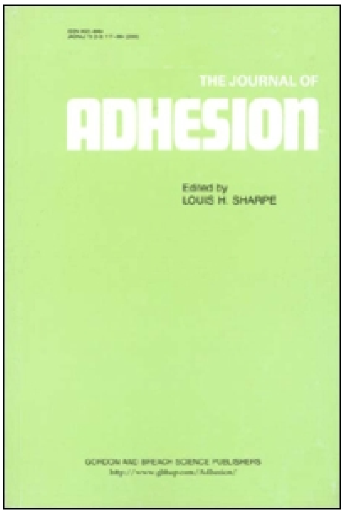

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Welding Mechanisms of Plastics: A Review

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Strength of welded joints is a function of technological parameters of the production process. The type of function is dependent on the welding mechanism. Different mechanisms were found under various welding conditions. The processes included in the plastic welding mechanism are divided into two groups:

1) Processes which realize the joining of the parts.
2) Processes which create conditions for the first group to proceed.

The first series of processes includes:

a) diffusion of macroradicals, molecular segments or molecules of the polymer which can be either in a solid, melted or dissolved state.

b) convective mass transfer.

c) recombination of macroradicals across the contact surface.

d) physical (surface) interaction.

e) any combination of processes described above.

The second group contains:

a) formation of the real contact surface.

b) formation of the macroradicals.

c) destruction and removal of inert layers which prevent real contact of active material.

Each process and the conditions of its proceeding are discussed individually.

INTRODUCTION

Plastic welding is a technological operation based on the autohesion process. Autohesion is the capacity to form stable bonds as two different surfaces of the same substance are brought into contact.[1] These bonds prevent peeling of the joined parts at the initial contact surface. The first description of the autohesion process appeared in 1935–7.[2–4] Several authors tried to explain the mechanism of autohesion.[5–7] The autohesion was related to the presence of free molecular ends on the contact surface.[5] J. R. Scott developed this theory supposing that these molecular ends could diffuse across the contact surface and form the autohesion bonds.[6] The diffusional nature of autohesion was confirmed by direct and indirect methods of research.[8–22] But several other authors determined that under certain conditions autohesion and plastic welding are not controlled by diffusion.[23–28] Autohesion is a basis and a component of plastic welding. Strength of welded joints depends on the rate and the mechanism of the overall process. Therefore, review of possible plastic welding mechanisms is necessary for predicting the mechanical properties of the welded joints.

The mechanism of plastic welding is a total combination of the processes which occur from the beginning until the end of the welding operation. Different experts assume several mechanisms of plastic welding. Variations in the welding mechanism are related to temperature range, condition of surfaces being welded and value of pressure employed during the joining process. Processes included in the welding mechanism can be divided into two groups:

1) Processes which realize the joining of the parts being welded.
2) Processes which create conditions for the first group to proceed.

The first series of processes includes:

a) diffusion of macroradicals, molecular segments or molecules of the polymer which can be either in a solid, melted or dissolved state.

b) convective mass transfer.

c) recombination of macroradicals across the contact surface.

d) physical (surface) interaction.

e) any combination of processes described above.

Physical (surface) interaction takes place in every mechanism. If the surface interaction does not conclude the joining process, the physical interaction should be attributed to the second group which contains:

a) formation of the real contact surface.

b) formation of the macroradicals.

c) destruction and removal of inert layers which prevent real contact of active material being welded.

Several of the processes mentioned above can be described by the same Arrhenius equation but with different constants. Therefore, if the equation for description of the entire process is known, it does not always mean that the limiting process can be determined. Moreover, the rate of different processes is a function of welding conditions but not the same function. So at two different temperatures of welding, two different processes can limit the rate of the overall welding operation. Such exchange is shown on the graph in Figure 1,[29,30] where both parts of the broken line can be described by the exponential equations, but activation energies in these equations vary by approximately a factor of ten. Autohesion processes occur on the closed surface and spread out to about 10–100 angstroms in depth from the contact surface. So it is difficult to study the welding mechanism by direct methods. The mechanism of plastic welding can be predicted if the rates of different processes that occur during the operation are known under various conditions. But there are not enough data for such calculations now. Therefore, each process will be discussed individually.

PROCESSES OCCURRING DURING WELDING

Diffusion

The fact that diffusion limits the autohesion in polymers under certain conditions was proved by many experiments. The direct measurements of the interdiffusion rates in polymers were made by F. J. Bueche.[12,13] Radioactive plasticized polystyrene produced from styrene marked by the radioactive isotope C_{14} was employed for study. The experiments have shown that:

1) The interdiffusion coefficient is approximately inversely proportional to the viscosity of the polymer tested.

2) Activation energy of diffusion is equal to the activation energy of the viscous flow.

Therefore, it was concluded that the intensity of the molecular motion in a polymer can be estimated by the viscosity of the polymer under the same conditions. Direct proof of interdiffusion in polymers during the welding process was obtained by employing tagged atoms, ultraviolet and electron microscopes.[14] Authors have established that the rate of diffusion is high enough to realize formation of autohesion joints by interdiffusion of the molecules.

The second group of data is attributed to indirect facts. Establishing the diffusion limiting influence on the rate of autohesion by these factors consists of comparing the effect of the same factors on both diffusion and autohesion. Limitation of the overall process by diffusion can be considered to be proved if

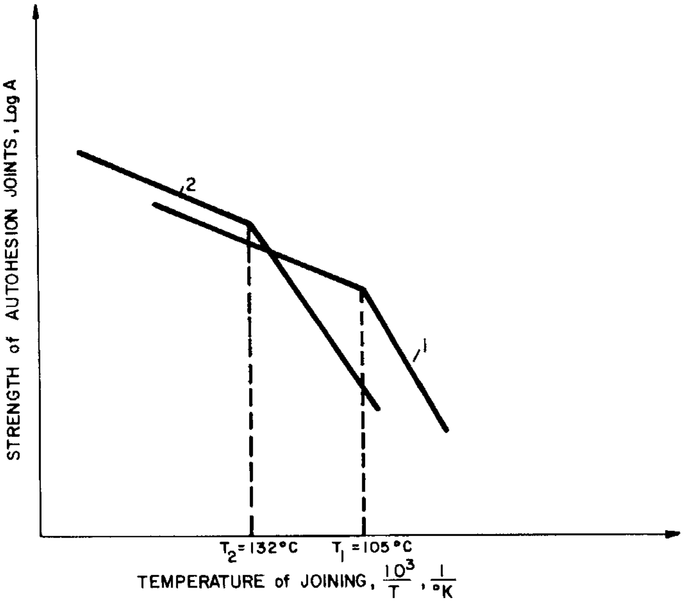

FIGURE 1 Dependence of strength of autohesion joints on joining temperature. (1) Low Density Polyethylene (LDPE); (2) High Density Polyethylene (HDPE).

all factors which accelerate or reduce the diffusion would accordingly increase or decrease the rate of autohesion process. Another limiting process had to be found if such correlation is not valid. Factors which increase the rate of diffusion are: raising the contact temperature, plasticization, and dissolution of the polymer. Increasing the molecular chain stiffness, the presence of strong polar groups, and crosslinking reduce the rate of diffusion. Many researchers have determined[15–22] that autohesion was limited by diffusion because the rate of the welding process varies according to the diffusion theory. Therefore the diffusion process usually limits the strength of welded joints when the plastic being welded is in the melted state and the contact surfaces do not have inert layers.

Convective Currents

The viscosity of melted polymers is too high to assume the presence of convective currents during the welding operation if the material being welded was not subjected to purposeful deformation. Welding of plastic pipes or articles having thin inert layers on surfaces often requires melt flow for obtaining sufficient joint strength. However, deformation of the plastic material under pressure is not a necessary result in formation of the convective currents. Deformation of polymer melts is not considerable during many types of welding and heat sealing operations. For example, heat sealing of polyethylene films, whose surfaces were covered with a monomolecular layer of gold, showed that such a barrier could prevent autohesion. The joining process in those experiments was carried out under a wide range of temperatures, times and pressures. Lack of autohesion testifies that deformation of the melted polymer was too small for destruction of the thin gold layer. The monomolecular layer could not prevent convective currents. Melt flow can be examined by cross-sectional cuts of welded joints which were formed from parts of the same plastic with different colors.

Recombination of Macroradicals

Recombination of macroradicals across the separation surface is one of the most difficult recorded processes. Besides, recombination is very hard to separate from diffusion of the macroradicals. This process was used most fully and effectively in chemical welding.[34, 35] Chemical welding is based on recombination of macroradicals which were formed as a result of degradation of special chemicals. Chemical bridges between two surfaces are the basis of strength for those joints.

Experimental data do not give information about the essential importance of macroradical recombination in welding of thermoplastics under normal conditions. Possibly the recombination takes place in the mechanism of autohesion together with diffusion of macroradicals during cold welding of polyethylene[28] or pressing of polymethylmethacrylate powder.[23–25]

Physical Interaction

The formation of physical interaction on the molecular level and the subsequent transition into diffusion was proposed as a possible mechanism of plastic welding by McKelvey and Strome.[36] Physical interaction is natural at real contact but the authors attach self-dependent importance to this process. They consider that instantaneous physical interaction can create strength of welded joints for producing special kinds of articles. This idea is probably valid in the case of full instantaneous contact of surfaces being welded. However under real conditions, formation of the contact surface is continuing in time and physical interaction can occur only in places where the contact is already obtained. However, after formation of the physical interaction process of interdiffusion starts then at least three processes occur simultaneously. Strength of welded joints is a complicated function of the rates of these processes. Results obtained from experiments on the durability of welded joints did not confirm the self-dependent importance of physical interaction. According to experimental data,[37,38] the durability of T-shaped welded joints can be described by the following equation:

$$t = t_0 \exp\left[\frac{U_0 - vp}{R}\left(\frac{1}{T} - \frac{1}{T_p}\right)\right] \tag{1}$$

where:

t = time to failure
t_0 = constant having the dimension of time
U_0 = activation energy of the process of failure
p = the load causing failure
T = temperature of the experiment, K
v = constant characterizing the physical structure of the material and the welded joint
T_p = the temperature at the point where the durability is independent of the load
R = the gas constant

Constant U_0 for T-shaped welded joints (which fail by peeling from the original surface of contact) relates to the type of bonds being broken down under the load. Therefore the value of the constant U_0 should be equal to the activation energy of destruction of the chemical or physical bonds created during the welding process. The activation energy of failure by peeling from the original contact surface was found to be the same as the activation energies of thermodestruction and mechanical failure of the polymer being welded. This means that destruction of chemical bonds is going on during the peeling of welded joints from the original contact surface. Joints were formed at a wide range of temperatures and times of welding. Some of the joints have had very low strength which is not sufficient for any article but the activation energy of failure remains the same.[37] Consequently, physical interaction is an important process in the welding mechanism but it does not create considerable strength of welded joints under real conditions, because other slower processes limit the influence of physical interaction, for example, formation of contact surface.

Of course, several of the named processes can occur simultaneously under specific conditions.

The first group of processes mentioned above can realize the joining of the materials being welded. But some subsidiary processes should occur to create conditions for carrying out the joining processes.

a) Formation of the Contact on a Molecular Level

Real contact on a molecular level should be obtained before any of the joining processes will start to occur. The area of real contact essentially depends on temperature, pressure and time of welding operation. Consequently, if the rate of increasing the contact area is less than the rate of diffusion or any other joining process, then under certain conditions the strength of the welded joints is limited by formation of real contact surface. The rate of formation of real contact surface is a function of the polymer viscosity described by the exponential equation of the Arrhenius equation type. Therefore formation of real contact can be described by the same type of equation.

The influence of welding parameters on area of real contact was observed on welded joints of plasticized PVC films formed at the heat stripe welding machine.[38] Different contact areas could be noticed after peeling of the welded joints at the initial contact surface.

b) Formation of Macroradicals

Several authors have found that under some specific conditions the joining process is not controlled by diffusion.[23-28] Formation of macroradicals sometimes limits the entire autohesion process and it does not matter if further radicals recombine or diffuse across the contact surface. A complicated dependence of autohesion on pressure and temperature was found during the pressing of polymethylmethacrylate (PMMA) powder. The degree of completion of the autohesion process was determined by the degree of transparency of the sample being pressed. Formation of the transparent samples was related to the stress relaxation.[26] Activation energy of the relaxation was calculated and, on the basis of its value, a conclusion was made that the mechanism of stress relaxation in PMMA consists of breaking chemical bonds. Besides, the correlation between degree of transparency and molecular weight of the pressed polymer samples was proved by experiment. The dependence of the molecular weight on pressure and temperature of pressing was also determined. Introduction of inhibitors into the polymer makes it difficult to produce transparent samples from PMMA powder. Inhibitors deactivate the macroradicals and prevent their recombination. These experiments have indicated the essential influence of formation and recombination of macroradicals on the autohesion process of PMMA powder subjected to pressing.

The influence of macroradical formation on strength of welded joints formed at low temperatures was researched by the process of cold welding of polyethylene.[28] Considerable autohesion interaction was achieved at room temperature under high pressure and deformation. But strength of the autohesion joints decreases if the temperature of welding is increased. Only at temperatures close to the melting point of polyethylene did the strength of joints start to rise again with increasing welding temperature. The author explains such complicated dependence by different mechanisms of autohesion at low and high temperatures. The autohesion is limited by the rate of destruction of chemical bonds at low temperatures but at high temperatures the diffusion of molecular segments limits the overall process.[28]

Two different limiting processes in the welding mechanism were also determined during the study of temperature dependence of the autohesion rate. Three polymers were tested.[29–31] The authors concluded that autohesion is limited by failure of the supramolecular structures at low temperatures and by the molecular diffusion at high temperatures. Such determination was made on the basis that temperature dependence of autohesion has a form of a broken straight line (*i.e.*, two lines with different slopes) in coordinates log A *vs.* $1/T$, where A is the strength of the autohesion joints. The temperature related to the break is higher than the temperature of the glass transition and very close to the melting points for all polymers tested. The activation energy of the process which limits the autohesion in the low temperature range is much higher than the activation energy of the process in the high temperature range. The activation energy of the low temperature process is related to withdrawal of the molecules from the supramolecular structures. This assumption does not seem valid, especially for high crystalline polymers like polyethylene. Another interpretation is that formation of macroradicals on the contact surface limits the rate of autohesion at low temperatures. Macroradicals are formed as a result of molecular chain destruction which is also a thermoactivated process and can be described by an exponential equation. Moreover, the activation energy of autohesion should be equal to the activation energy of thermodestruction of chemical bonds in the polymer. According to data obtained, the activation energy of the autohesion process[30, 31] and the activation energy of thermodestruction[32] coincide for both polyethylenes and polystyrene (see Table I). But the activation energy of autohesion is much higher than the activation energy of thermodestruction for plasticized poly(vinyl chloride).

The temperature dependence of autohesion has a break at the specific temperature which is near the melting point of the polymer. The authors[31] consider that the break relates to a change in the process which limits the rate of autohesion. Mobility of the molecular segments is so high at temperatures near the melting point that the number of segments available for diffusion is larger than the number of segments which can take place in diffusion. Therefore, diffusion limits the rate of the overall process and the strength of

TABLE I

Activation energy of autohesion (Refs. 30, 31) thermodestruction (Ref. 32) and mechanical destruction (Ref. 38)

Plastic material	Activation energy of autohesion in low temperature range (kcal/mol)	Activation energy of autohesion in high temperature range (kcal/mol)	Activation energy of thermodestruction (kcal/mol)	Activation energy of mechanical destruction (kcal/mol)
Low density polyethylene (LDPE)	60	5	63	63
High density polyethylene (HDPE)	64	5	63	63
Polystyrene	50	2–4	50–55	55
Poly (vinyl chloride) (PVC) + plasticizer	50–90	12–15	30–35	30–35

joints at the high temperature stage. Activation energy of diffusion is much lower than the activation energy of thermal destruction.

As discussed above, formation of macroradicals sometimes limits the autohesion process. Breakdown of the polymer macromolecules is going on with high activation energy. Consequently, this process appears limiting at low temperatures.

c) Destruction of the Inert Surface Layers and Removing Them from Welding Zone

Destruction and removal of the inert surface layers should precede the joining process because such layers prevent real contact of the active polymer. Destruction of thin surface layers can occur by mechanical vibration and impact during ultrasonic welding or by applying high pressure and by special design of electrodes in other welding methods. Surface layers can flow out from the welding zone if the melting point of these materials is lower than the melting point of the polymer being welded.[33] Sometimes, inert surface layers can be destroyed before the welding operation. Such treatment allows the obtaining of satisfactory strength of welded joints in cases where usual methods of welding are not effective.[39]

A review of the processes included in the mechanism of plastic welding shows that different processes can occur under various conditions. The strength of welded joints is dependent on the rate of the slowest process up to a certain level. But the most widespread mechanism under normal welding conditions (above the melting point of the polymer being welded) is:

1) formation of real contact
2) instantaneous physical interaction
3) diffusion

The strength of the welded joints and the type of failure as a function of welding technology were analyzed using the conclusion obtained.[38, 39, 41, 42] Testing of the strength of seams formed under different welding conditions showed that there are three possible types of failure of welded joints: peeling at the initial contact surface, tearing at the boundary of the seam, and partial peeling with tearing.[38] According to the theory, diffusion of the molecular chains or segments of the polymer through the boundary surface may continue until a cohesion structure is formed in the contact zone. It was assumed that peeling occurs in cases where the diffusion process during the welding operation does not reach completion, therefore, with an increase of temperature or time of contact the degree of completion of the process will increase. Under some conditions the strength of the seam in the welding zone is comparable with the cohesion strength of the polymer in the seam. The result of this will be that all seams welded at higher temperatures and longer contact times will not peel, but tearing of the material will occur. Thus, plastic films welded in a wide range of temperatures and times were tested and a change in the type of failure of the welded joints from complete peeling to peeling with tearing of the material was found. This change was related to completion of a certain stage of the diffusion process. It is known that diffusion in polymers is of an activation-kinetic nature. Consequently, the welding conditions which resulted in completion of the characteristic stage of this process was described by an equation of the type of the Arrhenius equation.

$$t = A_0 e^{Q/RT_w} \tag{2}$$

where:

t = is the time required for completion of a given stage of the diffusion process
A_0 = is a constant having the dimension of time, and depending on the thickness of the material
Q = is the activation energy of diffusion
T_w = is the welding temperature, °K [Eq. (2) is valid if the polymer is in the viscous-flow state]

The dependence of the strength of welded joints on the welding temperature was considered from the same viewpoint.[37] The degree of completeness of the diffusion process is of critical importance in peeling of the seam. Therefore, if the welding temperature is increased, the strength of the joint should increase to a value equal to the strength of the material in the seam. When this value is reached there is a change in the type of failure. A further increase in the welding temperature may increase the autohesion strength, but does not cause an increase in the strength of the joint as a whole, the latter strength being limited by the strength of the material in the seam. Therefore, the dependence of the strength of the joint on welding temperature should have a bend corresponding to the temperature of transition from peeling to peeling with tearing. However, the strength of the material of the seam decreases somewhat with an increase in the welding temperature. Consequently, the dependence may have a maximum at the temperature of change in the type of failure. The experimental data proved the correctness of the hypotheses and showed coincidence of the technological parameters of welding which resulted in a change of the type of joint failure and in the maximum of the joint strength. Consequently Eq. (2), which has a physical basis, makes it possible to establish the welding conditions which give the maximum strength of the welded joint.

Another way to verify the diffusion nature of the welding process was used. According to Eq. (1) given above, the constant v characterizes the physical structure of the material and the welded joint. Therefore, v in Eq. (1) is proportional to the stress concentration. Durability of the seams having failure by peeling from initial contact surfaces is determined by the number of molecular chains taking up the load over the contact surface; that is to say, those chains of molecules which crossed the interface during welding. The more advanced the process of diffusion, the greater number of molecular chains there are to be broken in the peeling of a welded seam, and consequently the lower coefficient of stress concentration there should be. The process of increasing the molecular chains during formation of the joint is essentially the same process as that described by Eq. (2). Therefore, it was proposed that constant v in Eq. (1), which is proportional to the stress concentration, should be inversely proportional to the coefficient of diffusion and should be an exponential function of temperature. Then for peeling seams:

$$v = v_0 e^{Q/RT_w} \tag{3}$$

where:

v_0 = is a constant of the welded joint
Q = is identical with constant Q in Eq. (2)

To check the correctness of Eq. (3), values of the constant v were used which were obtained in determination of the durability of the peeling seams welded at different temperatures.[40] The Eq. (3) is valid for welded joints

TABLE II

Activation energy of formation of the welded joints (Ref. 37)

Method of determination	Q, kcal/mol LDPE	Q, kcal/mol plasticized PVC
From data for the change of the type of failure	4.8	9.1
From dependence of joint strength on welding temperature	4.8	9.2
From dependence of constant ν in equation (1) on welding temperature	4.6	10.0
From data given in references 30, 31	5.0	12.1–14.8*

* Plasticizers used for PVC in experiments described in Ref. 31 and 37 were different and can explain variation in value of Q.

formed from LDPE and plasticized PVC. Activation energy of diffusion obtained from different experiments described above is given in Table II.

Values of the constant Q obtained from various experiments are very close and show the basic correctness of the hypotheses proposed. But these results still do not give an answer to the question—which process limits the rate of welding operation and strength of the welded joints? The same facts which were related to the diffusion process can be explained from the viewpoint that the rate of welding operation is limited by the rate of formation of the real contact surface.

Therefore, Eq. (2) can be used for calculating the parameters of welding technology.[41,42] However, the physical meaning of the constant Q in this equation is not clear so far and can be determined only by direct methods.

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