Microwave welding of thermoplastics

R. J. WISE, I. D. FROMENT *TWI, Granta Park, Cambridge, UK E-mail: ian.froment@twi.co.uk*

The aim of this work was to develop a new and versatile method for welding thermoplastics using microwave energy. A multimode cavity applicator was developed including features designed to deliver an even energy density and to apply weld pressure. A review of possible microwave susceptible implant materials was undertaken and results of welding trials using several candidate materials showed that this is a feasible welding technique. © *2001 Kluwer Academic Publishers*

1. Introduction

In the electromagnetic spectrum, energy in the frequency range 3×10^8 Hz to 3×10^{10} Hz is generally referred to as microwave energy. This wavelength range has found considerable use in industries such as cooking and drying [1] due to the response of water molecules to this excitation. In fact, the prevalent frequency for microwave cooking appliances is 2.45 GHz which has been selected as optimum for heating water in its free and bound states.

Microwave energy may be delivered to a material via several designs of applicator such as single mode applicators, multimode applicators, probes, antennae and lenses. A review of applicators can be found in texts such as Metaxas and Meredith [1] and Sisodia and Raghuvanshi [2].

The selection of the most appropriate applicator for any particular purpose is fundamental to the technical and commercial success of that application. Microwave welding of plastics has been developed as a versatile assembly technique suitable for exploitation in mass production industries such as automotive. For this reason the method of microwave energy input selected for microwave implant welding was by means of a multimode resonant cavity. In this case, a component is placed in a volume of space which is bounded by an earthed box from which no microwave energy can escape. Microwave energy is then launched into this volume and the energy distributes itself evenly, heating regions where microwave susceptible material is present.

This approach has one clear advantage over others using alternative applicator designs which is that it is capable of welding thermoplastic components of any shape. Provided that the thermoplastic is, to a first order approximation transparent to the microwave radiation in a multimode cavity, the only part to be heated will be a pre-positioned weld implant at the joint line.

When the energy is incident on the component, it propagates through the thermoplastic but is absorbed by the microwave susceptible implant causing heating (Fig. 1). As the temperature of this implant reaches the softening point of the surrounding thermoplastic, the material begins to flow across the joint and a weld is formed as the thermoplastic cools under pressure (with the microwave energy now off).

This approach embraces what is thought currently to be the most flexible, fastest and cost effective use of the microwave energy in a mass production situation.

The joint may have features in two or three dimensions but the microwave energy will still only heat the implant, thus creating a two or three dimensional weld. This makes the technique unique and is expected to give designers working in thermoplastics considerable freedom to generate components of increasing complexity in the future.

The success of the industrial exploitation of the process depends on a number of technical and commercial features including:

- Availability and cost of implant materials
- Equipment cost
- Ability to manufacture using this process
- Production rates
- Consistency of the welding process
- Joint strength

Central to many of these themes is the design, performance and interaction of the implant with respect to the production equipment and the parts to be welded. It is towards some of these issues that this work was directed.

2. Previous work

Some work in the field of the development of microwave welding of plastics is recorded in the literature. One development includes a five sided rectangular box into which microwaves are launched from an antenna [3]. Thermoplastic sheets to be welded are placed across the open side of the box and microwave energy is absorbed by the material placed at the joint line. This material could include copper powder, ferrous oxide, manganese oxide or carbon black. Microwave welding in this form is particularly suited to on site assembly of large scale plastic protective films such as those used to line the walls of an artificial lake.

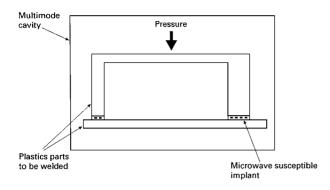


Figure 1 Schematic representation of equipment configuration for microwave welding using implants in a multimode cavity.

In another development, microwaves have been used to heat solvents for polyethylene [4]. Solvent welds in polyethylene have been made in this way in a multimode cavity.

Microwave welding of plastics has been reported where an implant of zinc oxide whisker is used as the heating medium [5]. Weld strengths are reported to be high and heating is said to occur at VHF, UHF or microwave frequencies.

A more sophisticated approach involving chiral materials as implants has been followed by Varadan and Varadan [6]. They report an approach where these specially formulated materials can themselves act as microwave susceptors or can be mixed in with the weld parent materials. Results of trials on ABS and polycarbonate show that joints with good mechanical properties can be achieved by this technique.

A welding technique for thermoplastics involving microwaves has also been developed by Piller [7] who uses "open and half open" cavities to apply microwave energy to plastics materials and suitable implants. Several implants are mentioned including fabrics with high electrical resistance, conductive polymers such as polypyrrole, polyaniline or polythiophene, or thermoplastic adhesives. This approach is said to produce uniform heating of the weld which could be particularly useful for joining long lengths of material such as floor covering, wall covering, roof covering and road laying.

Wu developed microwave welding in a multimode cavity using polyaniline as the implant material [8]. Welds in HDPE achieved over 80% of the strength of the unwelded parent material. He also welded both HDPE and nylon 6/6 using a single mode applicator and implants made from polyaniline. Weld strengths in nylon 6/6 exceeded 90% of the strength of the parent material.

3. Objectives

There were three main objectives of this work:

(i) To design and manufacture a multimode cavity applicator to permit even heating of welds in thermoplastic while under pressure.

(ii) To review possible microwave susceptible implant formulations and to manufacture samples of most likely candidate materials. (iii) To make and mechanically test microwave welds in several thermoplastics using the equipment and implant materials produced.

4. Equipment design and manufacture

A multimode cavity applicator, similar in conceptual design to a domestic microwave oven, was designed to permit even energy distribution throughout its volume. Two 1.1 kW magnetrons were positioned a certain distance diagonally away from opposite corners on the back wall of the cavity. Rotating metallic mode stirrers were placed a certain distance from the magnetron launchers to further randomise the direction of propagation of the microwave energy from the launchers. These devices randomised the direction of propagation of the microwave energy by providing a constantly varying angle of reflection for microwaves being launched from the magnetrons.

Weld pressure was supplied via an external pneumatic cylinder mounted on a rectangular frame. A thermoplastic rod then protruded into the cavity to supply pressure to the component being welded. Where the wall of the microwave cavity was breached to allow entry of the thermoplastic rod, a choke comprising a metallic tube of certain dimensions was clamped to the wall. The choke was designed to produce the correct orifice length to diameter ratio to avoid any leakage of microwave energy at 2.45 GHz.

The load capability of the weld pressure system was 0–4000 N. Jigging to accommodate single lap shear specimens was made in PTFE which has a very low loss at microwave frequencies. The door of the multimode cavity contained a panel of glass, for viewing the welding process, over which was placed some wire mesh. By this means microwave radiation was prevented from leaking through the glass. Around the edge of the door, a special microwave choke, made from slotted mild steel, was arranged to further minimise leakage. The safe closure of the door was ensured by two fail safe microswitches on the door catch.

The two 1.1 kW magnetrons feeding the multimode cavity could be run at any power level up to 1.1 kW each and an automatic timer device was also incorporated into the machine.

Power ratings for this machine were measured using a standard load of 3 litres of water. At full power, 1.1 kW was delivered by each magnetron to the cavity containing the load. Of this 1.1 kW, a certain proportion was delivered to the water load, the remainder being dissipated in the walls of the cavity or reflected back into the magnetrons.

Power levels quoted in the results of welding trials therefore strictly refer to equivalent water loads since the physical dimensions and electrical properties of weld implants would cause different amounts of power to be actually absorbed by these loads, the balance of power being dissipated in the walls of the cavity or reflected back into the magnetrons.

The prototype multimode cavity microwave welding machine is shown in Fig. 2.

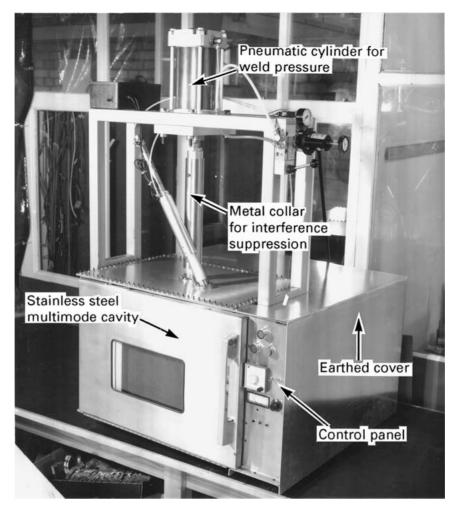


Figure 2 The 2.2 kW prototype microwave implant welding machine.

4.1. Safety issues

The main potential hazards associated with the equipment relate to exposure to microwave radiation or contact with high voltage. The UK recommended limit below which there is no proven risk to health is 100 W/m^2 of microwave energy at 2.45 GMZ. Levels of radiation around the cavity were measured and found to be undetectable using an Apollo leakage meter i.e. much less than 1 mW/cm². Accidental exposure to microwave radiation due to the door having been left open was thought unlikely due to the provision of a fail safe double microswitch on the door catch.

Magnetrons require a voltage of several kV in order to function. An earthed metallic cover was fabricated to protect personnel from accidental contact with components at high voltage.

5. Review of possible implant materials

5.1. Introduction

In order to research the available possibilities for introducing microwave energy to a joint in thermoplastics, all possible heating mechanisms were investigated. One of the more recent reviews of this field was given by Metaxas and included a summary of theory into each heating mechanism. These mechanisms can be divided into four broad groups, dielectric, conductive, magnetic and Maxwell-Wagner. It is possible that an implant could contain several materials each with a different loss mechanism. The possibilities are shown schematically in Fig. 3.

When a material is placed inside a multimode microwave oven operating at 2.45 GHz, a number of phenomena may occur as the microwave energy interacts with the material. If the material is a conductor of electricity, the magnetic field component of the microwave energy will try to generate eddy currents within the material. Depending on how good a conductor the material is, the nature of its geometry and its magnetic permeability, eddy currents will be generated to a certain depth in the material. If it is a very good conductor, eddy currents will only be generated in the surface of the material and the microwave energy will be largely reflected from the surface rather than penetrating into the material (e.g. copper). If it is a poor conductor, eddy currents will be generated to a greater depth and some energy may fully penetrate the material (e.g. carbon). In this case, less microwave energy will be reflected. If the material is an electrical insulator containing permanent dipoles having some ability to move, then the electrical field of the incident microwave energy will cause heating of the material. The microwave energy will penetrate through the material causing dielectric heating to a greater or lesser degree. There will, in general, be some microwave reflection from the surface of the insulator due to its dielectric properties.

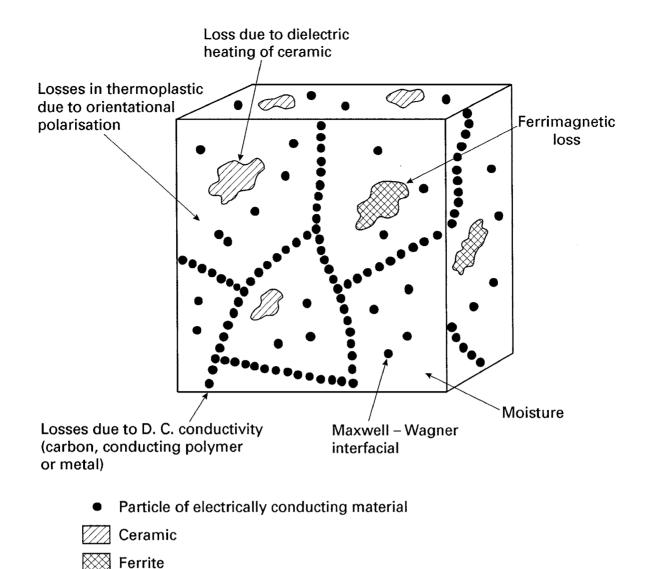


Figure 3 Possible microwave loss sources in an element of implant material.

If the material is ferrimagnetic, the magnetic field component of the microwave couples in to the magnetic moments of the ferrite particles. Characteristic losses can be calculated from knowledge of the complex permeability of the ferromagnetic material. If the material is an electrical insulator with little dielectric orientation (e.g. PTFE), the microwave energy will be unable to interact and will be transmitted through the material.

6. Dielectric loss

6.1. Thermoplastics

6.1.1. Previous work

A thermoplastic material may, in general, contain one or more homopolymer, some copolymer, an organic or inorganic filler, additives for things such as pigmentation or flame retardance, plasticisers and moisture or other environmentally absorbed substance. This section deals with dielectric losses due to microwave interaction with the homopolymer. There have been several reviews of this topic notably by Von Hippel [9], Tinga and Nelson [10], Bur [11] and Chen *et al.* [12, 13]. More general reviews of dielectric loss in thermoplastics at a wider range of frequencies have been given by Daniel [14], McCrum *et al.* [15], Hill *et al.* [16], Reddish [17] and Chen *et al.* [18].

In addition to these reviews, many papers have been written concerning the dielectric properties of individual thermoplastics at microwave frequencies [19–22].

6.1.2. Basic principles

When an electric field is applied to an insulating (dielectric) material, there will, in general, be some internal rearrangement within the material. The size of the change will depend on the material's ability to react but usually takes the form of a molecular alignment along the direction of the applied field. This is called polarisation.

The basis of the polarisation will generally involve movement of charged particles in the material to produce induced dipoles. The dipoles may be induced due to displacement of electrons around the nuclei (electronic polarisation), or displacement of atomic nuclei because of uneven distribution of charge in the molecule (atomic polarisation).

In fact, the electronic and atomic polarisations only occur at very high frequencies (infra red and visible) and, as such, play little or no part in the microwave heating process for thermoplastics.

The main mechanism responsible for dielectric heating of thermoplastic homopolymers is therefore orientation polarisation. However, in real situations this is likely to be complicated by additional heating due to the presence of electrically conducting particles and moisture (see below).

6.1.3. Theory

The theories for dielectric heating are complicated and dependent on many variables [1, 9, 14–17]. For the purpose of this work the following simple treatment is sufficient.

When an alternating electric field is applied to a thermoplastic homopolymer with a permanent dipole moment, at low frequencies (<100 Hz) it is likely that the orientation polarisation P within the material will be in phase with the change in the electric field E. At this point, there will be no heating of the thermoplastic. As the frequency increases, at some point the polarisation P will be unable to keep pace with the change in electric field E and a lag between the applied field and the polarisation will be evident. Now,

$$E = E_{\max} \sin \omega t$$

where E = the electric field strength (volts/metre), $\omega = 2\pi \times \text{excitation frequency (radians/second)}, t$ = time (seconds) and the polarisation

$$P = P_{\max}\sin(\omega t - \phi)$$

where ϕ is the phase lag (an angle) between *E* and *P*. Despite the fact that the thermoplastic is a bulk insulator, it is convenient to represent the lag as an electric current, given by:

$$\frac{\partial P}{\partial t} = \omega P_{\max} \cos(\omega t - \phi)$$

This current should be considered as a time dependent rearrangement of the charge distribution in the dielectric.

It is possible to calculate the power generated as a consequence of this phase lag:

$$P_{\text{average}} = \frac{1}{2} E_{\max} P_{\max} \omega \sin \phi$$

This equation is not very useful in this form because in general it is not easy to measure P. It can be shown [1] that the average power per unit volume,

$$P_{\rm av} = \omega \varepsilon_0 E_{\rm rms}^2 \varepsilon'(\tan \delta)$$

where ε_0 = permittivity of free space = 8.854×10^{-12} F/m, ε' = dielectric constant for the material and tan δ = effective loss tangent for the material.

$$\tan \delta = \frac{\varepsilon''}{\varepsilon'}$$
 and $\varepsilon = \varepsilon' - j\varepsilon'$

where $j = (-1)^{1/2}$ and ε'' is called the loss factor for the material. For any dielectric material, the important parameters describing the loss parameters are ε' and ε'' and these are the quantities which are often measured.

For thermoplastics, ε' and ε'' will, in general, be functions of both frequency and temperature. This means that during a microwave weld at constant frequency, the average power dissipated in the material will vary with temperature. This can be seen very well in Nylon 66 as reported by Huang [23] and as shown in Fig. 4.

Fig. 5 shows a graph of ε' vs log frequency for PMMA at room temperature and Fig. 6 shows ε'' vs log frequency at the same temperature [17]. Attempts have been made to show dielectric properties as a function of both temperature and frequency on the same plot by Reddish [17]. An example of such a graph can be seen

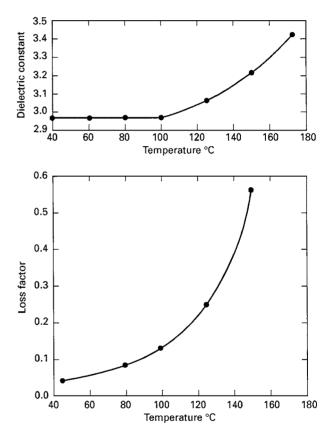


Figure 4 Dielectric constant and loss factor of nylon 6,6 at 3 GHz.

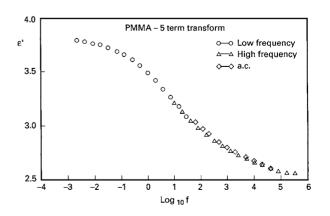


Figure 5 ε' versus log frequency for polymethyl methacrylate at room temperature.

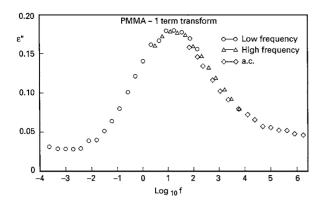


Figure 6 ε'' versus log frequency for polymethyl methacrylate at room temperature.

in Fig. 7, which shows ε'' vs temperature vs frequency for PMMA.

Reddish has produced similar plots for several other thermoplastic homopolymers [17]. These plots show a complex structure where the most important feature from the point of view of welding thermoplastics is the variation in loss as a function of temperature because this indicates that during the welding cycle the rate at which heat is generated in the material is not constant.

6.1.4. Relaxation time

From the explanation given so far, it is not obvious why there is a variation in orientation polarisation with temperature in thermoplastics. However, in general there are two major factors affecting the ability of materials to heat dielectrically. The first feature is the presence and size of the permanent dipole moment in the molecule. This determines whether or not a molecule will experience a force when an electric field is applied. PVC has a relatively large permanent dipole moment due to its polar molecular repeat unit. Conversely, polyethylene has a very small permanent dipole moment due to its non polar structure and experiences a very small force. Tables characterising these properties have been derived by Von Hippel [9] and Tinga *et al.* [10].

The other factor influencing the ability of materials to heat dielectrically is their ability to move within their surroundings. The way that this is measured is by a relaxation time which is characteristic of a polymer in a particular state. The relaxation time is a measure of how long the dipole takes to return to 1/e of its excited value. This is, in general, dependent on the viscosity of the polymer which is itself dependent on temperature.

In this way, thermal transitions such as T_g (glass transition) and T_m (melting point) can be seen in measurements of ε' and ε'' as a function of temperature in polar polymers [18]. The ability of some thermoplastics to heat dielectrically may also be affected by plasticisers since they will in general allow greater freedom of movement for the molecules. This is a complex topic and loss curves for thermoplastics in general require much interpretative effort.

Another way to envisage the dielectric heating mechanism is to consider the motion of individual segments of polymer chains. Below a certain temperature, called the glass transition point, T_g , segments of polymer chains are unable to change their conformation because there is insufficient free volume around them. As the temperature increases, a point is reached, T_g , where thermal expanison has created sufficient free volume around chain segments to permit changes

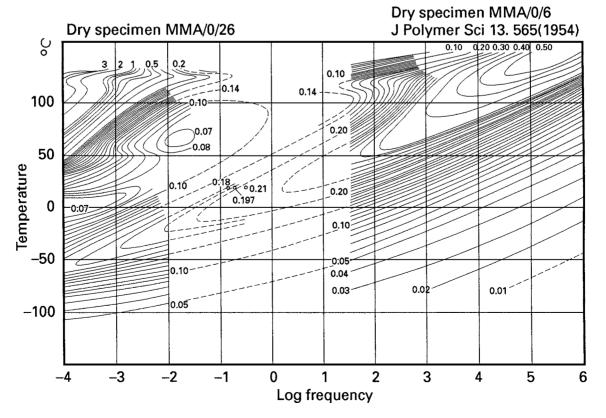


Figure 7 Contour map of dielectric losses of polymethyl methacrylate.

in conformation. The change in conformation usually takes the form of rotations around main chain chemical bonds causing that part of the polymer chain to take up a different position. The applied alternating electric field is able to directly cause such chain rearrangement at the glass transition temperature by acting on dipoles which would typically be on the scale of chain segments. Below the glass transition temperature, the applied electric field is unable to cause heating in this way because chain segments are not free to change their conformation. Since the motion of chain segments can be thought of as a measure of temperature, it can be seen how the application of a dynamic electric field can cause direct heating in certain polymers at their glass transition temperature.

This explanation is a gross over-simplification of the actual process occurring at the molecular scale, but serves to illustrate the principle of the dielectric heating mechanism. A comprehensive treatment of this mechanism is given by McCrum *et al.* [15].

6.1.5. Summary

It is possible to heat some thermoplastics using microwave energy due to a dipolar heating mechanism based on their permanent dipole moment. The degree to which they will heat also depends on a characteristic relaxation time which is generally a function of temperature. As a consequence, graphs showing dielectric loss factors against temperature are generally complex in structure making the design of a material suitable for a welding implant very difficult.

6.2. Ceramics

6.2.1. Previous work

The field of microwave heating of ceramics has not been fully developed but several case studies have been reported. A good overview has been written by Sutton [24] who describes the interaction of microwaves with ceramics in terms of a number of groups of materials. In one group lie the materials which readily heat up from room temperature, including Co_2O_3 and MnO_2 . In another group are ceramics which do not heat up in a microwave oven operating at 2.45 GHz until they reach a certain critical temperature. An example of this type of material is Al_2O_3 . Microwave heating of ceramics has been harnessed for the manufacture of refractories by mixing the 'lossy' ceramics with unfired material and then firing at high temperature in a microwave oven [25].

6.2.2. Basic principles

Some ceramics heat in a microwave oven in a similar fashion to dielectric polymers. Because these materials generally exhibit phase changes at much higher temperatures than thermoplastics, their dielectric properties remain more constant over the temperature range of interest for welding thermoplastics.

6.2.3. Summary

Implants for microwave welds in thermoplastics can be manufactured using blends of host thermoplastic and lossy ceramic material. The dielectric properties of these materials should remain relatively constant over the temperature range of interest, i.e. approximately 0–400°C. A formulation which can produce welds rapidly and is both commercially and environmentally acceptable has yet to be produced.

6.3. Water and other polar liquids *6.3.1. Previous work*

The dielectric properties of water were extensively reviewed by Hasted [26], and have been measured in many forms by researchers studying dielectric properties of foodstuffs containing moisture [1, 9, 10]. In the microwave region, the dielectric properties of free water and moisture are not simple and data require sophisticated interpretation.

Other polar liquids which may be useful to the welding process (for the production of solvent welds for example) should also be considered for their dielectric properties. Solvents such as methylmethacrylate (MMA), methanol, *N*-methyl pyrrolidone (NMP) and *m*-cresol are known to heat up readily in a microwave oven. As free liquids at room temperature they should not experience constraints on dipolar relaxation seen in some solid phases. Their interaction with the microwave radiation is only limited by their dipole moment.

6.3.2. Experimental data

A schematic graph showing the various contributions to the dielectric loss of a moist heterogeneous material [26] is shown in Fig. 8. The water content is responsible for peaks w and b where w is the loss peak due to free water and b corresponds to the bound water. In a material such as a thermoplastic, water may interact in one of two ways as it is absorbed. Some water will be held in pores or capillaries and remains chemically free or unbound. The remainder of the water will become chemically attached to the surface of the polymer by

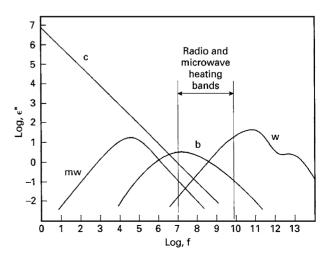


Figure 8 Loss factor of moist material as a function of log frequency.

hydrogen bonding or by another mechanism dependent on the chemical nature of the surface. The exact nature of the curve marked b will therefore be dependent on the surface under consideration.

Most domestic microwave ovens operate at 2.45 GHz which is not at the peak of the loss curve for free water which is at approximately 16 GHz. Instead this frequency has been chosen to lie mid way between the peaks in the curves for free and bound water as shown in Fig. 8 In this way, food containing a predominance of free or bound water will still experience a high degree of microwave heating.

6.3.3. Summary

Implants for microwave welding may contain some moisture or polar solvent which will be heated during welding. As this liquid approaches its boiling point, it is likely to evaporate from the substrate unless so deeply embedded in the material that superheating occurs. This latter effect could produce local porosity in the weld which is undesirable. Implants for microwave welding which depend on moisture content for microwave heating are unlikely to reach temperatures high enough to cause sufficient melting of the materials being joined. If they do cause melting of the parent materials, porosity in the joints is likely to be a problem due to superheating of the water trapped in the joint.

7. Conductive loss

7.1. Metals

7.1.1. Basic principles

It is well known that if metal objects with sharp edges are placed inside a microwave oven and the power switched on, some arcing from the edges will generally be observed, e.g. from a gold rim on a tea cup. The reason for this is that the free charges excited by the microwave fields in the surface of the metal will tend to concentrate at the sharp edges. This concentration of charge will produce a concentration of electric field at the sharp edge and when this electric field strength reaches a certain value, the air in the vicinity of the edge breaks down. This breakdown in the air is seen as the arc.

Arcing will generally involve some heating mostly due to the rapid motion of particles in the arc. However, for microwave welding, temperatures involved in this heating process are generally too high and cannot be controlled sufficiently to be of use. A system which does use controlled microwave generated arcing (or plasma) is described elsewhere [27].

Apart from arcing, microwaves will also interact with metal surfaces by causing eddy current heating. This is an interaction between the magnetic field component of the microwave energy and the metal and results in electric current being generated. This current can be sufficiently large to cause Joule heating and such could melt surrounding thermoplastic if used as an implant in a weld.

7.1.2. Theory

When microwave energy is incident on a good conductor, e.g. a metal, much of it is reflected. However, some of the energy gives rise to the generation of electric currents and losses can occur due to the resistive nature of the conductor.

The depth of penetration of currents into the conductor is given by the skin depth equation [28].

Skin depth
$$\sigma = \left(\frac{2}{\sigma \omega \mu}\right)^{1/2}$$

where skin depth, $\delta =$ depth at which the fields or currents have decreased to 1/e their magnitude at the surface (metres), $\delta =$ conductivity of conducting material (S/m), $\omega = 2\pi \times$ frequency of microwave radiation (rad/sec), $\mu =$ magnetic permeability of the conducting material (H/m).

It can be shown [1] that the power, P, per unit area dissipated in the conductor as a result of eddy currents is:

$$P/unit\,area = \frac{1}{2} \left(\frac{\mu\omega}{2\sigma}\right)^{1/2} |H_t|^2$$

where μ , ω and σ are as previously defined and H_t , is the magnitude of the magnetic field strength of the microwave tangential to the conductor.

7.1.3. Summary

Because the conductivity of metallic materials is generally high, much of the incident microwave energy is reflected. In addition, any sharp exposed metallic edges are liable to cause arcing which could result in problems with weld consistency. It is suspected that materials other than metals with electrical properties ideally suited to this application will be found.

7.2. Carbon

7.2.1. Previous work

Carbon black material has been mixed with uncured rubber prior to irradiation with microwave radiation for accelerated curing (vulcanisation). Early papers describing this work include those written by Ippen [29] and Meredith [30]. Ippen's work involved measurements of dielectric loss factors on various types of rubber filled with various carbon black types. Graphs summarising his results appear in Figs 9 and 10. Concentrations of carbon black were typically between 25% and 50%. Meredith developed the concept to a stage where equipment for continuous conveyer driven vulcanisation was built capable of processing 1000 lb of material per hour. Meredith also summarised Ippen's findings regarding microwave heating rate as a function of carbon black content [30]:

- Carbon black provides a route for Ohmic heating to occur by currents flowing through the particles.
- The contribution to heating rate is directly proportional to the proportion of carbon in the compound above a certain critical minimum carbon content.
- The contribution to heating rate is approximately inversely proportional to particle size.

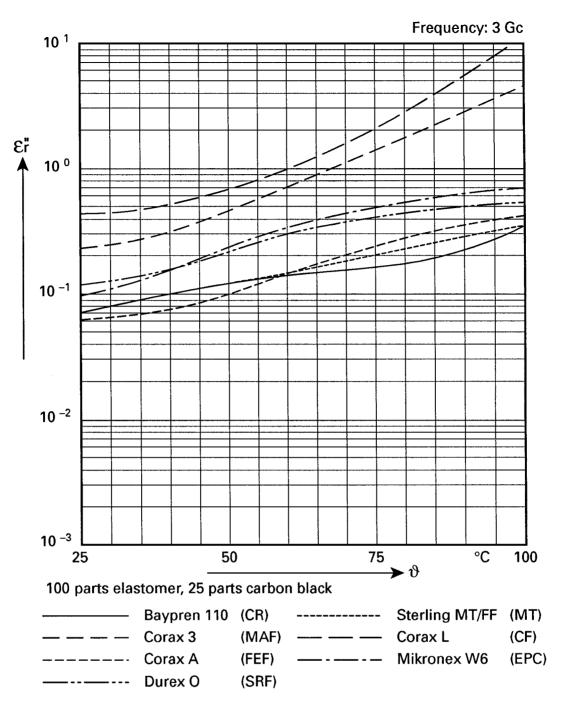


Figure 9 Loss factor as a function of temperature for various carbon blacks (25 parts carbon black, 100 parts elastomer) in polychloroprene.

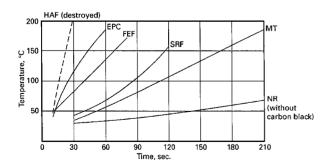


Figure 10 Dielectric heating of various carbon blacks (50 parts carbon black, 100 parts natural rubber) in natural rubber and of natural rubber without black.

7.2.2. Basic principles

If samples of thermoplastic (such as uncured rubber or polypropylene) containing different amounts of carbon black have their D.C. electrical resistively measured, samples with low carbon content have high resistivity. This is because each carbon particle is insulated by a layer of thermoplastic. As the carbon content increases, there will exist a thermoplastic to carbon mixture ratio where the D.C. electrical resistivity shows a sharp decrease. This is called the percolation threshold. For carbon black contents above this ratio the electrical resistivity will continue to decrease until the specimen is all carbon. The percolation threshold occurs when there is sufficient carbon to provide an unbroken path of electrical conduction all of the way through the material and is in general a distinct and well defined value.

In a microwave energy field, samples with carbon content below the percolation threshold may heat by:

- Dielectric loss in the thermoplastic
- Maxwell-Wagner loss

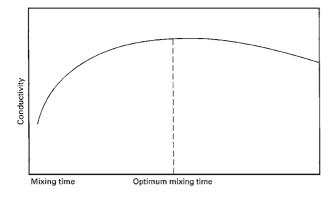


Figure 11 Electrical conductivity versus mixing time for carbon black in thermoplastic.

- Conduction loss in small zones of connecting particles
- Arcing between carbon particles

There are a number of other factors which will affect the heating rate such as:

- Particle size and shape
- Size of the applied microwave fields
- How well the carbon particles are dispersed in the thermoplastic

The dispersion of carbon particles is particularly important and a graph showing the effect of mixing time on conductivity [31] is shown in Fig. 11. In a semi crystalline thermoplastic, carbon particles will reside only in regions of amorphous material and so effects of thermal history (governing amounts of crystallinity) will be important to the electrical properties of the material.

7.2.3. Theory

Ippen's work involved the derivation of the dielectric loss factor ε'' for carbon filled material. It can be shown [29] that contributions to ε'' for a material may be made from conductive losses. In this case the contribution from conductive processes to ε'' is:

$$\varepsilon'' = \frac{\sigma}{\varepsilon_0 \omega}$$

where $\varepsilon_0 =$ permittivity of free space, $\omega = 2\pi \times$ microwave frequency and $\sigma =$ electrical conductivity.

This term can be added to other contributions to ε'' , such as that from the orientational polarisation. To give a total ε'' called $\varepsilon''_{\text{eff}}$.

The average power per unit volume is then calculated by the same formula as was used for the polar dielectric:

$$P_{\rm av} = \omega \, \varepsilon_0 \, \varepsilon_{\rm eff}^{\prime\prime} \, E_{\rm rms}^2$$

where this time $\varepsilon_{\text{eff}}''$ is dominated by the conduction term $\sigma/\varepsilon_0\omega$.

7.2.4. Summary

Carbon black has been used to aid the vulcanisation of rubber in proportions up to 50%. It is possible to calculate the electrical conductivity and average power dissipation of carbon black filled thermoplastics from measurements of ε''_{eff} . It should be possible to formulate implants for the microwave heating of thermoplastics using carbon black of a certain structure and percentage composition. This should offer a cost effective formulation using industry known processing routes.

7.3. Electrically conducting polymers *7.3.1. Introduction*

The existence of polymers which conduct electricity has been known for many years but it is only since the mid 1970's that interest in these materials has been rekindled. The reason for this was the accidental discovery of polyacetylene by chemists in Japan [32]. The material which they made had an electrical conductivity of less than 10^{-6} S/cm [33] which was later found to increase to almost 10 S/cm when dopants such as iodine [34] were added.

In a conducting polymer, the carbon atoms along the backbone of the polymer chain are connected by alternating double and single bonds (called conjugation). This structure can give rise to a delocalised electron, called a solition, which, under the action of an applied electric field can become a charge carrier moving along a chain.

When such polymers are doped, this introduces other charge carriers which increases the current carrying potential for the material [33, 35, 36].

The problem with polymers with a conjugated backbone structure is that they tend to be relatively rigid and intractable. Processing these materials from solution or by melting is therefore difficult or impossible and this is probably the main barrier limiting their widespread use.

Of the conducting polymers, polyaniline is probably the closest to being widely exploited on a commercial scale by virtue of its manufacturing cost and stability. Other conducting polymers which have been developed for potential applications include polythiophene, polypyrrole, polyphenylene and polyphenylenevinylene.

7.3.2. Previous work

The possibility of using polyaniline as an implant material for welding thermoplastics at microwave frequencies was first proposed by Epstein and MacDiarmid [37]. Later, several other groups have reported the use of polyaniline in microwave welds including Wu [8].

Electrically conducting polymers have yet to find a bulk application due to difficulties with their processability. Similar problems have been encountered with the application of these materials to weld joints. More recently electrically conducting polyaniline has become available in solvent [38] and has opened up the possibility of positioning the implant in liquid (by printing) or paste (by dispensing) form.

7.3.3. Basic principles

As an electrically conducting material, the same principles of microwave heating apply to polymers such as polyaniline as apply to carbon or metals. The main difference is at the molecular level where the fundamental charge carrying unit is different for the conducting polymer than for the other materials.

Electrically conducting polymers are often electrochemically synthesised into coatings on host polymers or as free standing films. The electrical properties of the conducting polymers may become highly anisotropic by the application of mechanical stresses which may cause orientation of the molecular structure.

There are versions of some electrically conducting polymers which are able to be processed in solution (e.g. polyaniline) but this is generally at the expense of suboptimal electrical properties.

It is generally possible to mix particulate solids of conducting polymer with thermoplastics to make blends whose electrical properties will depend on 'rule of mixture' and percolation theory as previously described.

7.3.4. Theory

Much of the theory relating to microwave loss in thermoplastic blends with carbon apply equally to blends containing electrically conducting polymer. As previously described, it is possible to define a component of ε'' (the loss factor) which is due to conductive loss. The term is:

$$\varepsilon_{\rm cond}'' = \frac{\sigma}{\varepsilon_0 \omega}$$

where, $\varepsilon_0 =$ permittivity of free space, $\omega = 2\pi \times$ microwave frequency and $\sigma =$ electrical conductivity. In the case of a blend of thermoplastic with conducting polymer, ε'' will generally comprise a term due to conduction and a term due to dielectric loss in the thermoplastic, i.e.

$$\varepsilon_{\rm eff}'' = \frac{\sigma}{\varepsilon_0 \omega} + \varepsilon''(\omega)$$

The power dissipated per unit volume in such a blend is then calculated from the formula:

$$P_{\rm av} = \omega \varepsilon_0 \varepsilon_{\rm eff}'' E_{\rm rms}^2$$

7.4. Ferrites

7.4.1. Previous work

Materials exhibiting high magnetic loss properties at high frequency can perhaps be divided into those which also conduct electricity and those which do not. Materials which conduct electricity are dealt with in the previous section. In the other group are materials such as ferrites which exhibit high losses at high frequency and have been harnessed for such purposes as drying plaster moulds [39] and curing of thermosetting materials [40]. A review of magnetic spectra of ferrites up to microwave frequencies is given by Rado [41].

7.4.2. Basic principles

Ferrite materials generally do not conduct electricity and are used in high frequency circuitry to give added inductance to coils without incurring losses due to eddy current generation. Objects made from ferrites are generally sintered from powder and are very hard and brittle in their finished form.

Losses will be incurred in ferrites due to hysteresis, domain wall and electron spin resonance effects. These losses are a function of frequency and temperature.

7.4.3. Theory

Losses in magnetic materials can be treated in a similar way to losses in dielectrics [1]. At high frequency,

$$B = \mu_0 \mu^* H$$

where B = induction field (Tesla), H = field strength (A/m), $\mu_0 =$ permeability of free space and $\mu^* =$ complex permeability.

$$\mu^* = (\mu' - j\mu_{\rm eff}'')$$

where $j = (-1)^{1/2}$, $\mu' =$ permeability and $\mu''_{eff} =$ magnetic loss factor.

It can be shown that [1] the average power per unit volume, P_{av} , generated in such a material in a microwave field is given by:

$$P_{\rm av} = \omega \mu_0 \mu_{\rm eff}'' H_{\rm rms}^2$$

where $\omega = 2\pi \times \text{microwave frequency and } H_{\text{rms}} = \text{rms}$ value of magnitude of a magnetic field strength.

7.4.4. Summary

It is possible to generate heat in ferrite materials in a microwave oven by harnessing the magnetic field component of the microwave energy. This has been done in the past for curing thermosetting materials and for drying plaster.

7.5. Maxwell-Wagner loss

7.5.1. Previous work

The Maxwell-Wagner or interfacial loss mechanism occurs at interfaces in heterogeneous materials, for example where a conducting phase meets a dielectric medium. Here a charge build-up can occur and dissipate energy if excited at high frequency. Studies of this loss mechanism have been made by Hamon [42] and Dryden and Meakins [43], who worked on copper phyhalocyamine in paraffin wax and water in wool-wax respectively. In both cases the peak in the loss factor was at approximately 10^6 Hz.

7.5.2. Basic principles

The conducting material in the dielectric medium certainly affects the polarisation of the material overall, particularly in the frequency range 10^5-10^7 Hz. The theory of the loss is complicated and comprises terms from both the dielectric and conductive parts of the material [42].

7.5.3. Summary

Maxwell-Wagner loss is a potential source of energy dissipation in heterogeneous materials excited at frequencies of over 10^5 Hz. The complexity of the theory of loss and the corresponding difficulty in formulating lossy material mean that it would be difficult to formulate a Maxwell-Wagner type implant for welding thermoplastics at microwave or high frequency.

8. Experimental

8.1. Materials

8.1.1. Parent materials

Welding trials were carried out on two materials, unreinforced polypropylene (Procom from ICI PLC) and transparent acrylic (Perspex[®] extruded acrylic sheet from ICI acrylics).

8.1.2. Implant materials

The following materials were employed as microwave susceptible implants:

- Non-woven polyester fabric coated with carbon, applied as a fabric strip implant.
- Indium-tin oxide (supplied by ERA Ltd) applied as a two part compound, mixed to make a paste and screen printed onto a glass slide. The material was dried at 180°C fired at 620°C. It was then physically removed from the glass slide and applied as an implant in powder form.
- Acrylic coated with gold by sputtering. The implant was therefore applied directly to the parent material in vapour form in a vacuum chamber.
- Acrylic specimens were coated with copper or iron from vapour in a vacuum chamber by evaporation of these metals using an electron beam.
- Five grades of polyaniline (supplied by the University of Durham) were employed in welding trials; 5–10% (weight) polyaniline in the solvent *N*-methyl pyrrolidone (NMP); 5–10% (weight) polyaniline camphur sulphonate in the solvent *m*-cresol; 5–10% (weight) polyaniline R6G hydrochloride in the solvent *m*-cresol; oriented polyaniline film doped with methane sulphuric acid.
- Implant materials based on polypropylene and filled with carbon blacks Vulcan XC72 or Vulcan P (supplied by Cabot) were prepared by injection moulding. Weight proportions of carbon varied from 18% to 30%.
- Implant materials based on acrylic (Diakon MG102) and filled with carbon blacks Vulcan XC72 or Vulcan P (supplied by Cabot) were prepared by mixing and extrusion to form granules. The granules were then further extruded to produce tapes with thicknesses up to 3 mm.
- Ceramics including lead dioxide (PbO₂), Chrome (III) oxide (Cr₂O₃), manganese dioxide (MnO₂) and cobaltic-cobaltous oxide (Co₃O₄) were applied as implants in powder form.

• Two manganese-zinc type ferrites were applied as implants in powder form. The approximate composition of these materials was Fe₂O₃ (50%), MnO and ZnO (50%). The two ferrit grades were supplied by Neosid Ltd and had the following properties:

	F5A	F10
Initial permeability, μ :	$2500\pm20\%$	$6000\pm20\%$
Curie temperature	200°C	130°C
Electrical resistivity	$100 \ \Omega \ cm$	$50 \ \Omega \ cm$

• The organic solvents methylmethacrylate (MMA), *m*-cresol, *N*-methylpyrrolidone (NMP) and water were all applied as implants in liquid form.

8.2. Welding procedure

Microwave welds were made in single lap shear configuration to ASTM 1002-72, that is, with an overlap area of 25.4 mm \times 12.7 mm. Implant materials were applied over this area, the joint assembled and the weld pressure applied. A weld pressure of 7.5 MPa was applied during welding in most cases and maintained for a predetermined time after the microwave energy had been switched off (usually 60 seconds).

Microwave welding power was applied in the range 440 W to 2200 W for times ranging from 10–120 seconds.

8.3. Examination and testing

Joints were visually inspected during the welding operation for signs of arcing and after welding for signs of uncontrolled or non-uniform heating patterns. Unless otherwise stated, single lap shear joints were produced and mechanically tested using an Avery Denison Universal Testing Machine (type 7152) at a cross head displacement rate of 5 mm/min.

9. Results

A brief summary of the results is presented in Table I.

9.1. Carbon loaded polyester tape

A graph showing results of single lap shear tests for microwave welds in acrylic using the carbon loaded polyester tape, is presented in Fig. 12.

Welds made in Acrylic using 2200 W of microwave heating power all failed in the parent material when tested giving failure loads up to 1.08 kN. In all welds, the implant could be seen to have broken up during welding. This was particularly noticeable in welds with heating times above 10 secs. The implants were torn apart as the molten material flowed under pressure. This is shown in Fig. 13. With the heating time at 25 secs,

TABLE I	Brief summary	of results of	microwave	welding trials

Mechanism	Implant material	Acrylic	Polypropylene
Dielectric	Ceramics	Heating trials only—lead dioxide best result	
	Solvents	Heating trials only	
Conductive	Carbon-Vulcan P/XC72	Not successful	Parent material failures in short heating times 5–10 secs
	Carbon polyester tape	Parent material failure, implant break up during heating gave visually poor weld	Failed in implant
Polyaniline	Metalised polymers	Too high conduction to allow bulk heating, i.e. all power reflected	
	Polyaniline	Parent material failure, solvent based weld	Did hot heat sufficiently to melt
	Indium-tin-oxide	Difficult to coat thermoplastics due to high drying and firing temperatures	polypropylene
Magnetic	Ferrite	Parent material failure—possible long heating times	Not attempted
Maxwell-Wagner		Not attempted	Not attempted

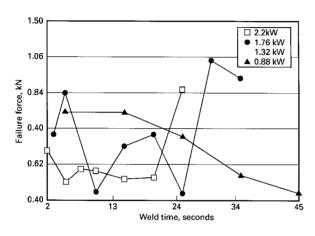


Figure 12 Results of single lap shear tests on microwave welds made in acrylic using carbon loaded polyester tape as an implant.

considerable arcing was observed around the implant during the final seconds of heating. Examination of the joint line after welding showed small areas of burnt acrylic.

Welds made at 1760 W microwave power failed in the parent material except when welded at 3 secs when failure occurred in the implant. For example when welded for 30 secs, failure load was 1.26 kN. No arcing from the implant was observed even with heating times up to 35 secs. At heating times above 15 secs the implant broke up during welding.

Welds made at 1320 W and 880 W microwave power failed in the implant when heated for less than 15 secs. At heating times greater than 20 secs the implant started to break up when welding at 1320 W and above 35 secs when heated at 880 W.

Welds made at 440 W microwave power failed in the implant when heated for less than 30 secs. At heating times over 30 secs the implant broke up during welding. Welds that failed in the implant gave failure loads of 0.59 kN when welded for 30 secs.

At all heating powers, where the failure occurred in the implant, the maximum failure loads achieved were higher than those achieved in parent material failures. This is a limitation of the test geometry.

A graph showing results of single lap shear tests for microwave welds in polypropylene using the carbon loaded polyester tape, is presented in Fig. 14. Results for lap shear joints made in polypropylene showed that all joints failed in the implant. Strengths up to 0.6 kN for the single lap shear joint were achieved at 2200 W microwave power with 20 secs heating time. The results showed that as the heating time was increased the weld strength improved. The results also showed that reducing the microwave power to 1760 W had little effect on the strength of the joints although some strength reduction was observed at 1320 W microwave power.

9.2. Indium tin oxide

The precoated glass sample heated rapidly in the microwave oven. The paste did not conduct electricity until it had been fired. Attempts to produce, in house, further ceramic samples coated with indium-tin oxide were not successful. Difficulties were experienced in reproducing the drying and firing temperature profiles. It was not thought technically feasible to produce implants by coating glass slides with indium tin oxide and then detaching the coatings for addition to the joint.

9.3. Gold sputter coated acrylic

The sample coated with gold for 1 min, heated insufficiently to produce melting of the interface. Slight arcing around the weld area was seen during the first 10 secs of welding. The samples coated with gold for 2 mins and 3 mins heated but not sufficiently to produce melting of the interface. Arcing around the weld area was seen during the first 10 secs of welding. The gold coating on the samples crazed during heating.

The sample coated with gold for 4 mins heated sufficiently to produce softening of the acrylic but was not sufficient to produce a weld. It was difficult to remove all of the gold from outside the weld area prior to heating. This gave the sample a poor visual appearance and some heating took place away from the weld. A lot of arcing was seen around the weld area during the first 20 secs of the weld cycle leaving small burnt patches at the interface. The sputter coated gold crazed during welding. This is shown in Fig. 15.

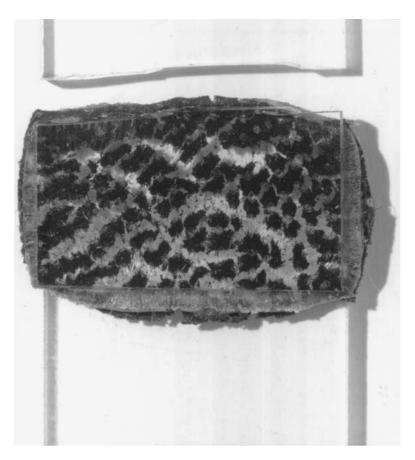


Figure 13 Acrylic with carbon polyester tape implant.

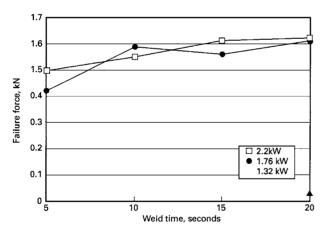


Figure 14 Results of single lap shear tests on microwave welds in polypropylene made using carbon loaded polyester tape as an implant.

9.4. Metallised polymers

A lap joint could not be produced between the acrylic sheets using the metal coated PET film. Microscopic examination of the weld interface after testing showed that there was no evidence of a joint forming between the metal interface and the acrylic but there was between the PET and acrylic. It was not possible to determine the mechanical strength of this joint. The samples of acrylic coated in copper and iron heated but no melting of the acrylic was observed in the 10 secs heating time before the heating cycle was abandoned due to severe arcing. The mirror surface of the iron coated specimen crazed during heating. This is shown in Fig. 16.

9.5. Polyaniline

Welds with parent material failure were produced in acrylic using polyaniline camphur sulphonate in *m*-cresol as the implant material. The results (Fig. 17) showed that as the microwave heating power was reduced, the time taken to achieve parent material failure reduced. For example, at 2200 W microwave power, parent material failure occurred with a heating time of 20 secs. Parent material failure loads were typically 0.5 kN. The heating time to achieve the same result was 50 secs with 880 W microwave power.

The effect of different solution application methods was to improve the appearance of the weld. With the solution dropped onto the interface using a glass pipette, for example, excessive heating occurred at the joint edges which resulted in a visually unattractive green weld flash. Visually, the best results were achieved when the polyaniline was deposited by paint brush or smeared with a tissue. In these cases, the interface was transparent.

Similar results were achieved in acrylic using polyaniline R6G hydrochloride in *m*-cresol solution (Fig. 18). This solution was red in colour. Again parent material failure occurred when welding at 2200 W microwave power for 30 secs. Parent material failure loads were typically 0.6 kN. The final visual appearance of the joint was poor compared to the pale green samples produced with polyaniline camphur sulphonate in *m*-cresol.

Attempts to produce conductive polyaniline by doping the polyaniline in NMP solution were not successful and no welds were produced.

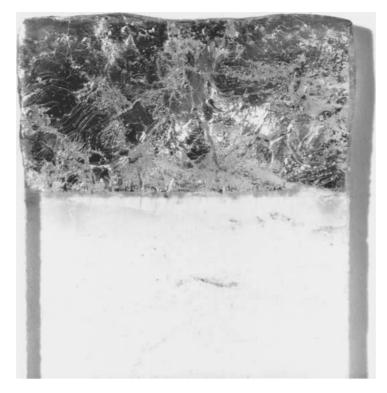


Figure 15 Gold sputter coated acrylic depicting crazing and material melt.

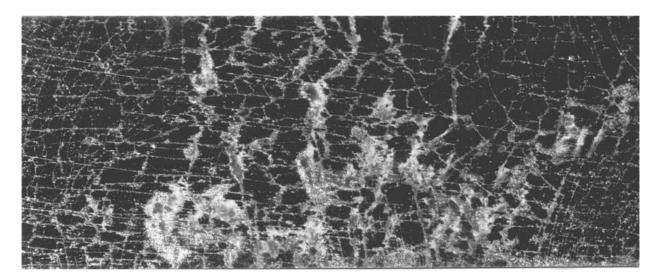


Figure 16 Electron beam deposited iron on acrylic.

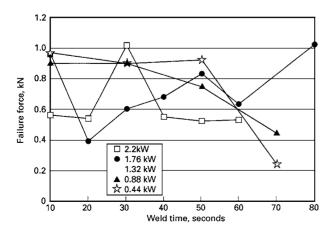


Figure 17 Results of single lap shear tests on microwave welds in acrylic using polyaniline camphur sulphate as an implant.

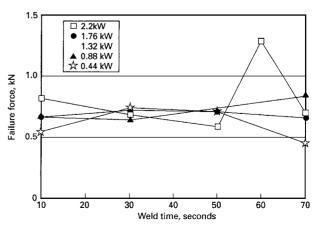


Figure 18 Results of single lap shear tests on microwave welds in acrylic using polyaniline R6G hydrochloride as an implant.

A single trial was attempted with polyaniline solution in a polypropylene joint. *m*-Cresol is not a solvent for polypropylene and therefore any weld would be by heating alone. Insufficient heat was generated to melt the thermoplastic.

9.6. Carbon filled materials

The most significant results in Vulcan P filled polypropylene implants were achieved with filler contents above 25% (Fig. 19). For example, at 25% Vulcan P loading, parent material failure was achieved at 5 secs heating time when welding at 2200 W microwave power. With a lower carbon content of 20%, parent material failure was only achieved with the relatively long heating time of 15 secs. In higher Vulcan P content (>25%) materials, overheating of the joint was regularly experienced, resulting in burning of the parent material. With 30% Vulcan P loading, high weld failure strengths (0.9 kN) were achieved using very short heating times (3 secs).

The best results in the thinner Vulcan P filled implants were achieved at heating times of 5 and 6 secs, where failure occurred in the parent material in both the 0.4 and 0.7 mm thick implants.

Similar results were achieved in Vulcan XC72 filled implants, where 25% filler gave the optimum results (Fig. 20). For example, welds gave parent material failure at 3 secs heating time. Other implants gave parent material failure above 6 secs for 22.5, 27 and 30% filler and above 9 secs for 18 and 20% filler. At a heating time of 9 secs, the 25% Vulcan XC72 filled implant had burnt significantly around the weld area, leaving a deposit of carbon.

Fig. 21 shows an example where burning of the parent material occurred in Vulcan XC72 with a filler content of 27%.

Welding trials conducted on the carbon black loaded acrylic were not successful. Although the acrylic heated in the microwave oven, there was not sufficient heat generated to allow the material to flow to form a weld. A number of factors govern the final conductivity of the composite material, all of which must be optimised individually [31]. These include thermoplastic type, car-

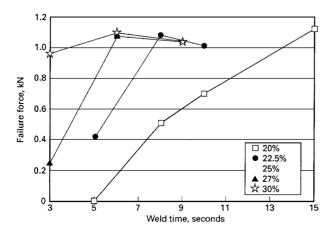


Figure 19 Results of single lap shear tests on microwave welds in polypropylene using polypropylene implants filled with Vulcan P carbon black and a welding power of 2.2 kW.

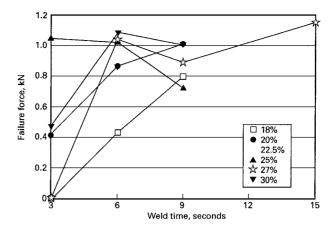


Figure 20 Results of single lap shear tests on microwave welds in polypropylene using polypropylene implants filled with Vulcan XC72 carbon black and a welding power of 2.2 kW.

bon type, percentage of carbon, surface area of carbon, surface chemistry of carbon and structure of carbon. Any one of these will affect the conductivity and may result in a failure to heat the material.

9.7. Powder ceramics

Of the powder ceramics tested for interaction with microwaves at 2.45 GHz, the strongest coupling (judged by estimation of temperature by operator) was achieved with the lead dioxide. The cobalt oxide heated to a lesser degree and the detectable temperature rise in the chrome(III) oxide and the manganese dioxide was negligible. Welds produced in acrylic using lead dioxide as the implant, failed in the parent material at the edge of the joint overlap.

9.8. Ferrites

Welds were made using the two grades of Neosid ferrite, F5A and F10. Welds made with the F5A ferrite in acrylic, produced failures in the parent material when tested in single lap shear mode. Parent material failure loads were 1.15 kN. Welds made using the F10 ferrite, failed in the implant material.

9.9. Solvents

Heating trials involving the organic solvents MMA, *m*-cresol, NMP and water revealed that they all interacted with microwave radiation at 2.45 GHz. Of the four liquids, the weakest interaction was observed with MMA while the other three solvents all interacted strongly with the microwave energy.

In the field of welding thermoplastics, these liquids could be used to create solvent welds in any polymer which is soluble in these solvents. Microwave heating could facilitate this process by causing a temperature rise in the solvents which would accelerate the polymer diffusion process believed to be the cause of the development of weld strength. Microwave assisted solvent welds were not attempted.

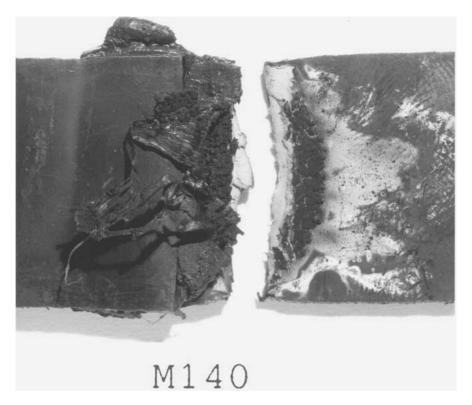


Figure 21 Polypropylene welded sample showing burning.

10. Implant formulation and introduction methods

10.1. Implant formulation

There are a number of attributes which must be optimised in the formulation of a successful implant for microwave welding of thermoplastics including heat generation, implant placement and joint mechanical integrity. Issues affecting heat generation have been discussed and those involved in implant placement are described below. The mechanical integrity of joints made using the microwave implant technique is a function of the composition of the implant because, as a part of the final structure, the implant contributes to the load bearing capability of the joint.

Depending on the physical form of the implant, the thermoplastic surrounding it may flow around and through it to cause molecular interdiffusion and hence weld strength. In this case, good adhesion between the implant material and the surrounding thermoplastic is not necessary and good joint mechanical strength will be achieved unless the volume of discrete particles in the implant exceeds the critical defect size required for rapid fracture. Implants falling within this category (Fig. 22) include fibrous or particulate materials (e.g. nonwoven fabrics and powders).

If the implant is continuous in nature, forming an imprenetrable laminate between the thermoplastic parts, welding of the parts may occur at the edges of the implant. Good adhesion between the implant and molten thermoplastic is required to ensure that the implant does not act as a large crack in the joint. (Fig. 23).

If the implant is composed of a composite of the thermoplastic being welded and fibres or particles of microwave susceptible material, strong joints will be made provided that the implant itself has good mechanical properties. Welding will cause molecular interdiffusion between the parts being welded and the thermoplastic matrix of the implant (Fig. 22).

10.2. Implant introduction

For the microwave implant welding technique to achieve commercial exploitation, cost effective and technically successful methods for placing the implant prior to welding are required. Other implant welding techniques, such as EMA weld [45] have sometimes been limited in their commercial exploitation due to difficulties in rapid implant placement. The optimum type of placement technology will depend on the implant type, parent material type and required production rate. Table I shows some of the forms in which individual implants could be produced and these may be important variables in the development of the production sequence for microwave welding of any component.

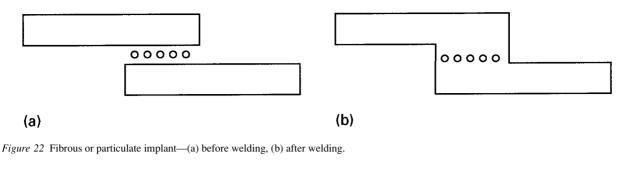
Since one of the main advantages claimed by the microwave implant welding process is that joints of complex geometry may be joined, the implant placement technology must also be capable of placing the implant along complex joints.

10.3. Possible implant introduction methods

10.3.1. Solid implants

Solid implants may be in the form of tape, powder, pellets, film or other. Some possible techniques for placing these material forms at the joint line include:

- In mould labelling technology
- Two part moulding



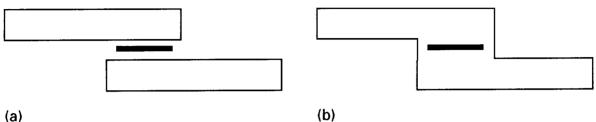


Figure 23 Laminar or film implant-(a) before welding, (b) after welding.

- Hot foil printing
- Spraying (powder)
- Sputter coating
- Metallisation by evaporation
- Friction surfacing
- Prewelding e.g. by hot gas
- Hot melt adhesion

It is technically feasible to introduce at least one of the solid implant types to the joint by each of these techniques. The selection of the optimum technique will depend on the selection of the optimum implant. This in turn will depend on the parent material, the weld geometry and the required production rate.

10.3.2. Liquid and paste (viscous liquid) implants

Some techniques for placing these material forms at the joint line include:

- Printing—automatic or manual
- Prewelding (deposition of a paste including a solvent for the parent material)
- Screen printing
- Dispensing from syringe (automatic or manual)
- Dispensing from tube
- Brushing
- Swabbing or application from absorbent material

10.4. Selection of implant introduction technique

The identified possible implant introduction techniques are listed in Table II together with some of their characteristics. This table could be used in conjunction with Table I to give guidelines for implant design.

For example, if an automotive component manufactured in polypropylene requires welding along a joint of complex shape, at a high production rate, consultation with Table I would tend to result in the selection of a carbon based implant for economic reasons. Liquid implants would not be recommended for polypropylene due to the low surface energy of this material and the limited choice of suitable solvents.

An implant introduction system would therefore be selected from the list for solid implants where automatic placement could be achieved in a complex joint. Possible processes could include in mould labelling, two part moulding, powder spraying or prewelding.

11. Discussion

11.1. Implant types and welding trials

The welding trials have shown that it is possible to make microwave welds using implants from the three main identified groups of loss mechanisms, i.e. conduction, dielectric and ferrimagnetic loss. Welding conditions vary from implant to implant but it has been possible to achieve good mechanical integrity in welds using each loss type. There was a good deal of scatter in the results of mechanical tests on specimens made using certain implant types, notably Figs 12, 17 and 18. This was attributed to transient discharging in the implants for certain combinations of welding parameters. This is generally an undesirable phenomenon because it degrades the polymer and leads to inconsistencies in weld strength. However, the occurrence of these events can easily be eliminated with the adoption of the most appropriate implant formulation and welding parameters.

The survey of materials which are lossy in microwave ovens at 2.45 GHz has shown that a considerable range exists. However, it is unlikely that one material type will be the optimum for all thermoplastic welds. On purely economic grounds, carbon is probably the optimum implant and the large range of carbon types available makes this a flexible option for technical reasons also. The correct formulation of carbon type and percentage content for each thermoplastic type is not an insignificant task. Carbon also suffers from one or two disadvantages such as colour, opacity and problems with application in liquid form.

Technique	Potentially manual (M) or automatic (A)	Implant material form	Comments
Tape or gasket	M + A	Solid	Automatic option very expensive
In mould labelling	M+A	Solid	
Two-part moulding	M + A	Solid	
Powder spraying	M + A	Solid	
Hot foil printing	M + A	Solid	
Sputter coating	А	Solid	Requires vacuum equipment
Vacuum metallisation	А	Solid	Requires vacuum equipment
Friction surfacing	M + A		Not widely used commercially
Prewelding	M + A	Solid + paste	Operator dependent
Hot melt adhesion	M + A	Solid	Parent material dependent for adhesion
Printing	M + A	Liquid	Parent material surface energy dependent
Screen printing	M + A	Liquid + paste	Parent material surface energy dependent
Dispensing from syringe	M + A	Liquid or paste	Parent material surface energy dependent
Dispensing from tube	M + A	Liquid or paste	Parent material surface energy dependent
Brushing	M + A	Liquid	Parent material surface energy dependent
Swabbing	M+A	Liquid	Parent material surface energy dependent
Spraying	M + A	Liquid	Parent material surface energy dependent

Welds with acceptable strength can be produced in short weld times, typically 15 seconds at 2200 W using the carbon impregnated polyester tape. This provides an attractive solution for welding complex geometries where the implant can be applied as a tape or pre-cut gasket. However, the breaking of the tape during welding leaves a visually unacceptable weld in the transparent acrylic and therefore would only be suitable for joints in opaque material. Indium-tin oxide would provide an opportunity to produce transparent welds in acrylic with very short cycle times. Current firing and drying temperatures make it impossible to join any thermoplastic materials due to their low melting points compared to the firing temperatures.

In the case of a metallic film implant, the metal deposited on the thermoplastic could act as a barrier between the samples being welded, preventing a polymer to polymer joint being formed.

The heating effect observed in the polyaniline solutions could have been caused by the polyaniline, the solvent or a combination of both. Subsequent control experimentation involving microwave heating of the solvent m-cresol and NMP revealed that they did heat independently. Therefore, the contribution that the polyaniline made to the heating effect was questionable. The two solvents for polyaniline were also believed to be solvents for PMMA and so the welding trials may have produced solvent welds accelerated by microwave heating of the solvents.

The practical significance of this project can perhaps be summarised as follows:

- A new welding technique exists for joining thermoplastics
- The technique has a number of unique features the most marketable of which is probably the ability to

weld complex joints or many components in one operation

- Possible implant types have been identified
- Possible implant placement technology has been identified

The commercial significance of this project is that an opportunity to exploit new technology exists and currently requires a marketing strategy to direct further development and industrial inception.

12. Conclusions

The following conclusions can be drawn from this project:

(i) Possible implant materials for microwave welding of thermoplastics have been surveyed and the best identified are probably those based on carbon black for commercial reasons. Polyanilines have shown good potential but require further development before commercial exploitation can be considered.

(ii) Welds have been made using implant materials identified in the survey and single lap shear strength data gathered. Failures away from the weld were recorded in many cases including with implants made from polyaniline and carbon black.

(iii) Possible implant introduction methods have been surveyed and applied on a laboratory scale.

Acknowledgements

Acknowledgements are due to Mr R. Hardy and Mr J. S. Littlechild who conducted the welding trials. The authors are also grateful to Dr A. Monkman of Durham

University for the samples of polyaniline, and to Dr G. E. Cooley of National Power for his contributions in the area of electrically conducting polymers. This work was funded by Shell Research SA, National Power Plc, ICI Acrylics and the Industrial Members of TWI.

References

- 1. A. C. METAXAS and R. J. MEREDITH, "Industrial Microwave Heating" (Peter Peregrinus Ltd., London, 1988).
- M. L. SISODIA and G. S. RAGHUVANSHI, "Microwave Circuits and Passive Devices" (John Wiley and Sons, New York, 1987).
- 3. F R Pat.No.2490057.
- 4. H. FUJIMATSU, S. OGASAWARA and S. KUROIWA, *Colloid Polymer Science* **268**(1) (1990) 28.
- 5. J P Pat.No.2261626.
- V. K. VARADAN and V. V. VARADEN, Polymer Engineering and Science 31 (1991) 7.
- 7. D E Pat.No.4036 876 A1.
- 8. C.-Y. WU, Ph.D. thesis, Ohio State University, 1996.
- A. R. VON HIPPEL, "Dielectric Materials and Applications" 1st ed. (MIT and John Wiley, Cambridge, MA 1954).
- W. R. TINGA and S. O. NELSON, Journal of Microwave Power 8(1) (1973).
- 11. A. J. BUR, Polymer 26 (1985) 963.
- M. CHEN, E. J. SIOCHI, T. C. WARD and J. E. MCGRATH, Polymer Engineering and Science 33(17) (1993).
- M. CHEN, J. W. HELLGETH, E. J. SIOCHI, T. C. WARD and J. E. MCGRATH, *ibid.* 33(17) (1993).
- V. V. DANIEL, "Dielectric Relaxation" 1st ed. (Academic Press, San Diego, CA, USA, 1967).
- N. G. MCCRUM, B. E. READ and G. WILLIAMS, "Anelastic and Dielectric Effects in Polymeric Solids" (Dover Publications, New York, 1991).
- N. E. HILL, W. E. VAUGHAN, A. H. PRICE and M. DAVIES, "Dielectric Properties and Molecular Behaviour" 1st ed. (Van Nostrand Reinhold, New York, 1969).
- 17. W. REDDISH, Bulletin of the Materials Science Club (1974).
- M. CHEN, E. J. SIOCHI, T. C. WARD and J. E. MCGRATH, Polymer Engineering and Science 33(17) (1993).
- W. MEYER, in Proc. Conf on Non Metallic Materials and Composites at Low Temperatures, July 1978 (Plenum, New York) p. 97.
- 20. S. OSAKI, J. Appl. Phys. 64(8) (1988) 4181.

- Idem., Journal of Polymer Science, C. Polymer Letters 28 (1990) 147.
- 22. Idem., Polymer 35(1) (1994).
- 23. H.-F. HUANG, Journal of Microwave Power 11(4) (1976) 305.
- 24. W. H. SUTTON, American Ceramic Society Bulletin 68(2) (1986) 376.
- 25. W. H. SUTTON and W. E. JOHNSON, US Patent 4219361, (1980).
- 26. J. B. HASTED, "Aqueous Dielectrics" 1st ed. (Chapman and Hall, London, 1973).
- 27. TWI Connect, September 1992.
- 28. P. LORRAIN and D. CORSON, "Electromagnetic Fields and Waves" 2nd ed. (W. H. Freeman and Co, New York, 1970).
- 29. J. IPPEN, Rubber Chemistry and Technology 44(1) (1971) 294.
- 30. R. J. MEREDITH, Journal of Elastomers and Plastics 8 (1976) 191.
- Conductive Carbon Black in Plastics, Cabot Corporation, Special Blacks Division, Boston, MA, Technical Reports S-39.
- T. ITO, H. SHIRAKAWA and S. IKEDA, Journal of Polymer Science, Polymer Chemistry Edition 12 (1974) 11.
- J. C. W. CHIEN, "Polyacetylene, Chemistry, Physics and Material Science" (Academic Press, San Diego, CA, 1984).
- 34. H. SHIRAKAWA, E. J. LOUIS, A. G. MACDIARMID, C. K. CHIANG and A. HEEGER, *Journal of Chemistry Society, Chemical Communications* (1977) 578.
- 35. T. A. SKOTHEIM, "Handbook of Conducting Polymers," Vol. I and II, (Marcel Dekker, New York, 1986).
- 36. R. H. FRIEND, RAPRA Review Reports 6 (1993) 3.
- 37. A. J. EPSTEIN and A. G. MACDIARMID, in Proc. Sixth Europhysics Industrial Workshop, 1990, Lofthus, Norway, edited by W. R. Salaneck, D. T. Clark and E. J. Samuelson (Adam Hilger, 1991).
- 38. Monkman A, Private Communication to R Wise, December 1994.
- 39. UK Pat.No.1481356.
- 40. Japanese Patent No.7428562.
- 41. G. T. RADO, Reviews of Modern Physics 25 (1953) 1.
- 42. H. V. HAMON, Australian Journal of Physics 6 (1953) 304.
- 43. J. S. DRYDEN and R. J. MEAKINS, *Proc. of Physics Soc.* London **70** (1957) 427.
- 44. R. TIDSWELL, ERA, Product Safety Data Sheet for TW1010 Electrode Paste
- 45. S. M. CHOOKAZIAN, in SPE ANTEC 1990, p. 1834.

Received 29 January 1999 and accepted 9 June 2000