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Microwave Processing of Ceramics and Metallic Materials: Developments at Penn State

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1. Purpose

Microwave energy has been in use for over 50 years in a variety of applications such as communications, food processing, rubber vulcanization, textile and wood products, and drying of ceramic powders, but it is only recently that it is emerging as an innovative technology to process various kinds of materials in an efficient, economic and effective manner with great commercial potential. Recent developments and innovations in microwave processing at the Penn State University in the last decade have attracted worldwide attention in academia and industry. Among the most prominent advances in the past few years are: sintering of tungsten carbide (WC) based composites, fabrication of transparent ceramics, sintering of powdered metals, effect of E and H fields on the processing of various kinds of materials, and design of continuous microwave systems enabling the commercialization of the technology. In this presentation some of these developments will be reviewed.

2. Microwave Processing

Microwave processing of materials, which includes heating and sintering, is fundamentally different from conventional processing which involves radiant/resistance heating followed by transfer of thermal energy via conduction to the inside of the body under process through thermal conductivity mechanism. It is rather a slow process and takes considerable time to achieve thermal equilibrium. In case of microwave heating, it is the absorption of the microwave energy followed by volumetric heating involving a conversion of electromagnetic energy into thermal energy. In this process there is no thermal conductivity mechanism involved, the heating is instantaneous and rapid, and is a function of the material under process. The heat is generated internally within the material instead of originating from external sources, and transmits towards outside. Hence, there is an inverse heating profile, inside-out unlike in a conventional heating outside-in.

Microwave heating has significant advantages over conventional heating in materials processing such as energy saving, very rapid heating rates, considerably reduced cycle time, and environmentally cleaner. It has been generally observed that microwave sintered products exhibit finer microstructures leading to considerable improvement in the mechanical properties, and hence better overall product performance.

3. Experimental

In this study various microwave systems including multimode and singlemode, 2.45 GHz frequency were used. The power of the system varied from 1 kW to 6 kW depending upon the temperature and mass of the workpiece. In the case of metallic materials, systems with capability of control atmosphere (H₂, H₂+N₂, Ar etc.) were used. The temperature was monitored either by using a special shielded thermocouple of optical/IR pyrometers.

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4. Results

Here, we present some of these developments and results of the important ceramics and metallic materials researched at our microwave-processing center:

**Microwave sintering of Important Ceramics:** Alumina, zirconia, electroceramics, hydroxyapatite, spinels etc. have been very successfully sintered in microwave to full density very rapidly. Alumina samples microwave sintered at 1400°C, with no hold time, were 98% dense. In conventional heating it requires at least 1600°C and 2 hours of soaking time to achieve the same degree of densification and with substantial grain growth. In microwave nearly full density was achieved at about 200°C less than the conventional temperature. The alumina grit developed by Carborundum Universal (Kerala, India) is a kind of agglomerated granule with an average particle size of 0.8-1.0 mm. Microwave sintering at 1500°C for 15 minutes provided density of 3.96 g/cc, which is near theoretical density. The abrasion index of this grit was 94 and Vicker's hardness 2317 kg/mm². In another study large objects of alumina with diameter of 1 to 10 cm and length up to 1-2 meters were also prepared using continuous microwave system. These parts were sintered at 1400°C very uniformly and homogeneously and about 98% density was obtained.

Fine grained zirconia ceramics were sintered in microwave at 1360°C for 2 minutes in a multimode, 2.45 GHz system. The sintered density was about 97.8% and average grain size was 0.25 μm. In another study of phase transition in Ca-stabilized zirconia using microwave heating, almost complete transformation from tetragonal to cubic phase was obtained at 1600°C in 10 minutes; conventionally it takes several hours (~10 hours) to achieve the same degree of transformation.

In microwave assisted process single phase perovskite PbZr₀.₅₂Ti₀.₄₈O₃:PZT was obtained at as low as 600°C. If one of the constituent precursors is a non-stoichiometric oxide then several fold enhancement in the reaction rates is observed in producing single phase PZT. The usage of non-stoichiometric precursors also leads to different reaction pathways for the formation of PZT. Further, PZT samples were microwave sintered at significantly lower temperatures than conventional processing resulting in finer grain size and minimal PbO loss.

Ba(Mg₁/₃Ta₂/₃)O₃ (BMT) with perovskite structure is a good dielectric material for microwave resonators because it possesses high quality factor (Q) and moderate dielectric constant. This remarkable material is, perhaps, the most refractory oxide (melting point > 3000°C), and therefore very high temperatures are required to sinter it in a conventional furnace. It takes several hours and over 1650°C to achieve reasonable densification. Therefore, to obtain BMT ceramics with high density, sintering aids such as Mn and Sn are used. But the sintering aids also influence the dielectric properties undesirably. We have synthesized and sintered BMT single phase material using reduced oxide precursors and microwave process without any sintering aids. Use of reduced Ta₂O₅-x remarkably enhanced the reaction kinetics and produced single-phase material at much lower temperature (1300°C/20 min) and higher densification than normally obtained by conventional processes. Microwave processed BMT samples exhibited density as high as 97% of theoretical when heated at 1600°C for 30 minutes. The average grain size in microwave sintered BMT was about 1 μm in contrast to 3 μm in conventional sintered material.
Transparent Ceramics

The conventional methods to fabricate fully dense and reasonably transparent ceramics involve high temperatures, lengthy sintering conditions, and various complex processing steps such as HIP and HIP, which make the processing of transparent ceramics uneconomical and time consuming. However, microwave method has been successfully used to fabricate transparent ceramics due to its ability to minimize the grain growth and produce a fully dense ceramic in a very short period of time with out utilizing high-pressure conditions. Several useful ceramics such as alumina, spinel, hydroxyapatite, AlN, AlON, MgO were fabricated into transparent and/or translucent form at room pressure and much lower temperatures and time conditions than normally required by conventional method. Figure 1 shows some of these samples fabricated in microwave.

![Microwave Sintered Hydroxyapatite Core](image)

**Figure 1:** Microwave processed transparent ceramics

WC-Co Composites

Hard metal composites, especially the tungsten carbide (WC-Co) based materials due to their unique combination of high hardness, toughness and strength, are universally used for metal and rock cutting and drilling operations. Conventional methods for sintering WC with Co as a binder phase involve high temperature (up to 1500°C) and lengthy sintering cycles (~24 hours) in order to achieve high degree of densification. Since microwave heating requires very little time to obtain nearly full density, the grain growth is relatively suppressed and finer microstructure is generally obtained. It is observed that microwave processed WC/Co bodies exhibit better mechanical properties than the conventional parts, fine and uniform microstructure with little grain growth, and nearly full density without adding any grain-growth inhibitors. Commercial products of WC-Co produced by microwave sintering process exhibit unprecedented improvement in abrasion resistance (15-30% better), erosion resistance (22% better), and corrosion resistance (20X better) without any noticeable loss in hardness or fracture toughness.

Microwave sintering and brazing of Metallic Materials

Earlier, it was well recognized by the researchers that microwave heating does not work in metals and is good only to oxide ceramics and semi-metals like carbides and nitrides. But
recently it has been observed that the microwave sintering can also be applied as efficiently and effectively to powdered metals as to many ceramics. Bulk metals are excellent reflectors in microwaves at room temperature and in general are not heated significantly. But in powdered and unsintered form virtually all metals and alloys would couple/heat in a microwave field very effectively to produce highly sintered bodies with improved mechanical properties. The microwave sintering of PM green bodies comprising various metals, steels and metal alloys produced highly sintered bodies in a very short period of time. Typically the total cycle time was about 90 minutes, sintering temperature ranges between 1100°C to 1300°C and soaking time 5 to 60 minutes. The mechanical properties such as the modulus of rupture (MOR) and hardness of microwave processed samples were much higher than the conventional samples. As an examples, copper steel (MPIF FC-0208 composition) was successfully sintered by microwave technique to obtain good sintered density, hardness, flexural strength, and near net dimensions, thus yielding equivalent or even sometimes superior mechanical properties than conventional sintering. In this material the Rockwell B hardness (HRB) as high as 82±2 was obtained for microwave processed samples sintered at 1260°C for 5 min. soaking in flowing forming gas atmosphere.

It is known that microwave selectively heats powder metals and reflects bulk metals at room temperature. This feature has been exploited to braze and join bulk metals using powdered metal/alloy braze materials. We have joined steels, W bulk metals and also brazed super alloys. An example is shown in Figure 2 in which regular steel and cast iron parts have been joined in microwave field in 2-3 minutes using a braze powder.

Figure 2: Microwave joining of steel and cast iron

Heating in separate $E$ and $H$ fields at 2.45 GHz

The results presented above were all produced using a multi-mode microwave cavity at 2.45 GHz frequency, in which the electric and magnetic fields cannot be separated out. Recently, we have designed and built single mode microwave cavity of TE103 mode in which electric and magnetic fields can be separated and materials can be exposed to reasonably pure $E$ and $H$ fields at 2.45 GHz. When processed various materials in $E$ and $H$ fields, dramatic results were obtained. This presentation will show some of these recent results and discuss various possible mechanisms to explain microwave-matter interaction.