time. This presumably is the reason for the entirely different picture of the tack-resin content relationship observed after 5, 10, and 20 min of open time

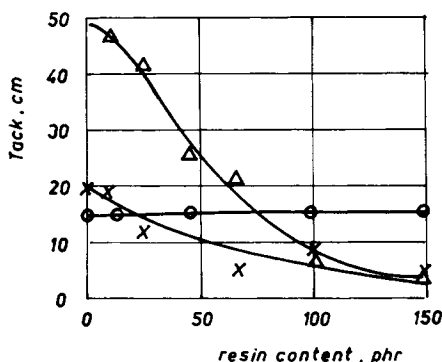


Fig. 6. Effect of adhesive open time on the tack of adhesive films for adhesives containing butylphenolic resin of $m_p = 60^\circ\text{C}$: (Δ) open time = 5 min; (x) open time = 10 min; (O) open time = 20 min.

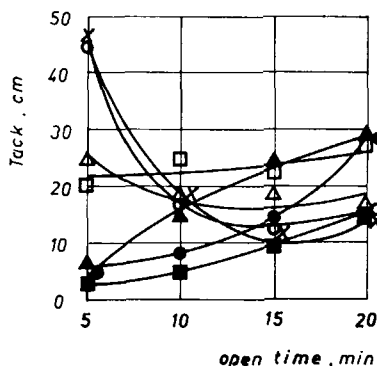


Fig. 7. Tack of adhesive films containing various amounts of butylphenolic resin of $mp = 77^{\circ}C$ vs. adhesive open time: (x) adhesive without resin; (O) resin content = 11.1 phr; (Δ) resin content = 25.0 phr; (\square) resin content = 46.8 phr; (\bullet) resin content = 66.6 phr; (\blacktriangle) resin content = 100.0 phr; (\blacksquare) resin content = 150.0 phr.

for resin of $mp = 60^{\circ}C$ (see Fig. 6). Similar behavior was observed for the two other resins investigated in this study.

It can be also noted in Fig. 6 that for adhesives containing no resin, or small amounts of resin only, the tack measured after short open time is much lower* than the one measured after a long open time. This relationship becomes, however, reversed for adhesives containing more resin. This phenomenon can be seen better in Fig. 7, where the tack is plotted against open time for adhesives containing different amounts of resin. It can be explained assuming that resin plays the role of tackifier only when solvent content in the adhesive film is high. Within the thin layer formed on the film surface during solvent evaporation, the resin loses its tackifying properties, and eventually the point is reached when the higher resin content in the adhesive film results in a lower film tack (20-min open time curve in Fig. 6).

Based on the results obtained in this work and reported previously,^{7,8} the following general interpretation for the observed significant effect of resin content in the adhesive film (i.e., elastomer/resin ratio) on the initial bond strength in solvent-based contact adhesives can be offered.

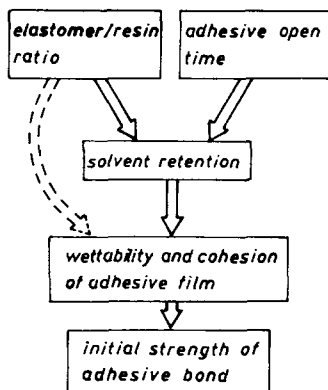


Fig. 8. Factors affecting initial bond strength in solvent-based contact adhesives.

* Corresponding to higher value in Fig. 6 (see the previous footnote concerning tack).

Initial bond strength depends on the cohesion and wettability of the adhesive film. These film properties are affected by resin and solvent content in the film, i.e., by the elastomer/resin/solvent ratio in the film. It was shown above that solvent retention is related not only to adhesive open time but also to resin content, and the pattern of this relationship is similar to the one observed when initial bond strength was plotted against resin content. Therefore, it can be concluded that the effect of resin content on the bond strength is significantly dependent on solvent retention. It seems that at certain resin content in the adhesive film, a elastomer/resin/solvent ratio reaches a specific value yielding film cohesion *still* sufficient and film wettability *already* adequate, resulting eventually in the maximum initial bond strength.

The general conclusion from these considerations is again that the most important factor affecting initial bond strength for solvent-based contact adhesives is solvent retention in the adhesive film. The schematic picture of this conclusion is presented in Fig. 8.⁸ Direct effect of the elastomer/resin ratio, which is not as important to the bond strength as complex effect of solvent retention, is marked by the broken line in Fig. 8.

Further investigations on the initial bond strength of fast-crystallizing polyurethane contact solvent-based adhesives are now being carried out in the authors' laboratory to check the validity of the interpretation presented above and to study the complex effect of crystallization and resin content in adhesive films on the initial bond strength.

References

1. B. Kujawa-Penczek, *Polimery* (Warsaw), **13**, 215 (1968).
2. P. Penczek, L. Walaszek, L. Stachurski, *Zeszyty Naukowe Kielecko-Radomskiej WSI, Ser. Garbarstwo, Z-1*, Kielce, 1971.
3. O. Ackermann, *Adhäsion*, **23**, 172 (1979).
4. C. W. Hock, *J. Polym. Sci. C*, **8**, 199 (1963).
5. F. H. Wetzel and B. B. Alexander, *Adhes. Age*, **7**(1), 28 (1964).
6. K. Kamagata, H. Kosaka, K. Hino, and M. Toyama, *J. Appl. Polym. Sci.*, **15**, 483 (1971).
7. J. Kozakiewicz and P. Penczek, *Adhes. Age*, **20**(7), 29 (1977).
8. J. Kozakiewicz, B. Kujawa-Penczek, and P. Penczek, *Prep. of 1st Joint Italian-Polish Seminar on Multicomponent Polymeric Systems*, Capri, 1979.
9. K. Kamagata, T. Saito, and M. Toyama, *J. Adhesion*, **2**, 279 (1970).
10. T. Tanno, I. Shibuya, and Y. Mukoyama, *Int. Symp. on Phenolic Resin Chemistry*, Washington, D.C., 1979.

Received October 8, 1980

Accepted March 19, 1981