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APPLICAZIONE DI ENERGIA NEL CAMPO DELLE MICROONDE IN PROCESSI INNOVATIVI DELL’INGEGNERIA CHIMICA E DEI MATERIALI

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Abstract

The objective of this PhD work is the investigation of a few applications of microwave heating in different fields of chemical engineering and material processing.

An analysis of two different chemical processes where microwave are employed as alternative heating system, has been carried out in this work: application of microwave to the remediation of soils by petroleum products and chemical vapour infiltration assisted by microwave to produce ceramic matrix composites. The reason of this choice was suggested by the interest in acquiring a wide knowledge on the different ways of interactions between microwaves and materials and the several potentialities of microwave heating in chemical engineering field.

Nowadays the application of microwave to the decontamination of soils is a well known application with still numerous potentialities to be investigated. The original of this study is concerned upon the results of the research relating to application of microwave-generated steam treatment on contaminated soils by linear hydrocarbons, mixture of them and a real petroleum product (kerosene). The purposes of the investigation were to verify the feasibility of a microwave treatment on contaminated soils from mixture of hydrocarbons or petroleum products as an alternative method respect to the conventional decontamination treatments. The previous knowledge of the temperature distribution in the irradiated soil is essential for the correct application of the treatment, then a mathematical model was developed to describe the heat and mass transfer evolution in a soil-water system during the microwave remediation. Results confirmed the necessity of an optimum moisture content in the soil in order to achieve a cost-effective use of the microwave irradiation in a feasible and efficient soil decontamination process.

On the other hand, work is already under way in the area of microwave chemical vapour infiltration. In literature a few examples are reported but only at laboratory level: an important characteristics of this research is that typical lab-scale technical solutions that are not suitable for industrial production plants, have been carefully avoided in order to easily carry out a scale up of this process. A new pilot scale microwave assisted CVI reactor was designed and built, in this work. Results obtained from the infiltrations carried out on preforms, not previously pre-treated, show the feasibility of the industrial scale-up of MWCVI process, with a satisfactory average weight increase respect to the initial sample in reduced microwave treatment

times. The silicon carbide deposition inside the sample was sufficiently homogeneous and compact, even if an evident inter-tow porosity was still present. The densification of the composites results deeper from the lower to the upper surface, because the gas reagent flux was not yet well optimised. The oxygen content in the MWCVI treated sample was considerably decreased respect the untreated fibres, therefore the plant was well-sealed and suitable in order to carried out operations in absence of moisture and air, which maybe responsible of silica and formation of compounds of silicon carbide affected by oxygen impurities.

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LIST OF ACRONYMS AND SYMBOLS

CENELEC: European Committee for Electromechanical Standardisation
CVI: chemical vapour infiltration
DBS: direct broadcasting satellite
DC: direct current
EIRP: effective isotropically radiated power
EMC: Electromagnetic compatibility
EMF: electric and magnetic field
EN: European Standard
ESD: electrostatic discharge phenomena
FD: frequency domain
FDTD: finite difference time-domain
FEFD: finite-element frequency-domain
FETD: finite element time-domain
IEEE: Institution of Electric and Electronic Engineer
IRPA: International Radio Protection Association
ISM: Industrial, Scientific and Medical
ITU: International Telecommunications Union
LSI: liquid silicon infiltration
MI: melt infiltration
MOL: line method
MOM: the moment method
MTS: Methyltrichlorosilane
MWCVI: microwave assisted chemical vapour infiltration
PIP: polymer impregnation/pyrolysis
RF: Radio-frequency
RFI: radio-frequency interference
RLC: resistance–inductance– capacitance
RMI: reactive melt infiltration (RMI or MI)
RMS: Root Mean Squared
TD: time domain
TESVE: Thermally-enhanced soil vapour extraction
TLM: transmission-line matrix
TWT: travelling wave tubes

VFMF: variable frequency microwave furnace
WHO: World Health organisation

ω = angular frequency
ε = complex permittivity
ρ = density
λ = latent heat of evaporation
δ = loss angle
ε'' = dielectric loss factor
ε'= dielectric constant
η_{SiC} =deposition efficiency of silicon carbide
α_t = the thermal diffusivity
$\dot{Q}_{abs}$ = generated heat by microwave
a = specific area
C = capacitor
c_p = specific heat
D = vapour diffusivity
D_p = Penetration depth
E_A = activation energy
f = vapour flow
I = current
k_0 = preexponential factor
k_c = general mass transfer coefficient
K_T = thermal conductivity
M = moisture
P = power dissipation
Ps = water partial pressure.
Q = Q-factor (ratio of energy stored in the cavity to the energy lost)
R = gas constant
T = temperature
t = time
V = voltage
X = moisture content
z = space coordinate

INTRODUCTION

The importance of minimizing the impact that chemical processing produces on the environment is growing. Optimal use of material and energy, and an efficient waste management can be recognised as important factors for environmental protection. Microwave processing of materials is a technology that can provide the material processor with a new, powerful, and significantly different tool with which to process materials that may not be amenable to conventional means of processing or to improve the performance characteristics of existing materials. Moreover, these technology is complex and multidisciplinary in nature and involves a wide range of electromagnetic equipment design and materials variables, many of which change significantly with temperature.

Microwave possess several characteristics that are not available in conventional processing of materials, including:

- Penetrating radiation
- Controllable electric-field distributions
- Rapid heating
- Selective heating of materials through differential absorption
- Self-limiting reactions.

These characteristics, either singly or in combination, present opportunities and benefits that are not available from conventional heating or processing methods and provide alternatives for the processing of a wide variety of materials, including rubber, polymers, ceramics, composites, minerals, soils, wastes, chemicals and powders. The characteristics of microwaves also introduce new problems and challenges, making some materials very difficult to process. First, bulk materials with significant ionic or metallic conductivity cannot be effectively processed due to inadequate penetration of the microwave energy. Second, insulators with low dielectric loss factors are difficult to heat from room temperature due to their minimal absorption of the incident energy. Finally materials with permittivity or loss factors that change rapidly with temperature during processing can be susceptible to uneven heating and thermal runaway. While the use of insulation or hybrid heating can improve the situation, stable microwave heating of these types of materials is problematic.

The most likely candidates for future production-scale applications will take full advantage of the unique characteristics of microwaves. For example chemical vapour infiltration of ceramics and solutions chemical reactions are enhanced by reverse thermal gradient that can be established using microwaves. Polymer, ceramic, and composite joining processes and catalytic processes are enabled by selective microwave heating. Powder synthesis of nanoparticles can take full advantage of rapid microwave heating to produce unique formulations and small particle sizes. Thermoplastic composite lamination and composite pultrusion processes are enhanced by rapid and bulk heating and by the ability to tailor the material's dielectric properties to microwave processes. The potential for portability and remote processing also make microwave processing attractive for waste remediation.

The objective of this PhD work is the investigation of a few applications of microwave heating in different fields of chemical engineering and material processing.

The research world (also industrial) is interested in microwave heating applications that regard particular sectors capable to create distinctive products difficult to produced by means of other technologies. In fact it is essential to considerate volumetric heating not as an alternative, but a way to obtain products and processes that take advantage of the specificness of this technique, eventually associated with other technologies.

Theoretical and experimental activity was focussed, in this work, on the analysis of two different chemical processes where microwave are employed as alternative heating system. In this way they could be enhanced not only with regard to saving time, but also improving the process or increasing the quality of the obtained products.

In this work two themes were examined: application of microwave to the remediation of soils by petroleum products and chemical vapour infiltration assisted by microwave to produce ceramic matrix composites. The reason of this choice was suggested by the interest in acquiring a wide knowledge on the different ways of interactions between microwaves and materials and the several potentialities of microwave heating in chemical engineering field.

Nowadays the application of microwave to the decontamination of soils is a well known application with still numerous potentialities to be investigated. The original of this study is concerned upon the results of the research relating to application of microwave-generated steam treatment on contaminated soils by linear hydrocarbons, mixture of them and a real petroleum product (kerosene). The purposes of the investigation were to verify the feasibility of a microwave treatment on contaminated soils from mixture of hydrocarbons or petroleum products as an alternative method respect to the conventional decontamination treatments. The previous knowledge of the temperature distribution in the irradiated soil resulted essential for the correct application of the treatment, then a mathematical model was developed to describe the heat and mass transfer evolution in a soil-water system during the microwave remediation. Microwave heating was performed on laboratory-scale experiments, measurement of temperature and residual moisture amount were carried out during the time. The agreement between the experimental values and the numerical results was appreciable both for the temperature and the moisture amount. Decontamination runs confirmed the necessity of an optimum moisture content in the soil in order to achieve a cost-effective use of the microwave irradiation in a feasible and efficient soil decontamination process. In particular 0.15 kg water/kg dry soil is an optimal condition to remove all the more volatile components of a commercial kerosene and almost of the 99% of the heavier compounds.

On the other hand, work is already under way in the area of microwave chemical vapour infiltration. In literature a few examples are reported but only at laboratory level: an important characteristics of this research is that typical lab-scale technical solutions that are not suitable for industrial production plants, have been carefully avoided in order to easily carry out a scale up of this process. A new pilot scale microwave assisted chemical vapour infiltration reactor was designed and built, in this work. Silicon carbide was infiltrated inside of pores that exist between the fibres tows by the decomposition at 1000°C of methyltrichlorosilane. Results obtained from the infiltrations carried out on preforms, not previously pre-treated, show the feasibility of the industrial scale-up microwave chemical vapour infiltration process. An average weight increase of about 70% respect to the initial sample was achieved in 18 hr of microwave treatment. The silicon carbide deposition inside the sample was sufficiently homogeneous and compact, with an evident inter-tow porosity was still present. The densification of the composites results deeper from the lower to the upper surface, because the gas reagent flux was not yet well optimised. The oxygen content in the treated sample was considerably decreased respect the untreated fibres, therefore the plant was well-sealed and suitable in order to carried out operations in absence of moisture and air, which maybe responsible of silica and formation of compounds of silicon carbide affected by oxygen impurities.

1 The Dielectric Heating

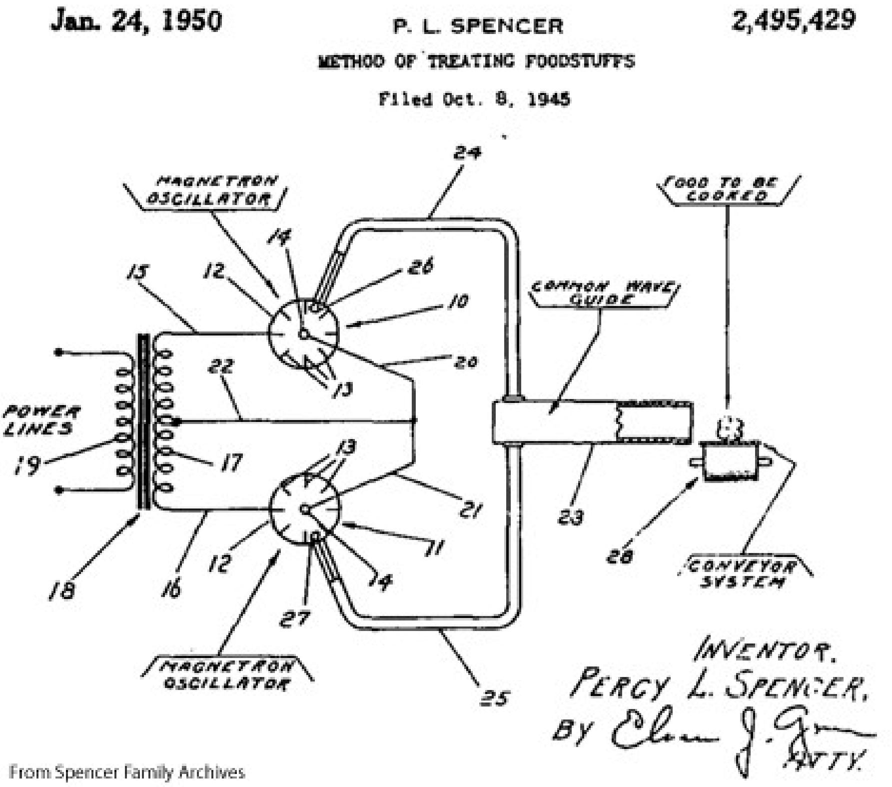
1.1 Introduction and fundamental concepts

In recent years, interest has grown rapidly in extending the application of microwave energy to the processing of a wide variety of new and engineered materials, including ceramics, polymers, composites, and chemicals. The growing interest is partly the result of increased awareness among materials scientists, processing engineers, and potential users of the many benefits of microwave processing. The use of microwave energy provides clean, rapid, and efficient heating over a wide range of temperatures (up to 2000 °C or more), as well as new degrees of freedom and flexibility over conventional processing methods. In some cases, microwave processing can synthesize new materials and microstructures that cannot be produced by other techniques.

Microwave processing is fundamentally different from conventional heating because electromagnetic energy is directly transferred to and absorbed by the material being processed. Consequently, this energy is converted into heat within the material and thus provides energy savings by eliminating the large thermal mass of conventional furnaces. Because microwaves are a penetrating radiation, materials exposed to them are heated from within and, when properly controlled, they can be heated more uniformly and rapidly than conventionally heated materials. This is particularly desirable for thick sections of low thermal-conductivity materials, such as polymers and ceramics. The penetrating radiation also gives rise to materials with interiors hotter than their surfaces. This method enables internal moisture to be removed from wet solids, as well as internal gases generated during binder burnout. Conversely, solid products of reactive gases can be deposited effectively into porous materials during chemical vapour deposition.

Because microwave heating mechanisms are different from conventional heating processes, microwave processing equipment and processing conditions often present new challenges. Employing recent advances in the understanding of microwave/material interactions is imperative for the successful processing of many materials and should be factored into the design of new microwave equipment.

Microwave processing first gained visibility in 1945, when the engineer Percy LeBaron Spencer, who worked for the Raytheon Corporation (Spencer, 1945) filed for patents for the heating of food with microwave radiation (Fig. 1-1). In 1952, Spenser (Spencer, 1952) received a patent for a conveyor microwave system, which led to industrialized microwave heating techniques. During the 1960s and 1970s, several companies pioneered the development of microwave equipment and applications for industrial uses, particularly for food products. It was not until the 1980s, however, that large-scale processes became commercially successful. To date, the major successes have been very limited and primarily include three large-scale applications: meat tempering, bacon cooking and rubber production. Smaller-scale applications can be found in the pharmaceutical, forest product, ceramics, textile, and chemical industries, mostly for the curing or drying of basic materials.



From Spencer Family Archives

Fig. 1-1 Picture of Original Microwave Oven Patent by Doctor Percy L. Spencer (Spencer, 1945)

In the area of processing advanced materials, only plasma generation for chemical processing and the curing of some polymers have yet progressed to the commercial stage in the United States. In Europe, however, the use of microwaves on an industrial scale is much more widespread, especially in the rubber industry.

The growth of microwave processing has been delayed for several reasons, including a limited choice of operating frequencies, a lack of understanding of microwave/material interactions, the reluctance of industry to accept the risk of a new technology and a lack of communication between microwave engineers and manufacturers. Each of these factors are changing, however. New developments in microwave devices are producing more versatile equipment for commercialisation, shown by the recently patented variable-frequency microwave furnace developed at Oak Ridge National Laboratories (McMillan et al., 1997) and the investigation of millimeter-wave devices at Los Alamos National Laboratories (Hardek et al., 1997). Moreover, improved data collection and modelling efforts are helping to illuminate the complex physical mechanisms of microwave/material interactions. As the influence of key processing parameters affecting the heating of materials with microwaves are better understood, process optimisation and control becomes more precise. Once materials can be engineered to meet industrial specification and customer needs, commercialisation should expand rapidly.

1.2 Electrical Volumetric Heating

By electrical means, volumetric heating is possible wherein all the infinitesimal elements constituting the volume of a workload are each heated individually, ideally at substantially the same rate. The heat energy injected into the material is transferred through the surface electromagnetically, and does not flow as a heat flux, as in conventional heating. The rate of heating is no longer limited by thermal diffusivity and surface temperature, and the uniformity of heat distribution is greatly improved. Heating times can often be reduced to less than 1% of that required using conventional techniques, with effective energy variation within the workload less than 10% (Meredith, 1998). Any material can be heated directly by electrical volumetric heating provided that it is neither a perfect electrical conductor nor a perfect insulator, implying that the range extends from metals to dielectric materials which could be considered quite good insulators. No single electrical techniques is effective in all cases and there are four methods used in practice, classified by the effective electrical resistivity and physical properties of the work piece.

1.2.1 Conduction and induction heating

These processes are used for heating metals with low resistivity and involve passing a heavy current through the workload to cause I^2R heating. The current may pass between physical electrical connections to the workload (conduction, or resistance, heating). The electrical frequency used ranges from direct current (DC) to 60 Hz. Alternatively the workload may form the secondary of a step-down transformer in which the induced electric and magnetic field (EMF) causes the heating current to circulate (induction heating), with electrical frequency of 50 Hz to about 30 kHz.

1.2.2 Ohmic heating

Ohmic heating is a conduction heating technique for liquids and pumpable slurries; it consists of equipment for passing an alternative current through the liquid between electrodes. Aqueous solutions, in particular, are almost always sufficiently conductive to permit a high power density to be dissipated, because dissolved salts provide ions as charge carriers. Ohmic heating invariably uses a power-frequency supply (50-60 Hz) and is extremely efficient as a converter of energy to heat in the workload, with efficiency of conversion over 95%.

1.2.3 Radio frequency heating

When the workload has high resistivity, the voltage required to pass sufficient current for a practical power-dissipation density becomes prohibitive at the low frequency used for conduction heating. This problem can be overcome by increasing the frequency to the range 1-100 MHz, most often 27.12 MHz, one of several internationally agreed frequency for the purpose.

Typical applications are plastics (welding and forming), wood (seasoning and gluing), textiles, paper and board (drying), food (post-baking/drying) and ceramics (drying). The workload is placed between electrodes in the form of plates or rods, to which is applied a high voltage (usually several kV) at the chosen high frequency.

Radio-frequency (RF) heating has been used in the industry since the 1930s and has grown to a substantial and important industry (Meredith, 1996).

1.2.4 Microwave heating

Intensive research during the Second World War into high-definition radar led to the development of microwave frequency (500 MHz to 100 GHz), and in particular the magnetron valve as a microwave generator of very high power output with exceptional efficiency (Meredith, 1996). In the post-war years further development resulted in microwaves being used for heating, especially for domestic purposes, but also significantly in industry, where there are some important advantages compared with processing at lower frequencies. Modern industrial-microwave-heating systems are used for a diversity of process in the food industry, tempering and thawing, continuous baking, vacuum drying, pasteurisation and sterilisation, and in ceramic, rubber and plastic industries, as well as many specialised processes in the chemical industry where there is great interest in vacuum processing.

Contemporary equipment has very high reliability and running costs are competitive with other heating methods, especially when the advantages of volumetric heating are included. Moreover high-power magnetrons, although initially expensive, are now rebuilt after normal end of life at a cost representing less than 10% of the energy they use. In industry, microwave heating is performed at either a frequency close to 900 MHz or at 2450 MHz, frequencies which are chosen by international agreement with the principal aim of minimising interference with communication services.

Most of the materials which can be heated at RF, can be also be treated at microwave frequencies together with some others which are difficult with RF because of their low loss factor. Because microwave heating operates at a much higher frequency than RF, the applied electric field strength is less, so the risk of arcing is less. The higher power density of heating can also be used, resulting in physically smaller plant. However the penetration depth is less at microwave frequencies than for RF, and, with the shorter wavelength giving greater prominence to standing waves, the uniformity of heating may be inferior. The overall efficiency of microwave heating systems is usually very high because of the exceptional efficiency of high-power magnetrons (85% at 900 MHz, 80% at 2450 MHz).

Because microwave frequencies have very short wavelength (33.3 cm at 900 MHz, and 12.2 cm at 2450 MHz), the electrical techniques used differ greatly from RF heating: RF equipment uses conventional electrical components such as inductors and capacitors, with open conductors for the electrical connections. Microwave equipment can not use these components because their size is comparable with the operating wavelength; under these conditions the components behave anomalously and the circuits would radiate most of the energy into space. Instead microwave heating uses waveguides (hollow metal tubes) to convey power from the magnetron(s) to the heating oven, frequently called the applicator. The applicator may have many forms but is almost invariably based on a closed metal stricture, with an access door or small open ports to allow the workload to pass through in a continuous flow.

1.2.5 The electromagnetic spectrum

Fig. 1-2 shows, on a logarithmic frequency scale, the frequencies of the electro heating techniques relative to those of other users. Frequencies allocated for electro-heating are often designated ISM “Industrial, Scientific and Medical” frequencies. The frequencies chosen for RF and microwave heating are the result of historical evolution and a complexity of international committees which constantly review the use of the electromagnetic spectrum.

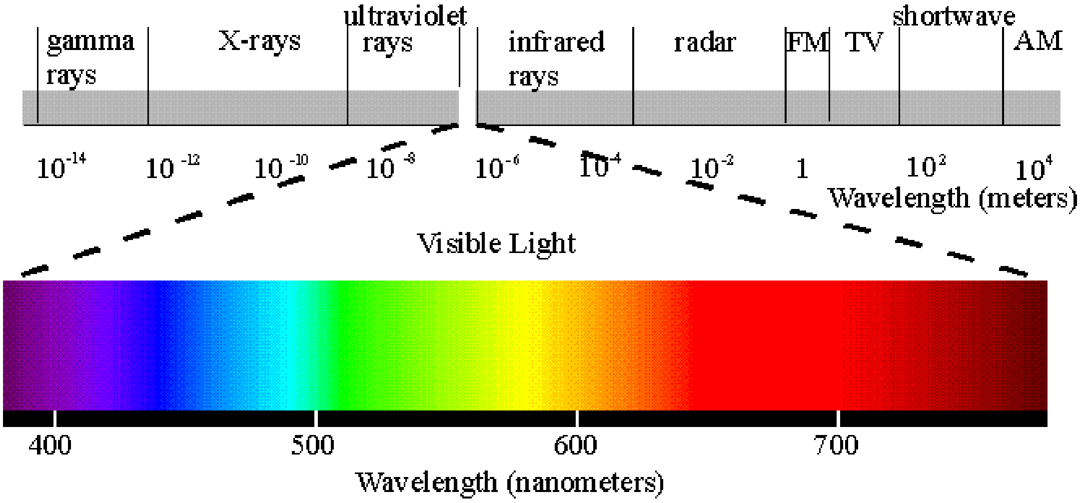


Fig. 1-2 The electromagnetic spectrum

All the electromagnetic techniques described above have particular operating frequencies for the electrical energy applied to the workload. This energy, which comprises combined electric and magnetic fields, is the same as that used for broadcasting, television, radar and satellite communications. Clearly great care must be taken to avoid interference between the electro-heating systems and these services, which essentially radiate and receive signals from locations spaced far apart. Whereas in communications the purpose is to radiate power into space, in electro heating the power is often generated at much higher power levels, but must be contained within the treatment equipment. Indeed the amount leaking away has to be controlled to within specified limits for safety of workers, and to avoid radio-frequency interference (RFI) to other services.

1.2.6 Electromagnetic compatibility regulations

Electromagnetic compatibility (EMC) is the ability of equipment or systems to function satisfactorily without introducing intolerable electromagnetic disturbances or without being disturbed to some extend. When EMC is achieved, components, devices and systems can perform their tasks according to specifications.

The European EMC standard deals with the safety of workers and the general public when exposed to EMF such as generated by heating equipment. The practical uses of electromagnetic energy at microwave and radio frequencies have been almost exclusively in the category of processing of information or communication, e.g., broadcasting, communication radar, etc. Only in the last half-century has there arisen significant "power applications" in which microwave energy is used to interact with materials for some end benefit, e.g., microwave diathermy or microwave heating. Recognition of this use of the electromagnetic spectrum was slowly recognized (Allen and Garlan, 1962) by authorities who regulate the use of the spectrum. These authorities, the Federal Communications Commission (FCC) in the United States, and the ITU worldwide ITU (International Telecommunications Union) worldwide, set aside specific bands of frequency, i.e., industrial–scientific-medical (ISM) for these uses. Allen and Garlan (Allen and Garlan, 1962) describe these actions as implementing a philosophy "*to provide frequencies on which unlimited radiation would be permitted and to prescribe severe limitations on radiation on other frequencies*". This was in recognition of the aspect of low cost and other factors inherent in economical ISM businesses.

1.2.7 Radiated emissions

The standard EN 55011 (BS EN 55011, 1998) details the limits of the radiated emissions of electromagnetic heating equipment. Unlimited radiation is allowed in some frequency bands, the ISM bands. ISM stands for science and industry. It means industrial, scientific, medical and domestic applications of electromagnetic energy or non telecommunication or non information technology applications of this energy. The standard designates the ISM bands, see Table 1-1 for the centre frequencies of these bands.

Table 1-1 Frequency Allocation of ISM Applications (Osepchuk, 2002)

Frequency [MHz]	Region	Conditions
6.765-6.795	worldwide	Special authorization with CCIR[c] limits both in-band and out-of-band
13.553-13.567 26.957-27.283 40.66-40.70	worldwide	Free radiation bands
433.04-434.79	Selected countries in Region I[a]	Free radiation band
433.05-434.79	Rest of regions I[a]	Special authorization with CCIR[c] limits
886-906	U.K. only	In-band limits
902-928	Region II[b]	Free radiation band

$2.40\text{-}2.50*10^3$	worldwide	Free radiation band
5.725-5.875	worldwide	Free radiation band
24.0-24.25	worldwide	Free radiation band
61.0-61.5 122-123 244-246	worldwide	Special authorization with CCIR[c] limits both in –band and out of-band

[a]Region I comprises Europe and Parts of Asia; the selected countries are the Federal Republic of Germania, Austria, Liechtenstein, Portugal, Switzerland and Yugoslavia

[b]Region II comprises the Western hemisphere

[c]CCIR="International Radio Consultative Committee" of the International Telecommunications Union (ITU)

1.2.8 EMC and human safety

As microwave power levels for industrial processing systems increase, potential hazards associated with exposure to radiation become important. Extensive work, as summarized in earlier review articles (Michaelson and Lin, 1987), indicate that the effects on biological tissue from exposure at microwave frequencies are thermal in nature. Unlike the higher energy, ionising region of the electromagnetic spectrum, including X-rays and γ-rays, the nonionizing bands from DC to visible light do not carry enough energy to break chemical bonds (Redhead, 1992). The only effects of nonionizing radiation in the microwave region on human tissue are those derived from the energy-matter coupling mechanism, particularly dielectric coupling. At present, the only confirmed effect is warming, from the conversion of electromagnetic energy to heat. Thus, microwave exposure standards are based on the thermal effects of exposure.

The currently accepted standard is the guidelines developed by the American National Standards Institute of 10 mW/cm^2 power density for exposure (ANSI C95.1-1991). The power density guideline is based on a maximum permissible exposure of 0.4 W/kg specific absorption rate, which is a factor of 10 less severe than the determined threshold absorption level (Redhead, 1992). Standards based on the institute's guidelines include the Food and Drug Administration's emission standard of 5 mW/cm^2 at 5 cm for microwave ovens (HHS, 1991) and the Occupational Safety and health Administration's exposure standard of 10 mW/cm^2.

For continuous whole body exposure (from 6 min and on) the reference levels at the important ISM frequencies for dielectric heating are shown in Table 1-2.

Table 1-2 Reference levels at the important ISM frequencies for dielectric heating (ANSI C95.1-1991)

Frequency [MHz]	**Workers [W/m²]**	**Public [W/m²]**
27	10	2
915	23	4.6
2450 GHz	50	10

To minimize exposure, the microwave system needs to be designed with effective leakage suppression, viewing or ventilation screens, and an interlock system on doors and access apertures to shut off power when doors are opened.

1.3 Microwave interaction with dielectric materials

In order to be able to heat via microwaves materials have to couple to the microwaves, i.e. they have to absorb electromagnetic energy. Basically three mechanisms are being applied here. The generally best-known mechanism is the excitation of a molecule covered with dipole moment. This is especially the case as concerns water, but also for a lot of solvents disposing of a characteristic dipole of their molecules. The molecules always try to align in the direction of the field lines in the quickly changing electric field, as shown in Fig. 1-3 and are thus brought to rotating swings. The energy absorption from the microwave field is the more intense the nearer the resonance rotation of the molecule lies at the frequency of the microwave. As far as water is concerned this would be a value near 15 GHz, which would, however, mean a very low penetration depth into the products and have the disadvantage not to heat up the product deeply enough.

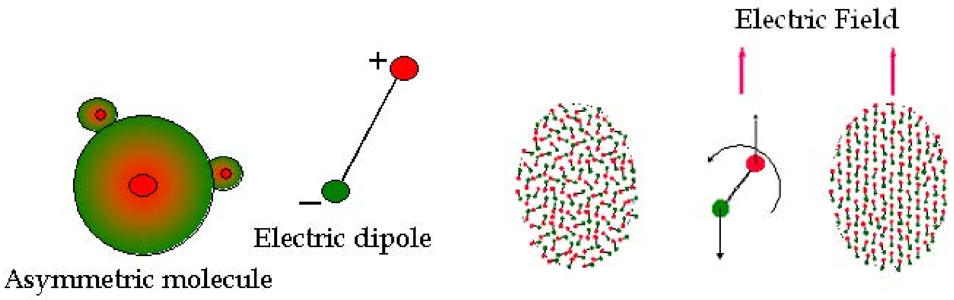


Fig. 1-3 Asymmetric molecule and its orientation respect to the electric field

Heating in the electromagnetic field also happens, if there are free ions in the product. This sort of heating takes place in electrolytes, but also in many glass or ceramic materials.

A third possibility of coupling is the magnetic absorption, however, having little importance in industry apart from the fact that such materials are often being used in absorptive locks for a damping of microwave energy. Well conducting materials such as for example metals also interact with the microwaves; in this case the penetration depth of the microwave is, however, only a few micrometers thus almost no energy is being absorbed from the microwave field due to the small resistance of these materials, i.e. the microwaves are being reflected at the metallic surface.

The ability of any material to absorb microwave energy is being expressed by its dielectric loss factor which is always combined with the dielectric constant. This dielectric loss factor depends on the type of material, frequency and temperature.

1.3.1 Relative permittivity

Relative permittivity (ε') is a dimensionless number which for a loss less homogeneous material is the same in all three rectangular coordinate directions (Metaxas and Meredith, 1983). It is simply related to refractive index for optical materials, and analogously with optics the lines of electric field entering the surface of the dielectric material are refracted (i.e. direction changed as well as magnitude) and also partially refracted. Other important related effects are the change in propagation velocity of a plane entering the material and the change in characteristic impedance.

Relative permittivity is a multiplying factor by which the capacitance of a vacuum capacitor would be increased if instead it were filled with a dielectric material of that permittivity; but it says nothing about the power absorption of the material.

Simple alternative current (AC) theory shows that a capacitor of value C connected to a sinusoidal generator of RMS (Root Mean Squared) voltage V at angular frequency ω, will draw a current I in the Equation 1-1:

$$I = j \cdot V \cdot \omega \cdot C$$ **Equation 1-1**

The operator symbol j signifies that the sinusoidal current leads the voltage by precisely 90°, and because there is no component of the current flowing in phase with the voltage there is no net power dissipation. Consider now the capacitor C as a parallel-plate capacitor for which the capacitance value is well-known and given by the Equation 1-2:

$$C = \frac{A\varepsilon_0\varepsilon^*}{d} \qquad \textbf{Equation 1-2}$$

where ε^* is the relative permittivity of the dielectric material filling the space between the plates, A is the area of one plate and d is the spacing between the plates. Supposing the permittivity having an imaginary component, it is could be written as the Equation 1-3:

$$\varepsilon^* = \varepsilon' - j\varepsilon'' \qquad \textbf{Equation 1-3}$$

Then the current I is expressed in Equation 1-4:

$$I = V\omega\frac{\varepsilon_0 A}{d}\left(j\varepsilon' + \varepsilon''\right) \qquad \textbf{Equation 1-4}$$

The first term in parenthesis shows the component of current in phase quadrature with the voltage as it would be with a loss loss capacitor , but the second term is a component in phase with the applied voltage, and therefore representing power dissipation. This power dissipation P is given by the Equation 1-5:

$$P = Re(VI) = V^2\omega\frac{\varepsilon_0 A}{d}\varepsilon'' \qquad \textbf{Equation 1-5}$$

The term ε'' clearly quantifies the power dissipation in the capacitor having a dielectric filling with relative permittivity defined by Equation 1-5 and it is called the "loss factor" of the dielectric.

The "loss angle" δ is another term used to quantify the lossyness of a dielectric. This the angle by which the resultant current differs from the ideal 90° phase angle relative to the voltage and it could be determined from the Equation 1-6:

$$\delta = tan^{-1}\frac{\varepsilon''}{\varepsilon'} \qquad \textbf{Equation 1-6}$$

1.3.2 The dielectric heating equation

Substituting in Equation 1-5 the volume of dielectric material between the plates: $V = Ad$, the voltage stress in the dielectric: $E_i = V/d$, the total power dissipated: $p = P/Ad$ and the angular frequency $\omega = 2\pi f$ it is possible to obtain the Equation 1-7:

$$p = 2\pi f\varepsilon_0\varepsilon'' E_i^2 \qquad \textbf{Equation 1-7}$$

Some very important features of dielectric heating equation are particularly evident from Equation 1-7:

a) The power density dissipated in the workload is proportional to frequency where the other parameters are constant. This means that the volume of workload in the oven may be reduced as the frequency rises, resulting in more compact oven.
b) The power density is proportional to the loss factor.
c) For a constant power dissipation density the electric field stress E_i reduces with $\sqrt{f}$. Then if ε'' remains constant with frequency the risk of voltage breakdown reduces as the chosen operating frequency rises.
d) In practice the value of ε'' varies with the frequency, generally, but not always ε'' rises with the frequency (see Paragraph 1.4.4). It also varied with temperature, moisture content, physical state (solid or liquid) and composition as detailed in Paragraphs 1.4.2-1.4.3. All these may change during processing, so it is important to consider ε'' and also ε' as variables during the process.

1.3.3 Penetration depth

Because of absorption, the electromagnetic fields decrease as the wave passes through the material. In the absence of reflected waves in the material, the field intensity and its associated power flux density fall exponentially with distance from the surface. Because the power absorbed in an elemental volume of material is proportional to the power flux density flowing through it, the power dissipation also falls exponentially from the surface. The rate of decay of the power dissipation is a function of both the relative permittivity ε' and the loss factor ε''. The "Penetration depth" D_p is defined as the depth into the material at which the power flux has fallen to $1/e$ (=0.368) of its surface value and it is given by the Equation 1-8 (Metaxas and Meredith, 1983):

$$D_p = \frac{\lambda_0}{2\pi\sqrt{2\varepsilon'}} \frac{1}{\sqrt{\left[1+\left(\frac{\varepsilon''}{\varepsilon'}\right)^2\right]^{0.5}-1}}$$

Equation 1-8

When $\varepsilon'' \leq \varepsilon'$ Equation 1-9 simplifies in:

$$D_p = \frac{\lambda_0 \sqrt{\varepsilon'}}{2\pi\varepsilon''}$$ **Equation 1-9**

The penetration depth is a very important parameter for a workload because it gives an immediate first-order indication of the heat distribution within it. In a semi-infinitive slab of ideal material, for example, having constant values of ε' and ε'' with temperature, and with a plane wave at normal incidence, the temperature rise T_z at depth z is given by the Equation 1-10:

$$T_z = T_0\, exp\left(-\frac{z}{D_p}\right)$$ **Equation 1-10**

The depth of penetration also determines the uniformity of heating, curing, etc., throughout the material. High frequencies and large values of the dielectric properties will result in surface heating, while low frequencies and small values of dielectric properties will produce a more volumetric heating.

1.4 Dielectric properties of materials

The dielectric properties of materials vary widely, not only with composition, but also with density, temperature and frequency. There is an extensive literature on methods of measurement (Von Hippel, 1954, Gallone et al., 1996) and on the results obtained (Bengtsson and Risman, 1971). Knowledge of dielectric data is essential in the design of heating systems because it enables estimates to be made of the power density and associated electric-field stress and equally important to the microwave-penetration depth in the material. It will be appreciated form the following that there can be substantial variation from the norm for a given material, and so it is desirable that measurements be made of each specific material to be processed.

Materials may be classified into three groups, i.e. conductors, insulators and absorbers, as illustrated in Fig. 1-4. Materials with a high conductance and low capacitance (such as metals) have high dielectric loss factors. As the dielectric loss factor gets very large, the penetration depth approaches zero. Materials with this dielectric behaviour are considered reflectors. Materials with low dielectric loss factors (insulators) have a very large penetration depth. As a result, very little of the energy is absorbed in the material, and the material is transparent to microwave energy. Because of this behaviour microwaves transfer energy most effectively to materials that have dielectric loss factors in the middle of the conductivity range. In contrast, conventional heating transfers heat most efficiently to materials with high conductivity.

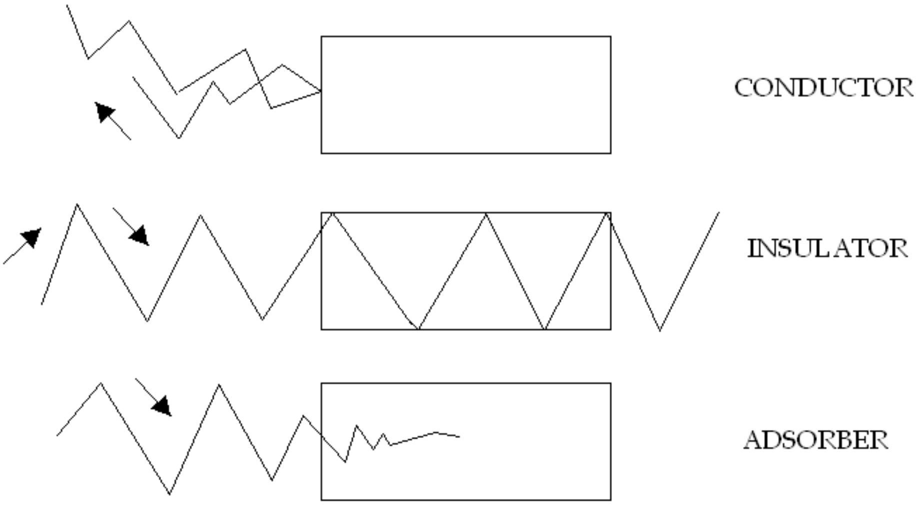


Fig. 1-4 Microwave absorption characteristics for conductor, insulator and absorber.

1.4.1 Measurement of dielectric properties

Because the dielectric properties govern the ability of materials to heat in microwave fields, the measurement of these properties as a function of temperature, frequency, or other relevant parameters is particularly important. Many authors have reviewed different techniques for dielectric property measurements at microwave frequencies. The two most common techniques are the resonant cavity, or cavity perturbation method, and the transmission and reflection method (Metaxas and Meredith, 1983). In the resonant cavity method, the ratio of energy stored in the cavity to the energy lost (often referred to as the quality or Q-factor) is measured in an empty cavity and a cavity with a small sample in it. Because microwave systems are often designed by replacing system components with equivalent electrical circuits (Thostenson and Chou, 1999), the Q-factor is based on the parallel RLC (resistance–inductance– capacitance) resonant circuit theory (Fig. 1-5). The Q-factor for a parallel RLC circuit is given by Equation 1-11:

$$Q = R\sqrt{\frac{L}{C}}$$ **Equation 1-11**

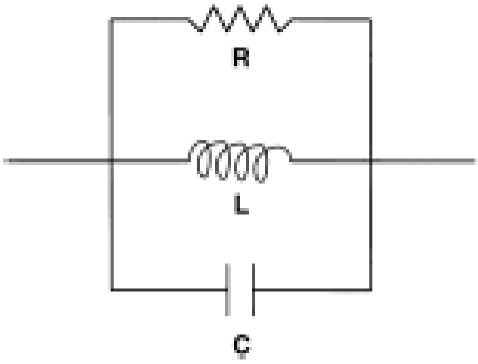


Fig. 1-5 Parallel RLC circuit.

The addition of the sample in the cavity perturbs the electromagnetic field, and it changes R, L, C, and, hence, the impedance of the equivalent circuit. The changes in the Q-factor and resonant frequency of the cavity can be related to the dielectric constant the loss tangent of the sample. In the transmission and reflection method, a sample is placed in a waveguide and the phase and amplitude of the transmitted and reflected waves are examined. The differences in these waves give information on the dielectric properties. For high-temperature dielectric property measurements on ceramics, factors, such as accurate temperature measurement, complicate the measurement of dielectric properties using these techniques (Tinga , 1992).

In fact, while standard methods have been established for dielectric property determination at low temperatures, characterization at elevated temperatures (up to 1500°C) has now been developed. A number of research programs in the world have focused on developing high-temperature measurement devices. These efforts are especially important for characterizing materials that display increased losses above a critical temperature, leading to a rapid rise in heating. This phenomenon, commonly known as thermal runaway, can result in poor product quality and cracking. In some applications, however (such as firing, sintering, melting, annealing, and fibre drawing), rapid heating at high temperatures can be beneficial. In such cases, developing temperature control techniques would be invaluable.

1.4.2 The variation of the permittivity with moisture content

Since many applications involve the removal of moisture (*M*) from the workload, the variation of ε, in particular ε'' with moisture content, plays an important role in the design of microwave heating/drying devices. For this a major effort was devoted to establishing the variation of ε' and ε'' with moisture for many industrial materials encountered in manufacturing industry, such as paper, board, foodstuffs, leather, wood and textiles.

Liquid water is strongly polar in its structure, causing readily to adsorb microwave energy and convert it into heat. When in contact with another material, liquid water is referred to as adsorbed water. However, the dielectric properties of adsorbed water show marked differences to that of liquid water. The principle relaxation for liquid water (i.e. maximum loss factor) occurs at about 18 GHz (Metaxas and Meredith, 1983) with other minor relaxations taking place further up the infra-red frequency band. On the other hand the relaxation peaks of adsorbed water occurs at frequencies will below the 18 GHz level. It appears therefore that the nature of the water adsorbed in a material has a marked effect upon its dielectric properties and consequently upon the interaction of the high frequency field with that material.

Adsorbed water in wet materials can exist in two principle states: free water, which resides in capillaries, cavities etc. and bound water, which is chemically combined to other molecules or physically adsorbed to the surface of the dry material. Fig. 1-6 shows qualitatively the variation of the loss factor with moisture content, M of a typical wet solid. The two states of water within the original material can be related to different regions of the ε'' vs. M curve, these regions being characterised by the slope ($d\varepsilon'/dM$).

The smaller slope at low moisture content is due to bound water (region I) whereas the higher slope at increased moistures is largely due to the presence of free water (region II). The water molecules 'bound' in the first uni-molecular layer at the surface of the material are less rotationally free than the water residing in capillaries and cavities. Thus the latter gives rise to much higher dielectric loss. The change of slope occurs at the critical moisture content, M_c. However some materials exhibit a gradual change of slope making the positive identification of the two regions fairly difficult. The critical moisture content for highly hygroscopic materials occurs in the region between 10%-40% (dry basis) whereas for non-hygroscopic materials (e.g. sand) it is about 1%.

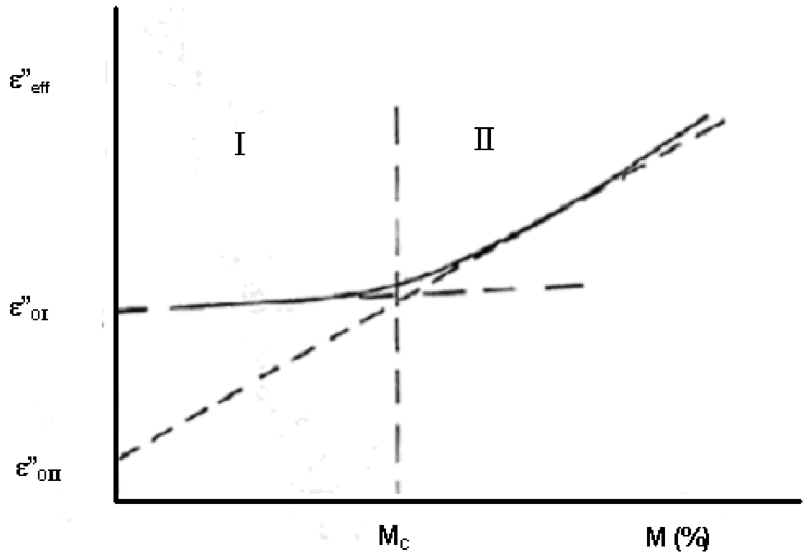


Fig. 1-6 The effective loss factor as a function of M (moisture content) (Adapted from Metaxas and Meredith, 1983).

1.4.3 The variation of ε″ with temperature

A number of investigations have been made in order to explain the temperature dependence of the loss factor in various materials. From Tinga's measurements, shown in Fig. 1-7 at low moistures the loss factor increases with temperature since the physical binding reduces and the dipoles are freer to reorientate (Metaxas and Meredith, 1983). For hydrations above 25% the loss factor decreases with increasing temperature.

The influence of the temperature and frequency on ε of many foodstuffs has been thoroughly investigated by Bengtsson and Risman (1971) in the temperature range [-20°C – 60°C] and by To et al. (1974) who have concentrated on temperatures above thawing to 65°C.

However, dielectric property data are important because they give values of the loss factor, ε'', which controls, along with other parameters such as the dielectric field and the frequency, the power that can be dissipated in a given material volume. Alternatively, for a given power dissipation, ε'' controls the rate of rise of temperature.

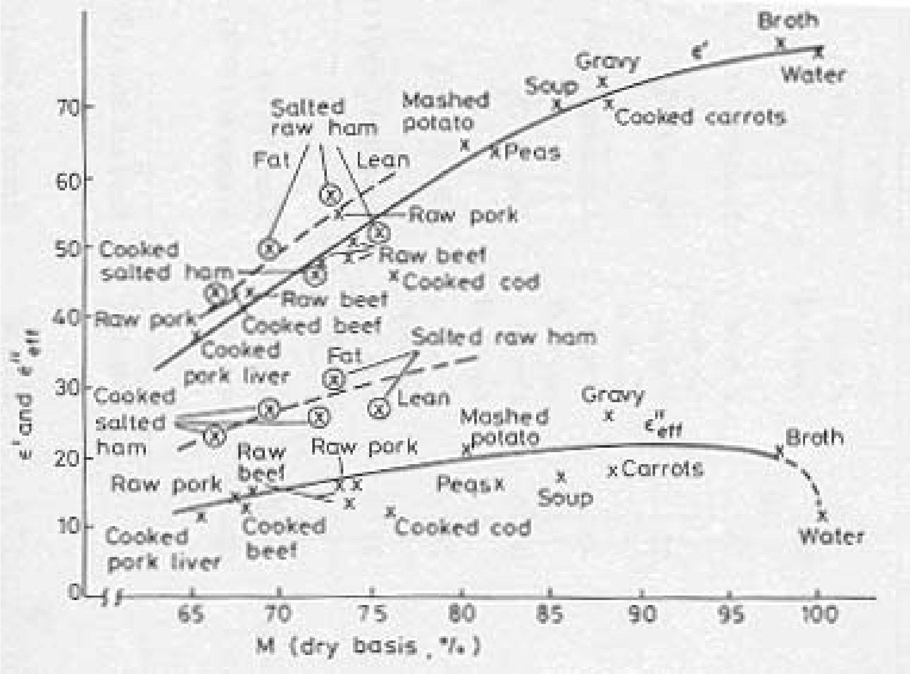


Fig. 1-7 Relationship between water content and dielectric data at 20°C and 2-8 GHz (points for salted material are circled) (Adapted from Metaxas and Meredith, 1983).

The larger the loss factor the easier the material absorbs the incident microwave energy. As a general practical rule, loss factors less the 10^{-2} require very high electric field strengths in order to ensure a reasonable rate of rise of temperature in the material; such low loss factor would almost certainly require fundamental mode resonant applicators as distinct from travelling wave or multimode applicators. On the other hand, loss factors of greater than 5 might present depth of penetration problems, in that because the material is highly absorptive to microwave radiation, most of the incident energy is absorbed within the first few millimetres, leaving the internal parts little affected. This causes non uniformities of heating which for in-depth heating are totally unacceptable. Therefore loss factors between the limits $10^{-2}<\varepsilon''<5$ would by presented by materials which, in general are good candidates for microwave heating applications.

1.4.4 The variation of $\varepsilon''$with frequency

Industrial use of microwaves calls for operation at discrete frequencies within given bands carefully selected so as not to interfere with other frequencies in use in telecommunications, defence and maritime applications. Therefore the measurement of the relaxation response of ε'in a particular industrial material, shown qualitatively in Fig. 1-8, is not in itself of particular significance other than to point out the relative values of the loss factor ε'', at the various allocated frequencies.

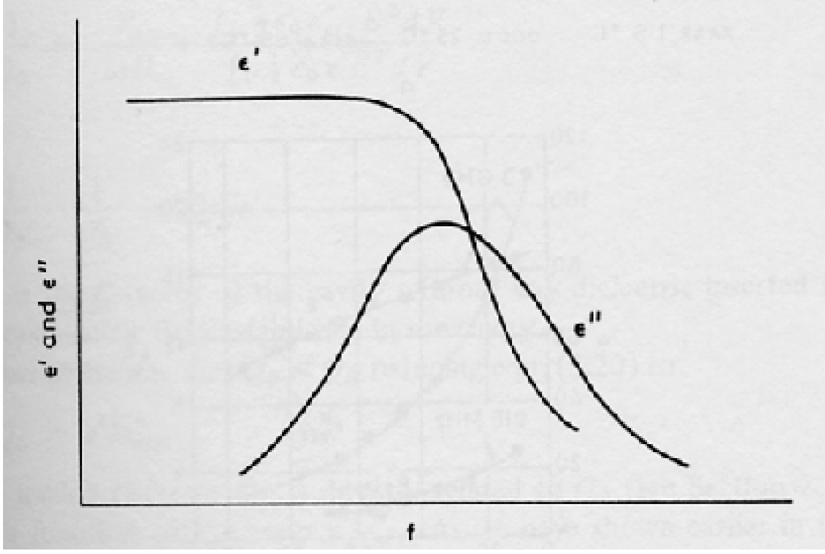


Fig. 1-8 Dielectric relaxation of a typical polar dielectric (Adapted from Metaxas and Meredith, 1983).

1.4.5 Thermal Runaway

Of great importance to microwave heating applicators is the "runaway" or the uncontrolled rise in temperature in a material brought about as a result of a positive slope, $+d\varepsilon''/dT$ of the ε'' vs. temperature response. A typical example is the case of Nylon 6.6: after an initial absorption of the microwave energy the temperature rise causes the ε'' to increase which, in turn, results in a further temperature increase, and so on. Damage of the material is possible unless steps are taken to avoid this cumulative effect by either interrupting the microwave energy or removing the material from the heated zone.

The techniques in the design of microwave heating systems are aimed to avoiding the risk of thermal runaway caused by the positive value of $d\varepsilon''/dT$. Generally the rate of rise of temperature of an elemental volume, is proportional to its heat input, $\varepsilon'' E^2 f$, but heat is conducted away from that element at a rate proportional to , where α_t is the thermal diffusivity. An equilibrium temperature is established when $\varepsilon'' E^2 f$ is equal to the rate of heat loss and is a low value consistent with achieving an adequately fast processing time.

1.5 Hardware Components

Microwave appliances have three major components: a microwave generator, a wave-guide and an applicator. Microwaves can be generated by several methods. Microwave signal transmission can be performed using transmission lines or wave-guides, which are inherently shielded and more widely used for low-loss transmission. There are many types of microwave applicators, each usually designed for a specific use. Examples include a resonant cavity, with dimensions chosen for a specific application, and an antenna for direct irradiation.

1.5.1 Microwave sources

Generation of electromagnetic radiation results from the acceleration of charge. To achieve the high power and frequencies required for microwave heating, most microwave sources are vacuum tubes. Some vacuum tubes that have been used for microwave heating include magnetrons, travelling wave tubes (TWTs), and klystrons. Magnetron tubes, which are used in home microwave ovens, are efficient and reliable (Kitagawa and Kanuma, 1986). Because magnetrons are mass produced, they are the lowest cost source available.

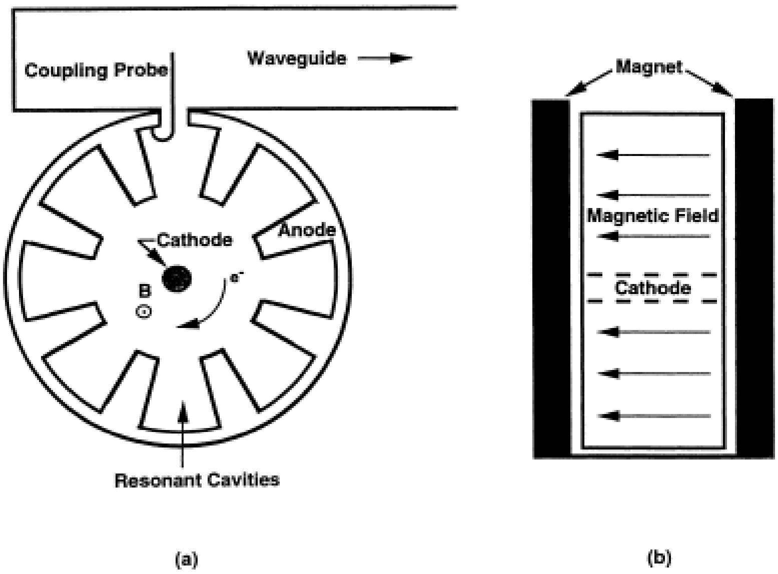


Fig. 1-9 Schematic diagram of the magnetron microwave tube (a) top view, (b) side view (Adapted from Roussy and Pierce, 1995, in Thostenson and Chou, 1999).

Magnetron tubes use resonant structures to generate the electromagnetic field, and, therefore, are only capable of generating a fixed frequency electromagnetic field. In variable frequency microwave, TWTs are used to generate the electromagnetic field. The design of the TWT allows amplification of a broad band of microwave frequencies in the same tube.

1.5.1.1 Magnetrons

In vacuum tubes, the anode is at a high potential compared to the cathode. The potential difference produces a strong electric field, and the cathode is heated to remove the loosely bound valence electrons. Once the electrons are removed from the cathode, they are accelerated toward the anode by the electric field. In a magnetron (Fig. 1-9), an external magnet is used to create a magnetic field orthogonal to the electric field, and the applied magnetic field creates a circumferential force on the electron as it is accelerated to the anode. The force causes the electron to travel in a spiral direction, and this creates a swirling cloud of electrons. As electrons pass the resonant cavities, the cavities set up oscillations in the electron cloud, and the frequency of the oscillations depends on the size of the cavities. Electromagnetic energy is coupled from one of the resonant cavities to the transmission lines through a coaxial line or waveguide launcher.

1.5.1.2 Traveling wave tubes

For variable frequency microwaves, high power travelling wave tubes are used as the microwave source. Unlike magnetrons, where the tube is used both to create the frequency of the oscillations and to amplify the signal, the TWT serves only as an amplifier. A voltage-controlled oscillator generates the microwave signal. The input voltage controls the frequency of the oscillator, and the signal is then sent to the TWT for amplification (Lauf et al., 1993). Because the oscillator determines the microwave frequency, these types of sources are able to rapidly switch the output frequency. The TWT (Fig. 1-10) consists of two main components: the electron gun and the helical transmission line. Because there are no resonant structures, it is possible for the TWT to amplify a large variation of frequencies (bandwidth) within the same tube. The heated cathode emits a stream of electrons that is accelerated toward the anode, and the electron stream is focused by an external magnetic field. The purpose of the helix is to slow the phase velocity of the microwave (the velocity in the axial direction of the helix) to a velocity approximately equal to the velocity of the electron beam.

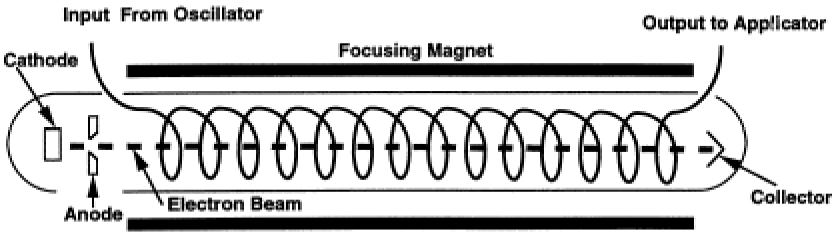


Fig. 1-10 Schematic diagram of the travelling wave tube ovens (Adapted from Collin, 1966, in Thostenson and Chou, 1999).

The wave propagates along the helix wire, and the pitch of the helix determines the phase velocity of the wave (Thostenson and Chou, 1999). When the microwave signal propagates along the helix, the axial component of the electromagnetic field interacts with the electron beam (Fig. 1-11).

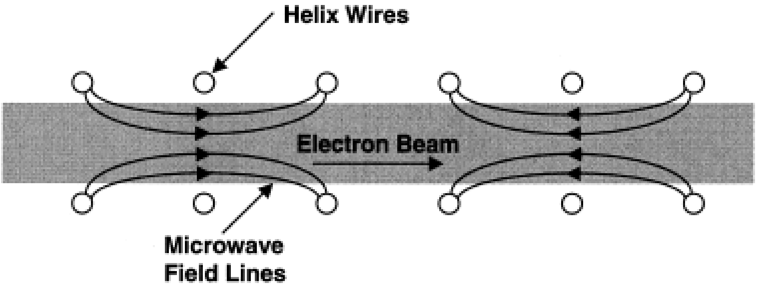


Fig. 1-11 Interaction of the microwave field with the electron beam in a traveling wave tube (Adapted from Veley, 1987, in Thostenson and Chou, 1999).

This results in acceleration and deceleration of electrons within the beam. For amplification of the signal to occur, the velocity of the electron beam should be just faster than the phase velocity of the helix. In this case, more electrons are being decelerated than accelerated, and the signal is amplified because energy is being transferred from the electron beam to the microwave field.

1.5.2 Transmission lines

The transmission lines couple the energy of the microwave source to the applicator. In low power systems, the transmission lines are often coaxial cables, which are similar to cables that are used on televisions. At high frequencies and output power, the losses that occur in coaxial cables are significant, and waveguides are often the transmission line of choice in microwave heating systems. Waveguides are hollow tubes in which the electromagnetic waves propagate.

The most commonly used cross-sections are rectangular. Two modes of microwave propagation are possible in waveguides: transverse electric (TE) and transverse magnetic (TM). For the TE mode, the electric intensity in the direction of propagation is zero. For the TM mode the magnetic intensity in the propagation direction is zero.

Every mathematical solution of the electromagnetic wave in a rectangular waveguide can be decomposed into a linear combination of the TE and TM modes. The most common waveguide mode is the TE_{10} mode. The subscripts specify the mode of propagation, and the mode indicates the number of maxima and minima of each field in a waveguide. In addition to waveguides, there are several other transmission line components that are used for equipment protection, sensing purposes, and coupling microwaves with the material in the applicator.

1.5.2.1 Circulators

When materials that are not good absorbers of electromagnetic energy are heated, a significant amount of power is often reflected back to the microwave source. Excessive reflected power can damage magnetrons. The circulator protects the microwave equipment by acting as the microwave equivalent to a diode in an electrical circuit; microwaves are only allowed to pass through the circulator in one direction. In the three port circulator, one port is connected to the microwave source, another is connected to the applicator, and the third port is connected to a dummy load. The power that is reflected back to the magnetron is diverted, and the dummy load, often water, absorbs the reflected power.

1.5.2.2 Directional couplers

In microwave heating, the ability of materials to absorb electromagnetic energy depends on the dielectric properties. Thus, the magnitude of the forward and reflected power is of interest to the researcher. Power measurement is accomplished through the directional coupler. Directional couplers are designed so that a small amount of forward and reflected waves are separated and measured by power meters.

1.5.2.3 Tuners

Tuners are used to maximize the power absorbed by the load through impedance matching. Several tuners, such as irises, three stub tuners, and E-H plane tuners, are used so that differences between the impedance of the microwave source and the load can be adjusted.

1.5.3 Microwave applicators and processing systems

The design of the applicator is critical to microwave heating because the microwave energy is transferred to materials through the applicator. The temperature fields within the material undergoing microwave heating are inherently linked to the distribution of the electric fields within the applicator. Common microwave applicators include waveguides, traveling wave applicators, single mode cavities, and multi-mode cavities. For processing materials, resonant applicators, such as single mode and multi-mode applicators, are most common because of their high field strengths.

The type of applicator used in a microwave processing system often depends on the materials to be processed. Commercially available single mode, multi-mode, and variable frequency multi-mode processing systems are all used for microwave processing research, and each of these systems has advantages and disadvantages.

1.5.3.1 Single mode

In the microwave applicator, or cavity, theoretical analysis can be performed to describe the response of microwaves. Given the geometry of the applicator, it is often possible to solve the Maxwell equations analytically or numerically with the appropriate boundary conditions. The design of single mode applicators is based on solution of the Maxwell equations (Equation 1-12) to support one resonant mode (Metaxas and Meredith, 1983).

Equation 1-12

Consequently, the size of single mode applicators is of the order of approximately one wavelength, and to maintain the resonant mode, these cavities require a microwave source that has little variation in the frequency output. Because the electromagnetic field can be determined using analytical or numerical techniques, the areas of high and low electromagnetic field are known, and single mode applicators have no uniform, but predictable, electromagnetic field distributions.

In general, single mode cavities have one ''hot spot'' where the microwave field strength is high.

This ability to design an applicator where the locations of high and low field strengths are known can offer some distinct advantages. Through proper design, single mode applicators can be used to focus the microwave field at a given location.

This technique has been exploited for joining of ceramics (Palaith and Silberglitt, 1989). In this application, it is desired to concentrate the microwave energy at the joint interface without heating the bulk of the ceramic. By placing the ceramic joint in the area of high electric field, localized heating of the ceramic joint will be achieved. Clearly, the size limitations of a single mode applicator limit this technique to relatively small joints that will fit within the area of high field strength in a single mode applicator. More recently, Tinga et al. (Tinga et al., 1995) have developed an open-ended single mode applicator for continuous joining of ceramic sheets that can overcome some of these practical limitations.

In addition, a knowledge of the electromagnetic field distribution can allow materials to be placed in the area of highest field strength for optimum coupling. Therefore, these cavities have been used for laboratory-scale studies of interactions between microwaves and materials. For example, single mode cavities are often used for studying the effect of microwaves on the curing kinetics of thermosetting resins (Wei and Hawley, 1993, Lewis et al., 1992). For cure kinetic studies, the single mode cavity offers a very controlled environment where the small samples can be placed for optimum coupling. In larger microwave cavities, small samples used for the kinetic studies do not represent a significant coupling load, and the process is more difficult to control for small coupling loads. An additional advantage of single mode cavities is the ability to monitor the dielectric properties during processing. Although the heating patterns of single mode cavities are non-uniform, in some single mode applicators it is possible to switch resonant modes. Sometimes the different resonant modes that are possible within the applicator have complementary heating patterns. Mode switching in single mode applicators is accomplished through altering the geometry of the applicator or by adjusting the frequency of the microwave source. By adjusting the height, for example, the applicator can be ‘‘tuned’’ to a different resonant mode. From a knowledge of these heating patterns in single mode cavities, the resonant mode can be changed during use to achieve a uniform in-plane temperature within the material (Ramakrishna et al., 1992).

Other applications for single mode applicators include use as preheaters for the pultrusion process and applicators designed for processing of ceramic filaments. Although single mode applicators have potential for some applications and laboratory-scale investigations of microwave/ materials interactions, single mode cavities are difficult to scale-up to many industrial applications due to the geometric limitations and non-uniformity of the fields. Therefore, single mode cavities are generally designed for specific purposes. Larger microwave applicators with more uniform fields are required to process large, complicated shaped components

1.5.3.2 *Multi-mode applicators*

Applicators that are capable of sustaining a number of high order modes at the same time are known as multimode cavities. This type of applicator is used in home microwave ovens. Unlike the design of single mode applicators, which are designed based on solutions of the electromagnetic field equations for a given applicator geometry, the design of multi-mode applicators are often based on trial and error, experience, and intuition (Metaxas and Meredith, 1993). As the size of the microwave cavity increases, the number of possible resonant modes also increase. Consequently, multi-mode applicators are usually much larger than one wavelength.

For a rectangular cavity, the mode equation for the resonant frequencies is given by Equation 1-12 (Collin, 1966):

Equation 1-13

where:

- f_{nml} is the TE_{nml} or TM_{nml} mode's resonant frequency
- c is the speed of light;
- n, m, l are the number of half-sinusoid variations in the standing wave pattern along the x, y and z axes
- a, b, and d are the dimensions of the cavity in the x, y and z directions.

The presence of different modes results in multiple hot spots within the microwave cavity. Like single mode cavities, local fluctuations in the electromagnetic field result in localized overheating. To reduce the effect of hot spots, several techniques are used to improve the field uniformity. The uniformity of the microwave field can be improved by increasing the size of the cavity. Because the number of modes within a multi-mode applicator increases rapidly as the dimensions of the cavity increase (Equation 1-13), the heating patterns associated with the different resonance modes begin to overlap. The rule of thumb to achieve uniformity within an applicator is to have the longest dimension be 100 times greater than the wavelength of the operating frequency (Kimrey and Janney, 1988). Unfortunately, at the common microwave frequency of 2.45 GHz, the largest dimension would exceed 12 m. It is possible to achieve a greater field uniformity by operating at a higher frequency. At higher frequencies, the wavelength is shorter, and the applicator required to achieve uniformity can be reduced to a practical size. Although higher frequency processing may seem to be the solution to creating greater uniformity, 2.45 GHz is able to penetrate deeper to create volumetric heating. Consequently, many attempts have been made to achieve more uniform heating within smaller multi-mode ovens at 2.45 GHz (Risman et al., 1987). A familiar example can be found in the home microwave oven. These ovens are often equipped with turntables that rotate during operation. The purpose of the turntable is to reduce the effect of multiple hot spots by passing the food through areas of high and low power and, therefore, achieve time-averaged uniformity. Another technique for improving the field uniformity is through mode stirring. Mode stirrers are reflectors, which resemble fans, that rotate within the cavity near the waveguide input. The mode stirrers ''mix up'' the modes by reflecting waves off the irregularly shaped blades and continuously redistribute the electromagnetic field. Like turntables, mode stirring creates time-averaged uniformity. In addition, adding multiple microwave inputs within a multi-mode cavity can further enhance the uniformity (Tran, 1991). Most techniques for creating uniformity depend on modifying the electromagnetic field within the microwave cavity.

Another method developed to achieve more uniform heating is hybrid heating. Hybrid heating can be achieved through combining microwave heating with conventional heat transfer through radiation, convection, or conduction. Variations of this method have been used successfully by researchers at the Oak Ridge National Laboratory (Janney et al., 1992) and the University of Florida (Arindam et al., 1990) to heat ceramics in a multi-mode furnace uniformly. Multi-mode applicators are typically more versatile than single mode applicators for batch operations and processing of large, complicated shaped objects. Thus, multi-mode systems are by far the most common processing systems used in industrial applications.

Recently, variable frequency multi-mode processing systems have been developed for materials research. The variable frequency microwave furnace (VFMF), developed by researchers at the Oak Ridge National Laboratory and Lambda Technologies, Inc. (McMillan et al., 1997; R.J. Lauf et al., 1993), has been able to overcome the problems of power non-uniformity within multi-mode cavities. The system makes use of the TWT source to sweep the frequency of the microwave field. The cavity used in this type of system is a multi-mode cavity. The result is time-averaged power uniformity within the microwave cavity. The ability to excite many different resonant modes by sweeping the frequency allows for uniform heating in a small cavity. It has been demonstrated that large composite laminates can be cured and post-cured (Demuse and Johnson, 1994) and large volumes of resin can be uniformly cured in a variable frequency microwave (Surrett et al., 1994). Similar to the fixed frequency multi-mode processing systems, uniformity will be further enhanced in a large applicator due to the greater number of resonant modes possible.

Several parameters determine heating patterns and rates, including wave-guide and applicator configuration and specimen properties, size, shape, and position. Obviously, the ability to predict electromagnetic field and temperature distribution patterns in the processed sample and cavity is vital for establishing processing guidelines and developing optimal processing procedures. Consequently, major efforts are underway for developing process simulation procedures using numerical modelling to compute electromagnetic and temperature distributions. These developments will allow improvements in both hardware design and fabrication, resulting in savings in process development and optimisation.

1.6 Temperature Measurements

One of the most difficult, yet important, parameters to measure in a microwave environment is temperature. Sample temperature is the most common process control parameter in microwave processing. Inaccuracies in temperature measurement or perturbation of the microwave field by temperature sensors can lead to erroneous indications of process temperature and misleading representation of process efficiency.

Fig. 1-12 identifies various temperature-measuring instruments and their applicable temperature ranges. Thermocouples and optical measurements techniques are most often used in microwave processing.

Temperature measurements in a microwave environment present several difficulties:

- Temperature measurements must be made directly within the sample and not in its vicinity. Microwave heats the sample itself (heat from within) and not the surroundings, and hence temperature probes must maintain good contact with the sample to achieve accurate temperature measurements.
- Thermal gradients developed during microwave heating make characterizing sample temperature using a single measurement difficult.
- Maintaining good contact with the sample might be difficult because of the changes in sample size during processing or due to motion of the sample.

- Conventional temperature-measurement procedures using thermocouples are not suitable for making these measurements in a microwave environment. The presence of a metallic temperature probe in a microwave environment can cause electromagnetic interference problems, causing distortion of the electrical field or affecting the electronics used for temperature measurements, as well as errors due to self heating, heat conduction, shielding and excessive localized heating, particularly at the tip of the probe.
- Optical measurements techniques such as pyrometers and optical fibre probes assume knowledge of emissivity.
- The "heating from within" property of microwave heating results in a surface temperature that is different from the core value.

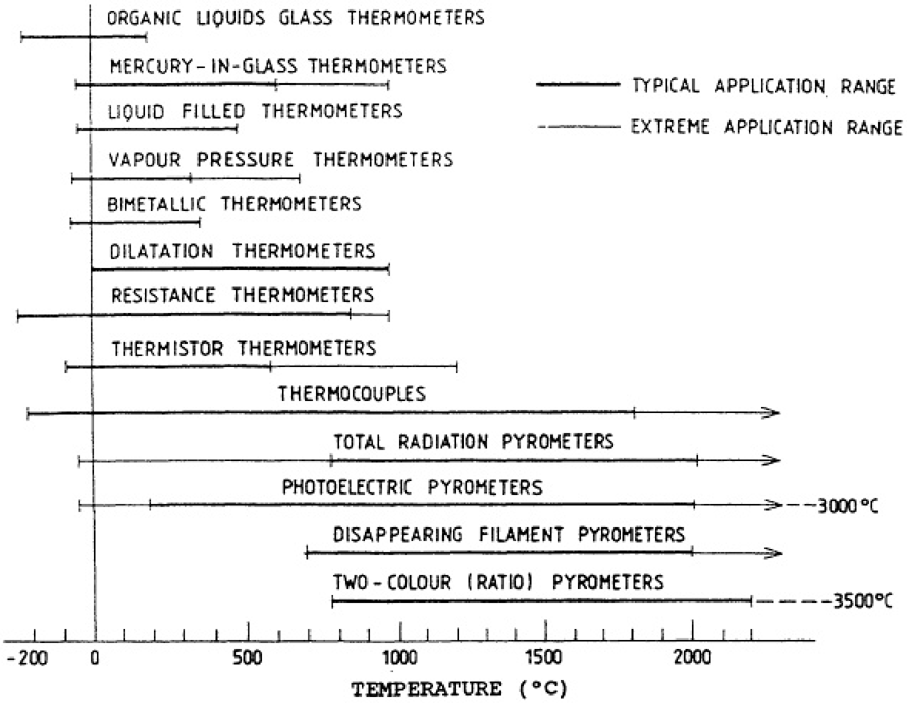


Fig. 1-12 Classification of temperature-measuring instruments (Adapted from Microwave Processing of Materials, 1994).

Depending on the optical properties and size of the sample, its emittance may make the optical radiation sensitive to the colder environment surrounding the sample.

1.7 Processing Materials

Microwaves present several characteristics that provide features not available with conventional surface-heating methods. These features as penetrating radiation, controllable energy distributions, rapid heating, selective heating, and self-limiting reactions, allow great flexibility and control for processing materials, and they can lead to improved quality and product properties, as well as reduced processing times, with resultant energy and labour savings. In addition, entirely new types of materials that cannot be produced by alternative methods can be created and synthesized. Microwave furnaces are quiet and safe and provide clean working conditions (which reduce contamination risks) with small floor-space requirements. Commercial opportunities for using microwaves to produce engineered materials are just beginning to be realized.

The range of dielectric responses of materials and their ability to absorb microwaves is perhaps one of the most widely used features of microwave processing. For example, water is a strong, broad-band absorber of microwaves and is, therefore, widely used in selective heating for processing food. Also, microwaves effectively remove water from all types of wood products, chemicals, and many other materials. The processing of rubbers, asphalt, ceramics, and polymer composites all depend on the selective heating of at least one of their constituents.

1.7.1 Polymer Processing

Microwaves were first used to process polymeric materials, such as certain rubbers, in the 1960s (Kirk–Othmer, 1995). In general, elastomers are poor thermal conductors and require complex and costly heating procedures. Microwave energy plays a considerable role in this field as the carrier of energy independent of the thermal properties of the material. Microwave processing has replaced traditional processes for certain polar elastomers-specifically, polychloroprene and nitrile butadiene rubber--and has the potential to provide new applications, such as the vulcanisation of thick and irregularly shaped sections.

For example, microwave energy has become the preferred method for foaming and preheating the rubber used in automobile seals. Through careful process development, the technology became a commercial success because of the high heating rates (substantially shortening process time) and enhanced product quality that results from uniform energy deposition. A significant aspect of this effort was tailoring the microwave absorption properties of carbon filler, a major component of filled rubbers. Carbon is generally a good microwave absorber, and the degree of absorption is dependent on the grade of carbon filler.

The use of microwave energy in polymerisation is in the development stage and plays only a minor role in industrial production, but this may change in the near future. Microwave technology has been applied to the processing of both thermosetting and thermoplastic polymers. Thermosetting materials (such as polyesters, polyurethanes, epoxies, phenolics, and amino resins) and thermoplastic materials (including polyolefines, vinyls, polyamides, and polycarbonates) have all been successfully processed with microwave energy. In the last 10 years, there has been much interest in using microwaves to process high-performance polymeric composites, such as carbon- and/or glass-fibre reinforced epoxies, which act as lightweight, high-strength structural components.

Flexibility in achieving temperature control is especially important in maintaining uniform temperature throughout a work piece, which is essential for curing polymer-matrix composites. For example, absorption properties of many polymeric materials depend on both temperature and frequency. Absorption characteristics throughout a processed sample may change as it is heated, making uniform heating more difficult. Changing the cavity mode, however, either by moving the cavity wall or changing the frequency, will change the energy deposition pattern within the sample. Frequency control enhances the ability to achieve more uniform temperatures. The prospects for developing mode-tuning devices to improve process control may have a profound impact on the number and quality of microwave-based applications for polymers.

1.7.2 Ceramic Materials

Among the most important microwave applications, a significant place is occupied by ceramics processing. The possibility of ceramics processing by microwave heating was discussed over 40 years ago by Von Hippel (Von Hippel, 1966), and experimental studies started in the middle of the 1960s by Tinga and co-authors (Tinga and Edwards, 1968). Since then the results of many investigations in microwave ceramics sintering and joining have been reported. In the majority of the papers the authors claim acceleration of microwave-driven processes as compared with processes performed using conventional heating. The acceleration commonly manifests itself as a reduction in the densification time of ceramic powder compacts, which is often accompanied by a decrease in the temperature of sintering. References to the published works on microwave sintering and joining of various ceramics and specific procedures and techniques can be found in the reviews by Sutton (Clark and Sutton, 1996), Katz (Katz, 1992) and Clark (Clark, 1997).

Therefore, high-temperature processing can benefit from such peculiar features of microwaves as reduced energy consumption and process time, rapid and controllable heating, inverse temperature distribution, and selective heating.

Specifically, high rates of volumetric heating, not limited by thermal diffusion, prevent recrystallization grain growth and result in a finer and more uniform microstructure of ceramic materials. It is well known that a fine, homogeneous, and fault-free microstructure is a necessary prerequisite for enhanced material performance. Similarly, a decrease in the duration of the high-temperature stage of the ceramics joining process leads to reduced grain growth in the joint zone and, as a result, to the higher mechanical strength of the joint. The inverse temperature profile with a maximum in the core of the body results from the combination of volumetric heating and surface heat loss. It is especially advantageous in processes that involve the reaction of a porous solid matrix with a gas or liquid phase, such as the synthesis of reaction-bonded ceramics or infiltration. Selective absorption of microwave energy and inertialess heating make up an added bonus of microwave processing. Selective microwave absorption can be purposely used for the synthesis of composite materials with desired functional properties. The inertialess nature of microwave volumetric heating offers the opportunity of the *in situ* control over the microstructure of the material undergoing processing.

Most of the early work on the processing of ceramic materials dates to the early 1960s, leading to commercial applications in the foundry and investment casting industry. Work on high-temperature applications, such as melting and sintering, began in the late '60s. Considerable interest was directed toward ceramics (in drying, calcining, sintering, and so on) through the '70s, when applications such as accelerated slip casting and acid digestion were developed.

Since many ceramics are transparent to microwaves at ambient temperature, microwaves can be used to detect moisture, pores, and defects that have different absorption characteristics. Microwaves can thus be used for non-destructive evaluation of materials. In fact, microwave devices were developed to detect, measure, and dry moisture around the tiles on the space shuttle.

The use of microwaves for processing solid ceramics can be divided into low-temperature (less than 500 °C) and high-temperature (greater than 500 °C) applications. While several applications of low-temperature processing have been commercialised, applications for high-temperature processing are mostly in the research stage or have been investigated in pilot tests. Moisture control and removal using microwaves is a common low-temperature process, and it has proven especially efficient and economical for low water-content applications. Low-temperature microwave processing of organic binders has found commercial success in removing waxes from ceramic shell moulds in the investment casting industry. This process uses only one-tenth the energy of the conventional autoclave process, minimizes poly-chlorobiphenyl contamination, reduces the breakage of shells, and reduces labour costs and storage space. It is also an excellent example of selective heating, because the shell, not the wax, is heated first.

High-temperature applications include ceramic joining, fibre drawing, melting, clinkering, sintering, and combustion synthesis. Combustion synthesis, in which two or more solids or a solid and a gas combine in the presence of heat to form a new material, has produced powders, carbides, nitrides, and oxides quite effectively using microwave energy. The basic difference between conventional and microwave combustion is that ignition begins at the centre of the sample under microwave irradiation, while in the conventional process, ignition generally begins at one or more surfaces. High-quality and high-strength joints can be made using microwaves because of the selective and localized heating characteristics of microwaves. In addition, the joining process can be many times faster than conventional methods because of microwaves heating rapidly.

1.8 Treating Hazardous Wastes

Commercial applications for treating hazardous waste with microwaves are not as well developed as applications in the materials processing industry. The use of microwave energy, however, has found certain niche applications that have been commercialised (systems for sanitizing infectious medical waste, for example), and a number of research efforts are underway to assess the potential for a range of environmental remediation applications.

Substances with low microwave-absorption characteristics can be treated indirectly by placing microwave susceptors (or absorbers) in a waste stream. Since only the substrate needs to be heated, the process can be conducted at lower temperatures, avoiding the formation of toxic by-products that often follow high-temperature incineration processes. Using suitable substrates, such as silicon carbide, this technique is effective in destroying trichloroethylene in gas streams.

Microwaves can also be used for regeneration and recovery processes. Microwave-regenerated spent activated carbon was found to produce a better product--with an estimated 10 percent lower cost--than that of conventional methods. Solvents have been recovered from waste polyurethane foam using microwave heat. The use of microwave heating to recycle used rubber tires via pyrolysis and devulcanization was studied in the 1980s, but poor economics discouraged development on a larger scale.

Contact less heating and remote operation make microwave technology an attractive alternative for processing toxic waste and both low- and high-level nuclear wastes. Microwave vitrification processes have been under study for a number of years. Once vitrified, hazardous materials pose less of a threat to the environment.

1.9 Continuing Research and Development

Microwave processing offers many potential benefits to industry, including time and energy savings, floor space reductions, and better use of assets. Major areas of ongoing research in microwave processing, include microwave/material interactions, dielectric measurements, microwave equipment design, process scale-up and evaluation, and new materials development. The establishment of multidisciplinary teams to research, develop, and market innovative approaches to employing microwaves will determine the rate and extent of the growth of a technology that offers much more than a convenient method for heating our food.

Processes based on microwave heating find many industrial applications. The main benefits of exploiting microwave energy in thermally activated processes depend on the specificity of microwave energy absorption. In contrast to all other commonly used methods, microwaves allow volumetric heating of materials. Microwave energy transforms into heat inside the material, which results, as a rule, in significant energy savings and reduction in process time. This factor plays a decisive role in most applications that have gained industrial acceptance to date. The wide availability of microwave power sources of frequencies <2.45 GHz and good microwave absorption properties of many materials have led to the emergence of industrial facilities for various applications with hundreds of megawatts in total of installed microwave power.