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List of Acronyms

MAT	Microwave Assist Technology
e”	dielectric loss
e”	permittivity
Tanδ	tangent delta, loss tangent
BC	boundary conditions
CFD	computational fluid dynamics
EM	electromagnetic
QW-3D	QuickWave-3D
EM-TH	electromagnetic – thermal heat
FDTD	Finite-Difference Time-Domain
Tpd	ton per day
BTU	British thermal unit

Executive Summary

The purpose of this project was to demonstrate the feasibility and evaluate the potential energy impact for implementation of Microwave Assist Technology (MAT) for industrial processes, with a specific focus on limestone calcination. Limestone calcination is a highly energy intensive process critical to the cement, steel, and glass industries. One industrial lime partner and one university partner participated in this project. The scope of work was designed to build on modeling and materials characterization, followed by MAT calcination experimentation, which was used to conduct a benefit analysis.

The microwave materials interactions for limestone were studied through dielectric property measurement and process modeling. Three different grades of limestone were provided by the industrial partner. The dielectric results indicated that the limestone would heat via microwave energy; however heating was temperature and impurity dependent. The ideal temperature range for microwave absorption was > 900 °C and higher impurity levels heated better. This information provided a baseline for understanding how limestone interacts with microwave energy and developing the MAT processing parameters. This information was also used as data input for the modeling work, which was conducted by the university partner. The model started with a microwave only simulation, and built a new module for incorporating simultaneous radiant heating. Modeling of various heating scenarios was performed, and the model is now a tool which can be used to provide a low-risk assessment of different experimental set-ups, and ultimately MAT kiln designs.

Seventy MAT and conventional experiments were conducted to initially develop the optimum processing parameters and then to conduct comparison studies. Laboratory scale testing was conducted in a lab-scale MAT kiln, which made MAT and conventional comparisons possible. Calcined materials were analyzed for weight-loss and reactivity, and it was determined that in general, MAT increased the rate of decomposition for limestone into lime and reduced energy consumption by 25%. Processing times were decreased by 34 %.

A visit to the industrial partner's manufacturing site helped to shape the design for large scale implementation. It was determined that due to the size and rotating nature of the industrial calciners, applying microwave energy throughout the entire calcining process was not feasible. Instead, the approach of a microwave post heating zone in the cooling box was explored.

Using this new approach of applying microwave energy only at the end of the calcining process, several pilot-scale MAT and conventional runs were conducted in a 27 ft^3 MAT kiln. Results indicated that MAT decreased energy consumption by 36% compared to conventional heating.

Energy consumption was monitored throughout the project for both the laboratory scale and pilot sale testing. Ceralink has demonstrated that the concept for using MAT™ as a means to efficiently calcine limestone has merit, due to the fact that MAT™ was shown to enhance the rate of calcining compared to conventional processing. Assuming a conservative 25% energy savings, estimates for large scale MAT™ calcining benefits were calculated for the lime and cement industries. This estimate showed that for the lime industry 7.3 TBTU/year, with 270 MMlbs CO_2 and $29 MM could be saved. For cement, the annual savings is approximately 5x greater due to the higher production rates yielding savings of 39 TBTU/year, with 3 Blbs CO_2 and $155 MM.

This project has furthered the commercialization effort of MAT by demonstrating energy savings on a pilot scale. However, one of the main barriers to commercialization of MAT for the lime and cement industries is the sheer size of production. Through this project, it was realized

that a production size MAT rotary calciner was not feasible, and a different approach was adapted. The concept of a microwave post heat section located in the upper portion of the cooler was devised and well received by the industrial lime company and an industry expert.

Commercialization of this technology will require 1) continued pilot scale calcining demonstrations, 2) involvement of lime kiln companies, and 3) involvement of an industrial microwave equipment provider. Ceralink identified two builders of lime kilns whom could be approached to partner with for continued technology and equipment development. Eventually, Ceralink would plan to license and transfer the technology to the kiln builders once the technical aspects are finalized. The lime company could then implement the MAT post heating sections in new builds and pursue retrofit of existing lime kilns in order to improve efficiency. Retrofits would likely be attractive to lime manufacturers, as the purchase of a new lime kiln is on the order of a $30 million dollar investment, where as a MAT retrofit might be on the order of $1 million. Ceralink would receive a royalty from the kiln builder.

Current scale-up designs indicate that four 100kW microwave generators (915 MHz frequency) will be required to provide sufficient energy to a chute measuring 6ft in diameter and 4 ft. long to finish calcining the limestone before the limestone is cooled. This is scale for a production size rotary calciner with 600 ton/day output. Ceralink has identified a microwave equipment supplier who could supply the required microwave equipment and engineering work to assist with design and integration.

Introduction

Microwave heating of materials occurs through internal friction when dielectric loss mechanisms respond in a microwave field. There are five different dielectric mechanisms (electronic, atomic, orientation or dipolar, ionic polarization and space charge) that can be activated in real materials related to crystal structure, defects, bonding, surface, grain boundaries, etc, see Figure 1. It’s been theorized that there should be an optimum frequency for maximizing the friction for each mechanism, but optimum frequency changes with temperature because certain movements become easier as the material heats up. This produces less friction in some cases and more friction in other cases, such as where a defect was frozen and couldn't previously contribute. This behavior causes a peak in the dielectric loss. Additionally, in practical terms there are certain frequencies that are allowed for industrial use. These are called ISM (industrial, scientific and medical) frequencies for applications in those areas.

Two frequencies were used in this project, 2.45 GHz and 915 MHz. The dielectric property testing indicated that the limestone coupled better with the microwave energy above 900 °C for both frequencies. Additionally, above 640 °C the limestone reaches its intrinsic conductivity, where electronic conduction becomes the dominant dielectric loss mechanism responsible for heating.

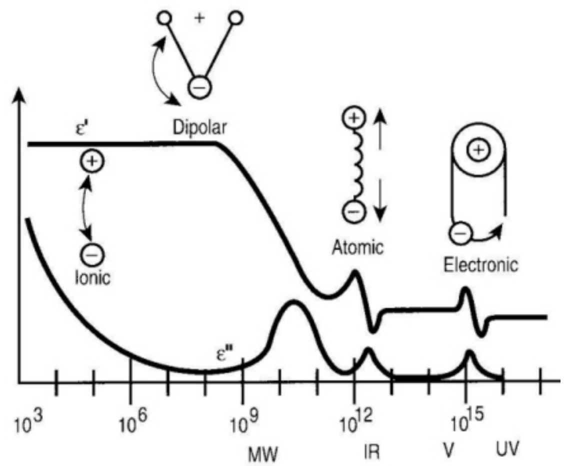


Figure 1. Dielectric mechanisms as a function of frequency[1].

In conventional heating, all heat must be transferred through the outer surface of the material to the interior. Microwave heating offers an important advantage of being able to place energy directly into the volume of the work piece. This requires meeting certain conditions where microwaves penetrate the material enough to cause volumetric heating. Very little heating occurs if microwaves are reflected or if they penetrate through the material too easily.

In this project, hybrid microwave processing was evaluated for energy reduction in the calcination of limestone to form lime and cement. Calcination of limestone ($CaCO_3$) to form lime (CaO) was selected as a focus in this study for three reasons. Firstly, calcination is an energy intensive process utilizing 90% of the energy consumed in the **cement and lime industries** (485 Tril BTU/yr). The cement industry produces about 100 million tons per year versus the lime industry at about 20 million tons per year and the energy consumption of both averages the same at about 6 mm BTU/ton. Therefore, the energy consumption for the lime industry is one fifth the consumption for the cement industry. Secondly, Microwave Assist Technology has been demonstrated to reduce energy consumption in similar processes by 50% or more. Thirdly, the development and commercialization of Microwave Assist Technology (MAT) in this industry will stimulate uptake in related processes, such as roasting of ore for easier comminution in the mining industry, calcining nano catalysts in the chemical industry, and calcining of proppants in the petroleum industry.

MAT is a method to simultaneously apply traditional radiant heat and microwave energy in the same kiln, leading to fast volumetric product heating. Microwave thermal activation is expected to target and directly heat the limestone, eliminating the reliance on thermal conduction as a means of energy transfer. In addition less energy is wasted in heating non-product, such as the atmosphere and kiln lining. This technology has the potential to increase the speed of lime production by decreasing both the reaction time and temperature, further reducing energy consumption. Full scale implementation of microwave hybrid calcining of limestone is expected to save 7.3 TBTU/year in the lime industry and 39 TBTU/year in the cement industry.

Background

The lime industry primarily uses rotary kilns (80%), mostly preheater kilns, and vertical kilns (20%). The kiln technology has developed over the years to reduce fuel consumption while meeting a variety of operational parameters. Kilns are designed based for the fuels available, limestone available, customer lime requirements, applications and environmental permitting limitations. Customer requirements include available calcium oxide, reactivity (how fast lime reacts with water), particle size (pebble or fines, <1/8 inch) and purity (impurities are usually customer specific such as sulfur).

Applications include steel manufacturing, water treatment, paper mills, environmental applications, flue gas desulfurization, etc. The *most efficient rotary kilns* are the larger preheater kilns with fuel consumptions of 4.5 MMBTU/ton lime while the most efficient verticals are the parallel flow regenerative (PFR) kilns at 3.02 MMBTU/ton lime. These fuel efficiencies have been accomplished by minimizing losses and recovering heat at both ends of the rotary kiln where the fuel is combusted. The vertical kiln does the same thing while combusting the fuel in the stone bed with heat recovery before and after combustion. Additionally, both technologies have improved fuel efficiency by continually improving refractory selection and design, by installing insulation with the refractory, by improving seals to minimize air in-leakage and improving operational control to maximize fuel efficiency.

Direct microwave heating overcomes the inherently slow thermal conduction in minerals such as limestone, metal ore, and ceramic powders. In conventional calcination processes, radiant heat is applied by burning coal or gas, or via electric heating elements. The product heats inwardly from the surface. This is a very slow process that requires long heat up and dwell times. Direct microwave heating enables reactions and diffusion to occur in shorter times and lower temperatures. Microwave power has not yet been applied to high temperature industrial processing due to many technical factors, which are solved by the combination of conventional radiant heat with microwaves (Microwave Assist Technology or MAT).

Ceralink is a materials engineering and technology commercialization company with eleven years of experience in developing microwave materials processing for drying, binder removal, calcining and sintering applications. Ceralink licensed Microwave Assist Technology in 2004 from C-Tech Innovations and holds an exclusive license in North America for MAT and is focused on commercializing this and other energy efficient technologies. Ceralink has extensive experience with microwave and materials interactions, microwave processing, and the design and building of MAT systems.

Ms. Morgana Fall, Operations Engineer (BS Alfred University), has 9 years of experience in ceramic engineering and microwave-materials interactions. Ms. Fall coordinates Ceralink's microwave equipment design work, and has designed several Microwave Assist Technology (MAT) kilns, from lab to production scale. Ms. Fall was responsible for the technology transfer for MAT when Ceralink licensed the technology in 2004. Ms. Fall has comprehensive knowledge of MAT process development and experimentation. Her ability to interpret microwave dielectric property data as it pertains to heating is a critical skill. Dr. Holly Shulman, founder and President of Ceralink, (PhD EPFL Switzerland, MS University of Pittsburgh, BS Alfred University) has 25 years of experience in ceramics and materials, with specializations in advanced ceramics, electroceramics, calcining of barium titanate, pigments and phosphors, as well as cutting edge microwave process development. Dr. Shulman has extensive experience developing and licensing ceramic materials and process technology. Mr. Shawn Allan, Senior Materials Engineer (MS Georgia Tech), has 9 years of experience in ceramic and metals processing. Mr. Allan is an expert in microwave dielectric properties, and microwave process development for calcination reactions. Patricia Strickland, CEO of Ceralink (BA Alfred University), has over 15 years of experience in finance and business management. She has a strong background in strategic partnering and has been successful in negotiating joint venture and licensing agreements for Ceralink. Ms. Strickland has been with Ceralink since 2001.

Dr. Vadim Yakovlev of WPI is an expert in electromagnetic modeling using QuickWave-3D software for modeling of microwave power applications. Dr. Yakovlev has several papers published on industrial microwave modeling, and has worked with microwave equipment manufacturers such as Ferrite Inc to help them model commercial systems for optimized energy efficiency, temperature uniformity, microwave energy input, and cavity design.

Mr. Johnney Bowers, a consultant of DCA Inc, assisted with the cost benefit analysis for this project, and understanding the needs of the lime industry. Mr. Bowers has over 17 years of experience working in the lime industry and has worked as the Vice President of Engineering one major lime company and Vice President of Operations for another US lime company. He also has experience in the kaolin and lithium industries where he worked with pyro processing applications involving a variety of rotary and vertical kilns

Results and Discussion

Materials Characterization

Dielectric property testing at microwave frequencies was conducted for various grades of limestone and for samples of kiln refractory materials. The purpose of the dielectric testing was to determine how materials would absorb microwave energy. Microwave heating occurs through dielectric loss mechanisms. The dielectric property which is most useful for predicting microwave heating behavior is the loss tangent (Tanδ). Tanδ is the dielectric loss (e") divided by the permittivity (e'). Dielectric loss is defined as a parameter of a dielectric material that quantifies its inherent dissipation of electromagnetic energy (or ability to store energy). Permittivity is directly related to electric susceptibility, which is a measure of how easily a dielectric polarizes in response to an electric field. Thus, permittivity relates to a material's ability to transmit (or "permit") an electric field. Interpretation of the Tanδ provides a guide to microwave heating behavior for developing a microwave process.

As a rule of thumb, when Tanδ is less than 0.01, the material is microwave transparent (none to weakly absorbing of microwaves). Weak microwave absorption causes the material to remain cool in a microwave field. A value of Tanδ which is equal or greater than 0.01 indicates the material will absorb microwave energy and heat. The higher the value of Tanδ is, the greater the microwave absorption will be, the faster the material will heat up. Microwave absorption is also referred to as "suscepting or coupling".

Dielectric property testing as a function of temperature was conducted for three grades of limestone. It was observed that there were differences in how the grades reacted with microwave energy. A graph of the Tanδ as a function of temperature is shown in Figure 2. The grade referred to as MLC1 had the highest Tanδ, followed by MLC3 and then MLC2. The higher the Tanδ the more strongly the material should heat in a microwave field. This information was used for developing a MAT calcining process for the laboratory experiments and was also used as input for the modeling work.

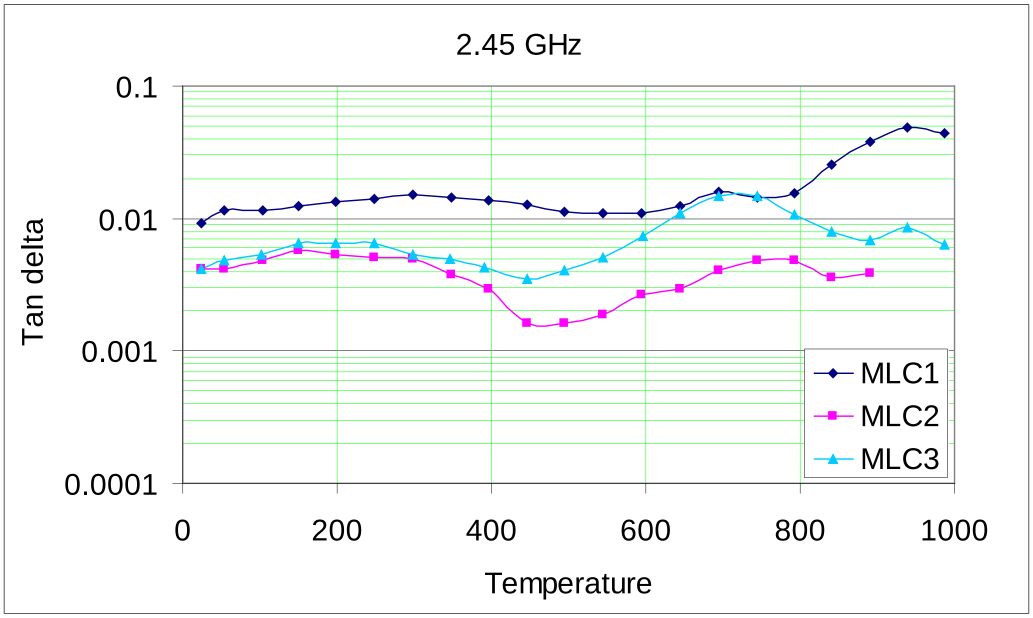


Figure 2. Tanδ as a function of temperature for 3 grades of limestone. The higher the Tanδ, the better the material should heat by microwave energy.

Samples of the lime kiln refractory used in different sections of the calcining kiln were procured from the industrial partner. Four different refractory materials were tested in order to evaluate their microwave interaction potential. The results are shown in Figure 3, which indicated that only a couple of the refractory materials are microwave compatible (low microwave absorption). The Reframag AF, a magnesia based refractory, had a much lower Tanδ value compared to the limestone (MLC1), making it compatible for use in the cooler section, where the microwave energy would be applied. Additionally, the ProGun 26Li has potential, as its Tanδ is slightly lower than the limestone. The other refractories tested had higher Tanδ, indicating that they would compete with the limestone for the microwave energy, and lower the overall heating efficiency of the system.

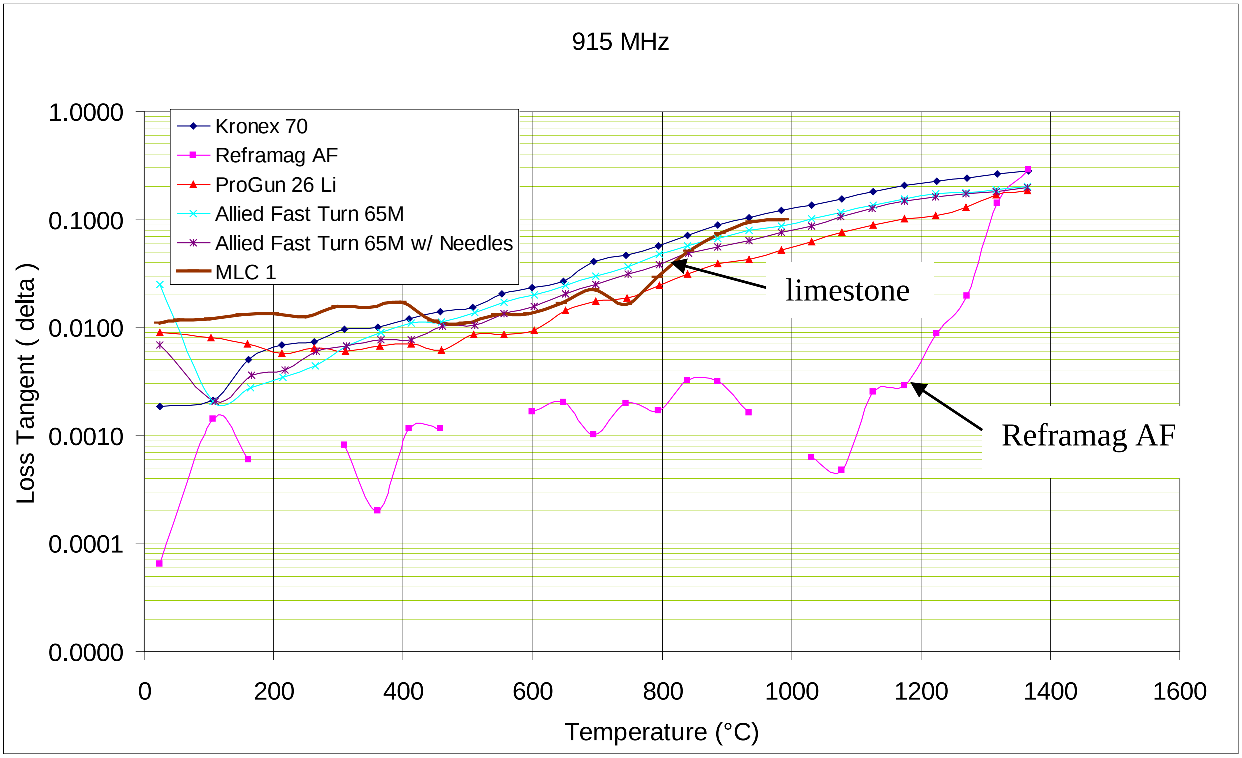


Figure 3. Graph of the Tanδ as a function of temperature for several grades of kiln refractories. Only the reframag AF and ProGun 26 Li had lower Tanδ compared to the limestone, making those compatible for microwave processing.

Modeling

The university partner has developed an advanced modeling technique capable of simulating thermal processes in hybrid systems of combined heating by heat radiation and microwaves. Simulation of heating scenarios in the systems of hybrid heating requires certain advancement beyond the current state-of-the-art in multiphysics modeling currently adopted in microwave power engineering [2-4]: such a technique should account for thermal radiation within the accurately and precisely reproduced geometry of the cavity. Appropriate approaches were employed in [5, 6], where Robin (convective) boundary conditions (BC) were applied at the sample walls with the effective heat exchange coefficient based on approximate formulas describing natural convection for several simplified geometries. Robin boundary condition is a general form of the insulating boundary condition for convection–diffusion equations. In [7], a custom electromagnetic-thermodynamic model accounting for radiant heat transfer has been proposed.

However, these techniques have limited capabilities in terms of both electromagnetic (EM) and computational fluid dynamics (CFD) analyses and thus cannot be easily extended to include the models routinely available in advanced EM and CFD modeling tools. This makes application of the earlier models to practical systems performing hybrid microwave processing not possible.

Basic Principles and Features of the Developed Technique

The approach to extending applicability of the modeling algorithms to problems involving both microwaves and heat radiation suggests coupling of an advanced EM solver with a full-fledged CFD package offering various models of radiation.

The developed computational tool is built using the 3D conformal FDTD simulator *QuickWave-3D* (*QW-3D*) ver. 7.5 [8] and the finite element solver *ANSYS Fluent* ver. 8 [9]. Functionality of these two simulators coupled on an elementary level has been earlier described in [10] and is thus regarded fully operational. In this project, the coupled simulator has been extended to account for the radiant BC and applied to hybrid radiative-microwave heating of limestone in a hybrid furnace (MAT™ kiln shown in Figure 4).

The developed technique can be characterized as a piece of software for automatic translation of computational mesh from the *QW-3D* environment to the *ANSYS Fluent* environment. It is designed for using in modeling of problems where a microwave absorbing object is heated in a hybrid oven fed with microwaves and also containing a heat radiating element. In such setting, microwave volumetric heating is combined with radiative surface heating. Any loads are allowed as long as their properties (permittivity and thermal properties) are correctly defined.

Figure 4. Internal space of the Ceralink MAT™ furnace

From the viewpoint of practical exploitation of the coupled modeling tool, it is operational with any versions of *QW-3D* and *ANSYS Fluent*: a specific model to be computed is prepared and set up manually, so this is up to the user to accommodate any version-related features of both simulators.

Brief Theoretical Description of the Modeling Technique

In this section, we present the EM-TH model which handles not only the heat flow within the sample, but also the radiation heat exchange between the heaters and the material sample. The following heat flow equation

$$c_p \rho \frac{du}{dt} - \nabla\left(k \nabla u\right) = \frac{1}{2} \omega \varepsilon_0 \varepsilon_r''(u) \,|\, E(u) \,|^2 \qquad (1)$$

where E is the intensity of the electric field, and the medium properties, thermal conductivity k, specific heat c_p and density ρ, are all functions of temperature u, is solved in the volume of the lossy materials where heat is induced by microwave energy. The TH-EM coupling results from the temperature dependence of the electric properties (dielectric constant ε_r' and the permitivity ε_r''), while the reverse EM-TH coupling stems from the source term function.

The BCs of the TH part are modified so that they include incident and emitted radiative heat fluxes given by equations (2) for the sample walls. Thus, the stage at which the solution of (1) is obtained needs to be preceded by solution of (2) for all surfaces of the sample-cavity system resulting in a modification of the BC's at the kth surface at the sample wall according to equation (3). Total radiative fluxes emitted from and incident on the kth surface are given by

$$(q_r^{out})_k = e_k \sigma u_k^4 + r_k (q_r^{inc})_k \qquad (2a)$$

$$(q_r^{inc})_k = \sum_j (q_r^{out})_{j,k} \qquad (2b)$$

where $(q_r^{out})_{j,k}$ is the heat flux emitted by the jth surface and incident on kth surface, e_k and r_k are the emissivity and reflectivity of the k-th surface, respectively, and σ is the Stefan-Boltzmann constant. In this paper, it is assumed that $r_k = \boldsymbol{r} \equiv 0$ and by Kirchoff's law, all energy of the incident heat flux is absorbed, i.e.:

$$(q_{total})_k = h_k (u_k - u_\infty) + (q_r^{inc})_k - e_k \sigma u_k^4 \qquad (3)$$

where h_k is the heat transfer coefficient at the kth surface, and u_∞ is the temperature of the surrounding fluid.

In cavity heat exchange, the largest difficulty lies in evaluating what portion of the total heat flux emitted by the kth surface reaches the jth surface. In the reported technique, the surface-to-surface (S2S) model [11] of radiative heat exchange is employed to solve the equations (2) for the radiative fluxes. In the S2S model, the heat flux exchanged between the jth and kth surfaces is evaluated based on their mutual orientation in space using the configuration factor $F_{j,k}$. As a result, the total heat flux incident on the kth surface is given in the S2S model by

$$(q_r^{inc})_k = \sum_j F_{j,k} (q_r^{out})_j \qquad (4)$$

Operational Characteristics of the Modeling Technique Evaluation

Software Performance

The developed modeling tool is believed to be original; related literature reveals no analogous algorithms or computational procedures. Moreover, there are also no appropriate benchmarks which could be used for testing the performance of the developed technique:

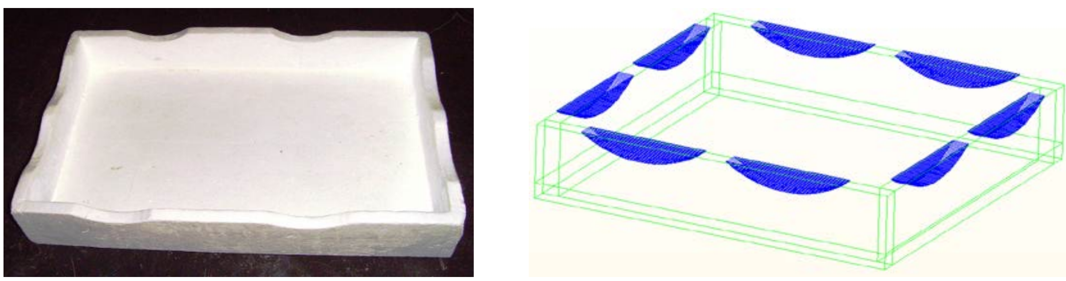
computational results produced by the technique described in [7] have been found inconsistent with independent experimental verification and this cannot be considered a credible source.

Therefore, Ceralink currently works on preparation of a series of experiments in hybrid thermal processing of limestone samples – the output of this work will allow for direct comparing with computational results and thus verification of the modeling technique and its implementation.

In this situation, the accuracy of the modeling technique was analyzed by considering the modeling results in comparison with reasonable physical expectations. On a more fine level, a special effort was put into checking if the model is stable for any temperature of the heating elements. It was established that with correct setting up of the thermal initial condition such stability can be ensured.

Software Implementation

In terms of hardware requirements, the technique described above is operational at a regular modern PC platform suitable for running *QW-3D* and *ANSYS Fluent*. For high productivity of simulations, advanced multiprocessor systems and GPU processing would be desirable for most practical problems. An example of the translation from real objects into the model representation is shown in Figure 5.



(a) (b)

Figure 5. Mullite tray for the limestone samples – general view (a) and representation of the model (b).

At the present stage of development of the modeling technique and its implementation, no special documentation (user's manual or guide, etc.) is available. Many aspects of practical development of the models, however, are reflected in the related documentations of the *QW-3D* and *ANSYS Fluent* packages. To run a model with the reported modeling tool, a skilled operator/user with experience in running *QW-3D* and *ANSYS Fluent* is required.

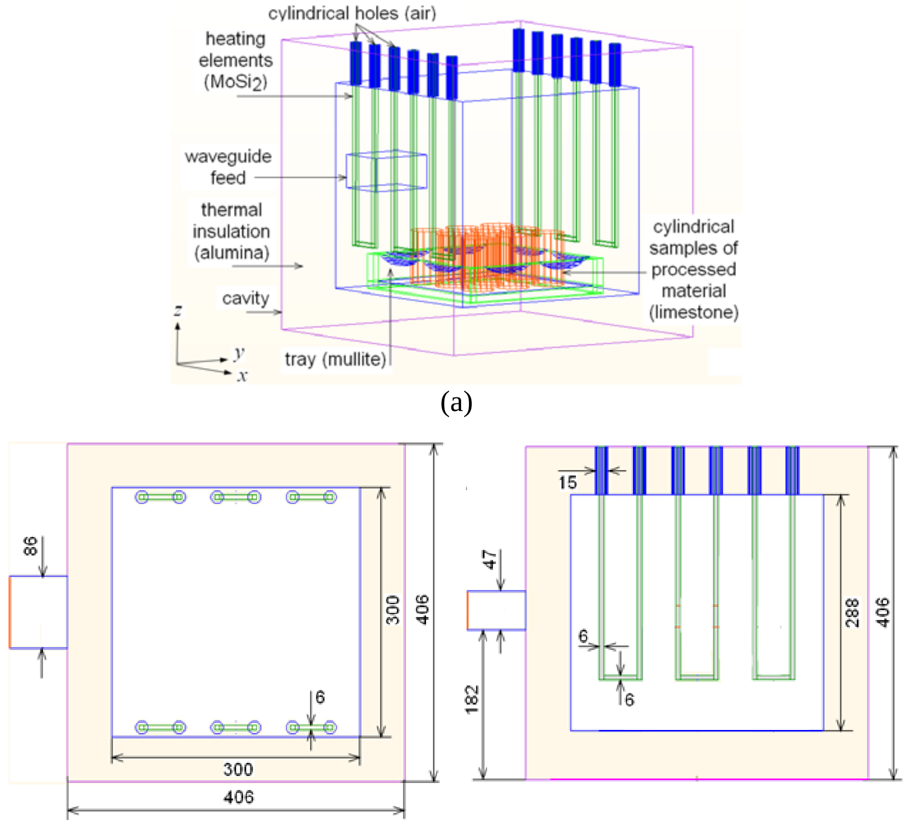
Application of the Modeling Technique to Hybrid Heating of Limestone

Microwave System

The hybrid microwave furnace considered in this study is Ceralink's MAT™ kiln (Figure 4) consisting of a cubic metal cavity with edge length 406 mm. The cavity walls are lined with a 51 mm thick layer of ceramic insulation with 80 wt% alumina (Al_2O_3). A mullite tray (Figure 4) is used to hold the limestone samples in the cavity. In order to speed up the calcination process, six U-shaped heating elements made of molybdenum disilicide ($MoSi_2$) are installed in the cavity near the two side walls. The microwaves are delivered into the furnace through a rectangular waveguide connected to the vertical wall opposite to the door. In a typical process, 2 kW of microwave power is delivered to the cavity, and the heating elements are kept at constant temperature of 1,000 °C.

Details of the Model

The system was represented in a 3D, fully parameterized model developed for *QW-3D*. All geometrical features of the furnace were precisely reproduced in the model, except two elements: the cylindrical rounded tubes of each U-shaped heater were approximated by three straight segments of square tubing (Figure 6b), and limestone pieces of normally irregular shapes were represented by some geometrically simple bodies; in this paper, we consider a set of nine vertical cylinders symmetrically located in the tray (Figure 6a).



(a)

(b)

Figure 6. **3D view of *QW-3D* representation of the MAT^{TM} furnace (a) and geometrical characteristics of the furnace's elements (b).**

The material parameters (dielectric loss-ε_r', permittivity-ε_r'', density-ρ, heat capacity –c, and thermal conductivity-k) of all microwave absorbing materials in the furnace (i.e., processed limestone, alumina-based ceramics, and mullite) were experimentally determined prior to modeling as temperature-dependent characteristics in the interval 20-1000 ^{o}C. Because of the high electric conductivity-σ of molybdenum disilicide, the material of the heaters was assumed to be metallic with “imperfect” (finite) σ (5×10^5 S/m) represented only by the surface.

Referring to the magnetron’s pulling effect, operating frequency of the system was determined to be 2.431 GHz: dedicated computations showed that the reflection coefficient turned out to be lowest at this frequency regardless of the temperature of the sample and the insulation. To ensure high accuracy of the solution of the primary electromagnetic problem, the FDTD mesh was optimized and made non-uniform through a special sensitivity analysis; nearly 800,000 cells were employed in the EM and TH models. As many as 30,040 surfaces were considered during calculations of the view factors F. In all the simulations, the heating time step $\Delta\tau$ was set at 10 s. FDTD stands for Finite-Difference Time-Domain. The physical volume of the problem to be solved with the FDTD method is discretized into small sub-volumes, or cells, and the FDTD mesh (or grid) is what is used to partition/discretize the volume into those small cells.

Modeling Results

Initial modeling was conducted for microwave only heating (no radiant heat). The examples in Figure 7 and Figure 8 show the temperature evolution of the inside of the MAT kiln with 9 cylinders of limestone for microwave only heating. The microwave only heating is being optimized for lowest reflection coefficient (most efficient usage of microwave energy), which will be used as input for the hybrid heating model. The microwave only heating (Figure 9 and Figure 10) shows that the temperature rise for the limestone is fairly slow with some non-uniformities for the microwave only. It was expected that the addition of radiant heat would increase the heating rate and also improve the heat distribution.

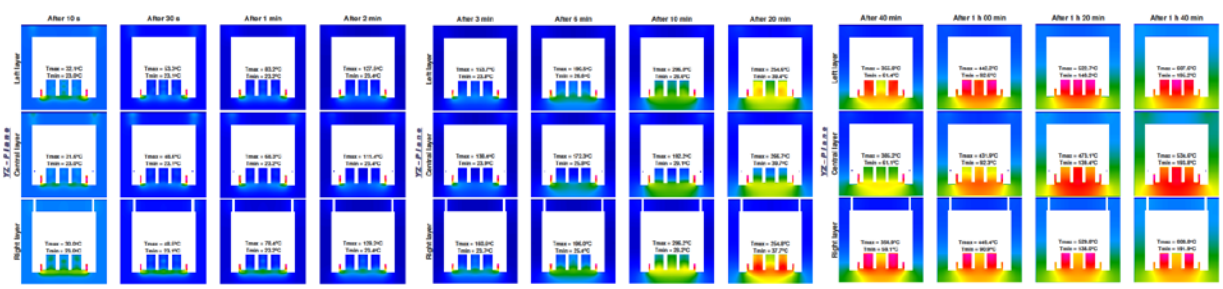


Figure 7. View of the temperature evolution through the YZ plane (side view) from 10 seconds up to 1 hour 40 minutes for microwave only heating.

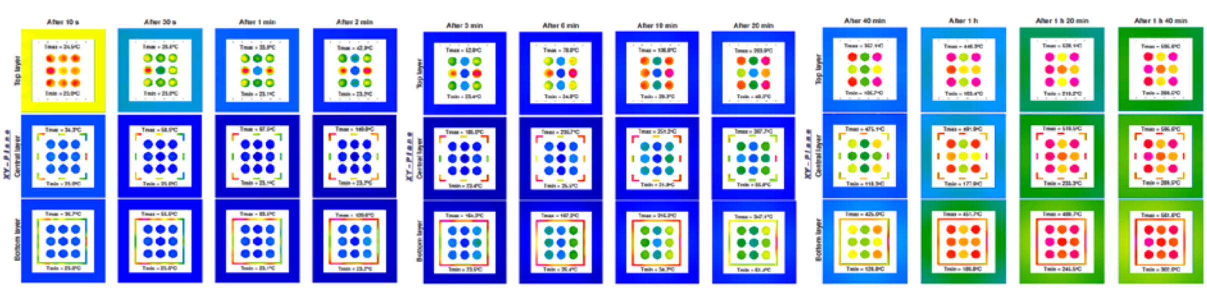


Figure 8. View of the temperature evolution through the XY plane (top view) from 10 seconds up to 1 hour 40 minutes for microwave only heating.

The main features of the heating process are characterized by the computed time-temperature history and 3D temperature field. The results of temperature simulations performed for pure microwave and hybrid heating are presented in Figure 9 and Figure 10. As expected, the hybrid heating greatly contributes to faster growth of the temperature in cavity and notably reduces the differences between the minimum and maximum temperatures. After the initial temperature rise observed for the hybrid heating scenario, the heat exchange becomes less intensive. This also agrees with expectations, as the radiative heat flux is proportional to the temperature difference between the heating element and the sample surface. As the latter becomes hotter, the radiative heat exchange ceases to dominate, and the micro-wave heating contribution becomes more pronounced making the two sets of temperature history curves look similar.

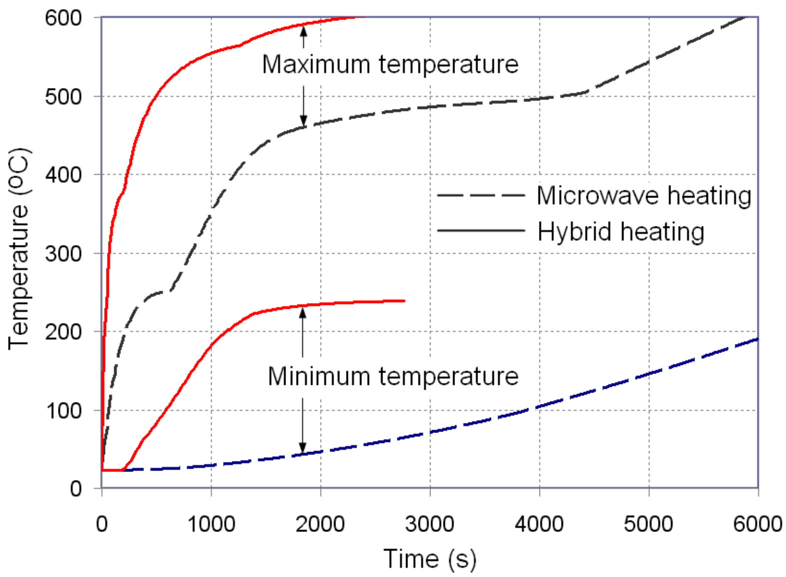


Figure 9. Time-temperature histories of the maximum temperature in the furnace for microwave and hybrid heating with 2 kW power.

The temperature distributions in the cavity cross-sections (Figure 10) suggest that with pure microwave heating, the mullite tray that absorbs most of the energy whereas the limestone samples remain relatively cool. In the case of hybrid heating, the heat exchange between the heating elements and the cavity mostly affects the temperature of the sample. Strong heating of close insulating layers is also observed.

These results provide an eloquent illustration of functionality of a new computational tool designed for modeling of hybrid (heat radiation and microwave) heating in specialized applied systems. The solver enabling coupled operations of two commercial EM and CFD simulators are used to model the high-temperature processing of limestone in Ceralink's MAT^{TM} hybrid furnace. This required the development of new linking mechanisms accounting for radiant BC's. The resulting tool allows simulating temperature distributions generated by hybrid and by purely microwave heating. Comparison of the heating regimes gives us an insight into the operation of the hybrid system and thus should be beneficial in their computational analysis and computer-aided design.

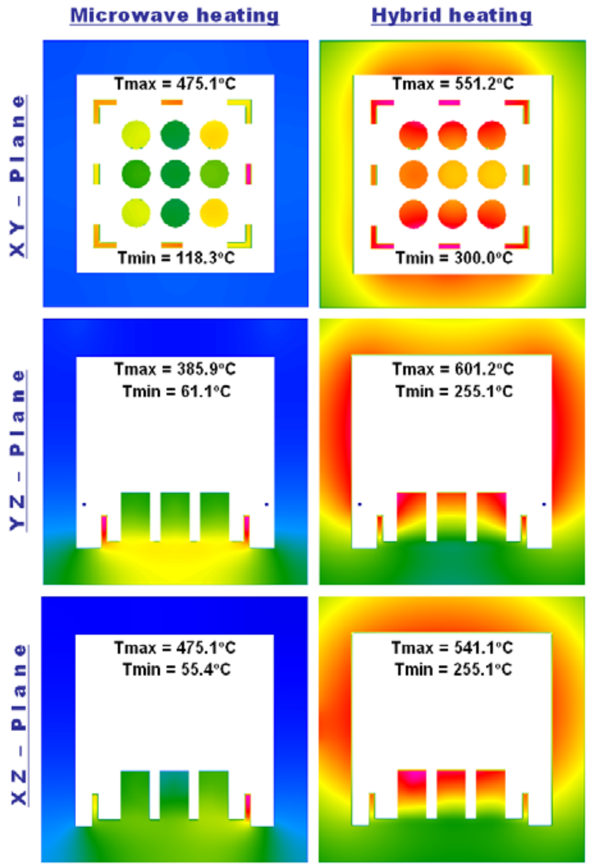


Figure 10. **Temperature distributions in the central coordinate planes after 40 min of heating.**

Pending Publications

Several papers dedicated to the developed modeling technique, its implementation and modeling results are currently under preparation. The first paper has been recently included in the program of one of the most prestigious forums in microwave technologies – the 2012 IEEE MTT-S IMS [11]. The paper was peer reviewed – it was accepted as a result of a scrutinizing evaluation of the 2012 MTT-S IMS Technical Review Program Committee. The second one will be submitted within two months to a major event of 2012 in the field for microwave power engineering – the 2nd Global Congress on Microwave Energy Applications, Long Beach, CA, July 2012.

Development of Laboratory MAT Calcination Method

Ceralink conducted research on both microwave and conventional calcining, with specific emphasis on the dissociation of limestone. Lime is created by heating the mineral limestone (calcium carbonate, $CaCO_3$) to temperatures exceeding 900 °C, which causes a shift to a favorable Gibbs free energy for the reaction of $CaCO_3 \rightarrow CaO$ (lime) + CO_2. The reaction is strongly endothermic; therefore, the material absorbs heat energy as it reacts. As a result, heat does not transfer into the limestone until outer layers have reacted. The reacted lime does not change volume significantly, and remains very porous and thermally insulating after calcinations, which impedes the flow of heat to the unreacted limestone. This creates a slow process to fully calcine the lime. The evolution of carbon dioxide creates a further impediment by raising the temperature at which the reaction is favorable. Ultimately full calcination is reached resulting in a 44% weight loss, without significant change in volume.

Over 65 laboratory scale calcining runs were conducted for 3 different grades of limestone supplied by the industrial partner (see Appendix A for a list of the runs). MAT and conventional processing conditions were developed to achieve 43-44% wt. loss, which indicated full conversion of limestone into lime. Initially, 1 kg and then 5 kg batches were processed in a laboratory kiln and analyzed. The set-up of the MAT kiln allowed for direct comparisons to be made between MAT and conventional processing, by simply having the microwave energy turned off, or on. All other variables, such as sagger, load, material type, and kiln were kept the same.

Results of these calcining comparisons indicated that the addition of microwave energy enhanced the disassociation reaction rate, e.g. shorter time required for MAT processing vs. conventional. This was tested in two ways, 1) by running same processing parameters and comparing the weight loss, and 2) by optimizing the weight loss for each method and comparing the time and energy consumption.

Evidence of the enhanced reaction was observed when a series of 1 kg samples were heated to 1200 °C and the weight loss, time and energy consumption were monitored. For the same processing conditions, (1200 °C, 0 dwell time) the MAT processed sample had a weight loss of 43.1%, while the conventionally processed sample was lower at 35.7%. This can be observed visually in Figure 11A and Figure 11B, where the gray area represents remaining uncalcined limestone. There are a couple small specks of gray in the MAT samples, while a large gray core remained in the conventional sample.

A) MAT- 0 min dwell, 43.1% wt loss **B) Conventional - 0 min dwell, 35.7% wt loss**

Figure 11.Images of calcined lime "stones". The gray core indicates some uncalcined material remaining.

In another set of runs, where the weight loss was optimized for each heating method, a 1 kg sample heated using MAT required a 10 min dwell for full calcination (43.3%), while conventional required 60 min dwell (43.5%). The energy and time savings is shown graphically in Figure 12, where the MAT process used 18% less energy and 16% less time compared to the 60 min conventional run for the 1 kg sample size.

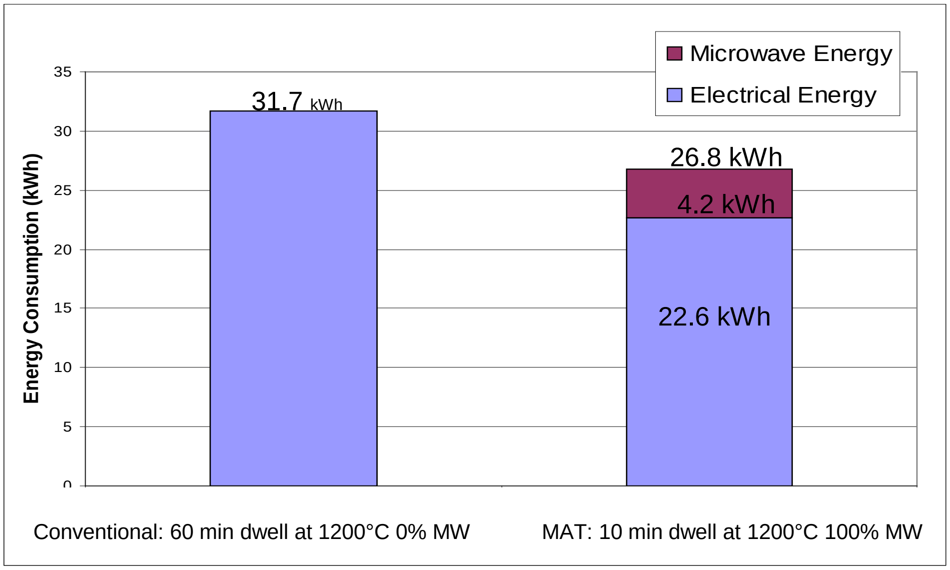


Figure 12. Energy consumption for 1 kg of limestone calcined to 1200 ºC for a weight loss of 43.5%. The MAT run consumed ~18% less energy and 16% less time (0 min dwell time) vs. the conventional with a 60 min dwell time.

Lime reactivity testing was conducted according to ASTM C 110 – 08. This test measures the reactivity of the lime by measuring the temperature rise when lime is mixed with water over a certain time interval (e.g. 30 sec, 3 min). Reactivity is an important property for lime, as some applications require a very high reactivity (water treatment) and others very low (food additives). A summary of the test results are shown in Table 1.

Table 1. Summary of lime reactivity testing

MRF Run	MLC	Weight Loss %	30 sec temperature rise (°C)	3 min temperature rise (°C)	total temperature rise (°C)	Run Parameters:
101	3	43.7	30.3	41.1	42.7	1 kg 30 min dwell @ 1200°C No MW
102	1	43.4	15.6	20.5	26.5	1 kg 45 mid dwell @ 1200°C No MW
103	2	43.0	41.1	46.3	50.1	1 kg 45 mid dwell @ 1200°C No MW
104	1	43.4	22.8	38.6	39	1 kg 10 min dwell @ 1200°C 100% MW
105	2	42.4	19.2	44.4	44.4	1 kg 10 min dwell @ 1200°C 100% MW
106	3	42.8	13.7	48.2	48.2	1 kg 10 min dwell @ 1200°C 100% MW
107	1	41.0	21.9	34.6	34.9	2 kg 10 min dwell @ 1200°C 100% MW
108	2	37.1	14	31.6	40.2	2 kg 10 min dwell @ 1200°C 100% MW
109	3	36.5	13.5	25.6	29.3	2 kg 10 min dwell @ 1200°C 100% MW
110	1	43.3	12.8	33.5	34.1	2 kg 45 min dwell @ 1200°C No MW
111	2	41.3	11	41.6	41.6	2 kg 45 min dwell @ 1200°C No MW
112	3	42.7	24.2	37	39.7	2 kg 45 min dwell @ 1200°C No MW

Refine MAT Calcining Method

It was theorized that the percent energy savings should increase as the load was increased, as a large mass is more difficult to heat by thermal conduction. More time is required time to move the heat through the load, while MAT should be more efficient at transferring heat into the material through volumetric microwave heating.

A series of larger scale 5 kg MAT calcining runs were conducted. A comparison of MAT vs. conventional was conducted where the MAT run achieved a 42% weight loss with a

120 min dwell and the conventional a 41% with a 240 min dwell. Overall, the 5kg MAT run consumed 23% less energy and had a 34% reduction in time, Figure 13. When compared to the 18% energy reduction for the 1kg run, the larger load resulted in a 27% higher energy reduction, indicating an increase in efficiency with a large load size. In addition, the larger load had a 140% increase in time savings (240 min) vs. conventional, compared to the 50 min time reduction for the 1kg run. It was anticipated that the time and energy savings (% difference) would increase further as the load size continued to increase.

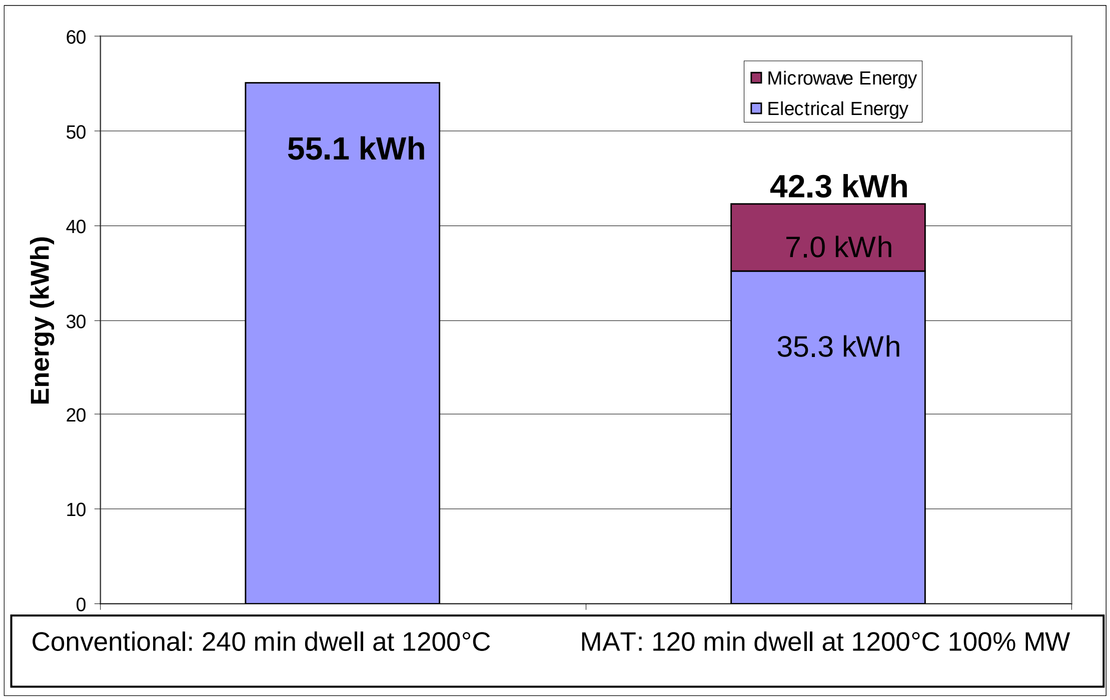


Figure 13. **Energy consumption for 5 kg of limestone calcined to 1200 ºC. The conventional weight loss was 41.2%, while the weight loss for the MAT run was slightly higher at 42.3%.**

At the beginning of the project, it was envisioned that microwave energy would be applied to the entire calcining process (e.g. along the length of the rotary kiln). However, after a trip to the industrial partner's manufacturing site, it was decided that this approach was not feasible due to 1) the thickness of the insulation in the rotary kiln, and accompanying issues of bringing microwave energy through the insualtion, 2) the rotating nature of the kiln, and the problems of attaching microwave equipment along the length.

The concept of microwave preheating and post-heating was then explored. A series of laboratory scale runs were set-up to examine microwave pre and post heating alternatives, where on the industrial scale, the microwave energy could be applied either in the stationary pre-heater or cooler sections.

The microwave pre-heating runs simulated adding microwaves to the pre-heating section of a rotary kiln by only applying the microwave energy from room temperature up to 700 °C. A conventional run with the same parameters was also conducted for comparison. The microwave pre-heating run only showed an increase of 1.4% weight loss over the conventional run.

Next, microwave post heating was explored as an alternative. The dielectric properties of the limestone are higher (better microwave absorbing) at higher temperatures, and therefore it was anticipated that the addition of microwave energy could provide more benefit as post heating treatment. Microwave post-heating runs were conducted by applying microwave energy from 900-1200°C. This run showed a weight loss increase of 3.5% from the conventional which is more than double the weight loss increase of the pre-heating run. It also had the equivalent weight loss of the MAT run, where the microwave energy was applied throughout the entire run. Therefore, the microwave post heat run provided the greatest energy efficiency for the highest weight loss. A comparison of these methods is shown in Table 2. These runs were all conducted with 1 kg of limestone.

Table 2. Summary of laboratory results comparing different methods

	Calcining Temp (°C)	Dwell time (min)	Microwave Temp range (°C)	Wt loss (%)	Energy (kWh/kg)
MAT	1200	0	25 -1200	39.0	24.1
MW Pre heat	1200	0	25 -700	36.9	23.5
MW Post heat	1200	0	900 -1200	39.0	23.6
MW Post heat	1200	5	900 -1200	42.2	24.6
Conventional	1200	0	0	35.5	23.4
Conventional	1200	20	0	43.2	26.5

While not in the original scope of work, Ceralink was able to conduct a set of scale-up runs testing microwave post heat experimentation in a 27 ft^3 MAT kiln (Figure 14), where the load size was increased to 22 kg. A series of calcining runs were conducted comparing MAT post heat and conventional. High alumina kiln furniture was positioned on the kiln car to form two troughs in which the limestone was placed as shown in Figure 15. The two troughs were labeled as front and back to keep track of the two different weight losses. The runs had a ramp rate of 5°C/min up to 1200°C. The damper stayed open and there was flowing air for all three runs. Microwave energy (20kW) was applied during the dwell only of the MAT runs to simulate a microwave post-heat. Energy data was collected and the weight loss was measured. The results are shown in Table 3.

The MAT pilot scale (22kg) post heat (run 2) with a 120 min dwell had a ~25% decrease in energy consumption compared to the conventional run (run 3); however, the MLC2 limestone grade was known to couple the least with microwave energy (MLC2 was available from industrial partner at the time). For run 4, MLC3 was used, and comparing the weight loss between runs 2 and 4, it’s evident that MLC3 heated more strongly by microwave energy and was fully calcined under the same conditions.

It was observed that in these larger batch runs, the stones calcined faster on the top layer of the stone bed, compared to the stones on the bottom. This concern is not seen on the production level, because the kiln rotates while energy is applied for dissociation. In the laboratory testing, microwaves are mainly applied from the top. In the solution being proposed, microwaves will be applied from all directions.

Figure 14. **Image of the 27 ft^3 MAT electric 1620 °C kiln at Touchstone.**

Figure 15. **The experimental setup which shows the high alumina kiln furniture positioned into troughs with limestone loaded on the kiln car.**

Table 3. **Summary of the results of the scale up runs performed at Touchstone**

Run	Grade lime stone	Dwell Time (min)	MW Power (kW)	Initial Mass (kg)	Final Mass (kg)	Weight Loss %	Initial Mass (kg)	Final Mass (kg)	Weight Loss %	Average Weight loss %	Energy (kWh)
1	MLC2	30	20	11.95	10.28	13.94	11.96	10.13	15.34	14.64	226
2	MLC2	120	20	11.97	7.31	38.91	11.96	7.13	40.43	39.67	317
3	MLC2	300	0	11.97	6.69	44.09	11.95	6.68	44.1	44.09	423
4	MLC3	120	20	11.95	6.65	44.35	11.95	6.65	44.35	44.35	252

Additional testing on a pilot scale will be required to determine the best application of microwave energy to existing lime kiln technology. This testing is necessary to determine the savings in a dynamic kiln. While the laboratory testing showed a potential savings based on the energy measured for the calcinations in a laboratory muffle furnace, there is no direct correlation between the lab furnace and a rotary or vertical kiln. The lab tests showed energy inputs of 31.7 and 55.1 kWh for 1 and 5 kg sample sizes. The energy required for the carbon dioxide disassociation is 0.977 kWh and 4.835 kWh respectively for the above two sample sizes. These numbers represent 3,244 % and 1,128 % above the minimum energy input required to disassociate carbon dioxide from limestone. These numbers show a large energy loss in the muffle furnace, but the improved efficiency of the sample size will diminish as the sample size increases.

Scale-up Design

The scale-up concept design was centered around MAT post heating, where the limestone would be discharged hot from the rotary section of the kiln at 1037 °C (1900 °F). The stones would fall into the upper part of the cooler section, which would be retrofitted with a narrower neck, measuring 6ft in diameter and 4 ft. long. A grate at the bottom of this chute would control the residence time of the stones, and allow them to dwell in a region of microwave energy for approximately 20 minutes, see Figure 16.

Assuming that the limestone is 70% calcined once it leaves the rotary part of the furnace, it was calculated that the energy of four 100kW microwave generators with 915Hz frequency would be necessary to impart enough energy to calcine the last 30% of limestone into lime, see Appendix A. The microwave generators would be situated on the platform, within 20 feet of the cooler section. The microwave energy would be delivered from the generator to the cooler section through microwave waveguide, which is hollow rectangular channeling. The waveguide from each generator would be split with a Y splitter, and attach to the chute in two locations. This will help to mix the microwaves, ensuring uniform microwave distribution from all sides.

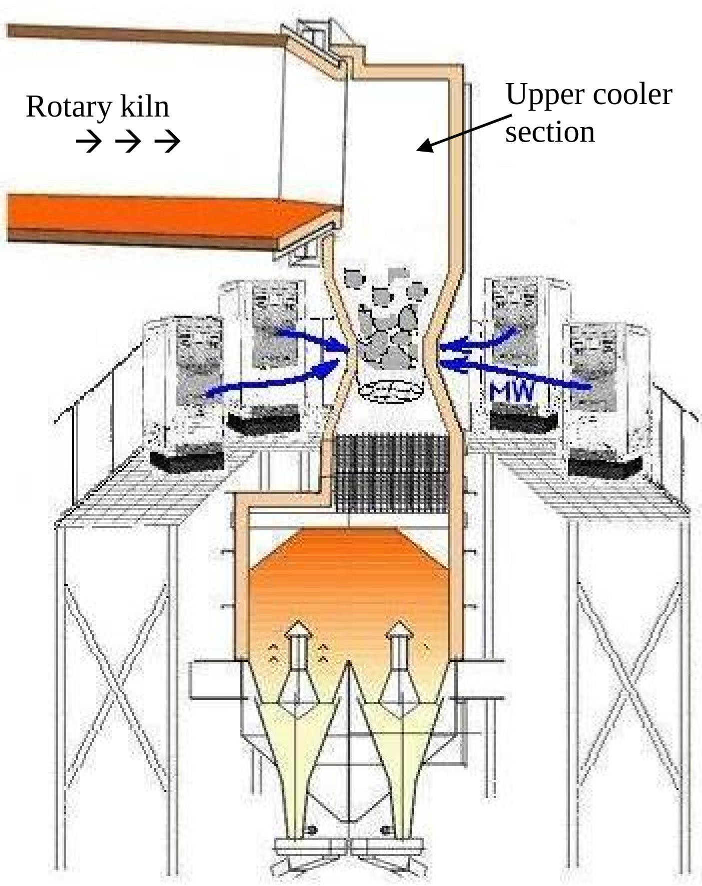


Figure 16. **Sketch of the post-heat section of a rotary kiln with microwave energy applied by four microwave generators and a slimmer section with a diameter of 6 ft and a length of 4 ft.**

Benefits Assessment

Ceralink worked closely with Mr. Johnney Bowers, who was able to provide information regarding the energy consumption for different size rotary calciners, along with a better understanding of the process flow. He also helped to examine and vet the implementation of microwave equipment to rotary and shaft kilns.

The impact of commercialization of MAT for the lime industry will serve as an important demonstration for many other energy intensive industries. For example, rotary calciners are used in processing of cement, metal ores, structural and electroceramic powders, and catalysts. MAT kilns can be used to save energy in the high temperature processing of thousands of products, such as refractories, insulators, metal casting molds, and filters. Ceralink worked with industry consultant to help examine energy implication of MAT for lime calcining.

The fuel consumption for a typical 1,200 tpd preheater rotary kiln being offered by the two major kiln system vendors[12] is 4.5 MMBTU/ton and 3.02 MMBTU/ton for a Maerz PFR twinshaft vertical kiln[13]. Since the fuel consumption in the kiln is a combination of the fuel to generate the heat energy for the disassociation of carbon dioxide from the limestone and the energy losses for the kiln system. The energy required to disassociate the carbon dioxide is 2.73 MMBTU/ton of lime. The amount of energy that can be reduced is the difference between the total energy being consumed by the system minus the disassociation energy (chemical energy required to cause CO_2). The disassociation energy cannot be reduced no matter whether it comes from heat or microwave energy, only the heat energy lost as latent heat in the product or waste gases other heat losses from the hardware. Thus a conservative estimated for the potential energy that can be reduced is 1.77 MMBTU/ton lime for the preheater rotary kiln (4.5 minus 2.73 mm BTU/ton) and 0.29 MMBTU/ton lime for the vertical kiln (3.02 mm btu/ton minus 2.73 MM BTU/ton)[14].

Based on demonstrated laboratory scale savings, a reduction of 25% in fuel consumption above the energy absorbed in the disassociation reaction, then the potential fuel reductions are 0.44 MMBTU/ton lime for the preheater rotary kiln and 0.07 MMBTU/ton lime for the vertical kiln. Assuming an average fuel cost of $4.00/MM BTU, then the savings for fuel consumption reductions are $1.76/ton lime and $0.28/ton lime, respectively. Assuming annual production rates of lime of 400,000 and 200,000 tons, in rotary and vertical kilns respectively; then the annual savings are $704,000 and $56,000 *per kiln* [14]. Based on an estimated retrofit cost of $1 mil (including the microwave equipment), the payback for a microwave post heat section on a rotary calciner is 1.4 years, and on the order of 18 years for the vertical. Implementation for the rotary calciners would make the most energy impact, as 80% of lime is produced using these types of kilns and the payback is less than 2 years.

MAT implementation for the total US lime production represents a potential $29 million dollars in savings for the US industry and a 7.3 trillion BTU of energy savings, see Table 4. Even though this project was focused on lime calcination, the process for cement calcination also includes the calcination of limestone, and with some development work MAT should be equally applicable. Using the energy and economic estimates for the rotary lime kiln, if the US cement industry is included in the energy and economic savings, the estimated potential yearly savings is an additional 39 trillion BTU and $155 million as cement production is roughly 5x that of lime, see Table 5. *Note, calculations for the energy, economic and environmental benefits are shown in Appendix C.*

Table 4. Projected Annual Energy and CO_2 Savings for MAT Implementation for US Lime Production

Kiln type	Production MM tons lime/yr	MAT Energy reduction (MMBtu/ton)	Energy savings/yr (TBtu)	Environmental Benefit CO_2 (Mlb)	Yearly $million savings
Rotary (80%)	15.8	0.44	7	258	$28
Vertical (20%)	4	0.07	0.3	10.3	$ 1.1

assuming US production is19.8 million tons lime per year [15], and a 25% energy savings

Table 5. Estimated Annual Energy and CO_2 Savings for MAT Implementation for US Cement Production.

	Production MM tons cement/yr	MAT Energy reduction (MMBtu/ton)	Energy savings/yr (TBtu)	Environmental Benefit CO_2 (Mlb)	Yearly $million savings
Rotary	88	0.44	39	3,250	$155

assuming US production is 88 mil tons cement per year [16], and a 25% energy savings

The addition of MAT to the discharge of rotary kilns, then the potential benefits can be in the increased capacity of a system, product quality improvements, and environmental issues. With the ability of MAT to complete the calcinations of the inside of lime particles then the capacity of existing or new kilns can be increased for less capital than standard systems. A significant amount of energy is required to drive the heat into the center of the particles and maintain that temperature long enough for the carbon dioxide to migrate to the outside of the particle while the density of the outer shell is decreasing making it harder for the carbon dioxide to migrate. Assuming a 1,200 tpd preheater rotary kiln system would cost about $30,000,000 or $25,000 per tpd of capacity. If the MAT addition to this system could increase system capacity by 10% (120 tpd) the capital savings would be $3,000,000 versus an estimated $1,000,000 cost. Thus a potential for capital savings may be a benefit of MAT retrofit or inclusion in kiln systems.

Another potential benefit of MAT if it indeed can complete the calcinations of the interior of the largest particles or pebbles then the quality of the lime will be improved thus allowing increased prices for the higher quality. This benefit and the value will be site specific dependent on the applications and the competition. Additionally if all of the available limestone can be calcined then a slightly lower quality limestone could be used to produce the target lime product quality. The potential savings would be related to limestone reserve utilization, again specific to the quarry, reserves and product requirements.

Application of MAT reduces the fuel consumption thus reducing all of the criteria pollutants, including carbon dioxide, nitrogen oxides, carbon monoxide and sulfur oxides, which are limited in permits. While MAT has potential to reduce fuel consumption, there is an even larger potential reduction in nitrogen oxides, as their generation increases with higher temperatures in the burning zone. Normally in the firing zone the flame temperature will be maximized to provide the delta temperature to drive the heat to the center of the particles or stones. Reduced pollution emissions can make permitting easier or in some cases make the difference between getting and not getting a permit.

<u>Limitations or Impediments to MAT</u>

The application of microwave energy to preheater rotary kilns is limited to the preheater area or the cooler section since the rotary kiln section is constantly rotating. However the application of microwave energy to preheater rotary kilns or vertical kilns depends on additional information about the application of the microwave energy to the limestone. Application information needed to determine how to apply the energy to a specific kiln is as follows:

- How much exposure is needed?
    - Is there a length of exposure required (time required for microwave energy to be absorbed by the limestone)?
    - Is there an energy or microwave intensity that is most effective?
    - What are the controlling parameters for absorption?
        - Temperature, Particle size, Particle size distribution
        - Type of limestone
            - Grain size
            - Fractures
            - Impurities

- What are the dynamics of the beam/stone interface?
    - How deep into a stone bed does the microwave penetrate?
    - Is the microwave a point beam, fan shaped or other geometry?

Commercialization

The MAT technology that is being developed can be applied to several mature commercial industries related to the research performed under this project. This includes the primary industry of lime for cement, steel, and glass manufacturing, as well as secondary industries such as iron ore, catalyst, petroleum coke, and ceramic proppants manufacturing. Each of these industries utilizes "pyroprocessing", or calcination of raw materials to effect specialized chemical reactions to achieve the final product. As well, these industries utilize large rotary kilns that are thermally inefficient that could benefit from incorporation of the targeted microwave heating that MAT offers.

Some of the major target industries and applications where MAT could be effectively integrated:

- ***Lime:*** Calcining limestone into lime for use in cement, steel, and glass industries.
- ***Steel:*** Sintering of iron ore pellets for primary steel production in blast furnaces
- ***Aluminum:*** Calcining bauxite for aluminum smelting
- ***Catalyst:*** Calcining of industrial catalysts
- ***Petroleum Coke:*** Calcining petroleum coke for use in aluminum smelting anodes and electric arc furnace steel electrodes
- ***Proppants:*** Sintering of ceramic proppants for use in oil and gas drilling
- ***Kaolin:*** Calcining kaolin to produce anhdrous clay with high brightness
- ***Carbon Black:*** Production of carbon black

Market Analysis

The market can be grouped into (1) minerals/ore processing, (2) specialty chemical calcining, and (3) sintering processes. Minerals and ore processing comprises the lime, bauxite, and other heavy ore processing, falling into the pyrometallurgical classification. These processes generally consist of taking raw ores and subjecting them to intense heat for the purposes of causing decomposition or other reactions. Specialty chemical calcining encompasses such applications as petroleum coke, catalysts, pigments, and other industrially important materials. The sintering processes, such as iron ore and ceramic proppants, are used to fuse together powder mixtures into pellets or other shapes for ease of use in other industries. Together, these target market industries mostly utilize large and rather inefficient rotary kilns to process their materials and could greatly benefit from the MAT research completed under this project.

A preliminary U.S. market analysis of just the cement industry, which includes the process of calcining limestone, demonstrates that this alone is a $10B market with a large global affiliate presence. The U.S. currently has an estimated 118 cement manufacturing facilities operating 192 kilns, which is by far the biggest use for lime calcination processes. The five largest companies operating in the U.S. (in terms of capacity) control nearly 55% of the market and are Holcim (Switzerland - 13.1%), Lafarge (France - 12.7%), CEMEX (Mexico - 12.2%), Heidelberg Cement (Germany - 8.25%), Ash Grove Cement Co. (United States - 6.5%). [17]

The production trend over the last 5 years for the US lime production is shown in Figure 17 [18]. This shows that lime production for 2010 was on the order of 18 million metric tons per year, after a drop in 2009. The cost of quicklime and hydrated lime are also included, and show the value of lime has been increasing over the last 5 years.

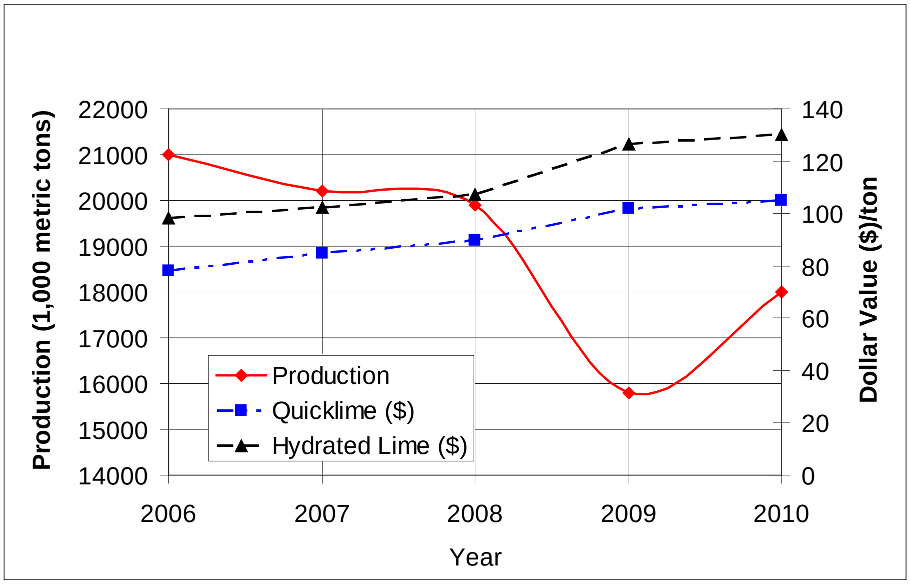


Figure 17. Graph of US lime production over the past 5 years, showing an increase for 2010. Also showing the dollar value of lime had increased from 2008 to 2009, but had a smaller increase 2009-2010.

The critical need that the MAT technology offers is that of producing significant energy savings in the form of reduced natural gas consumption and the potential for higher throughput using existing infrastructure. The value proposition being offered by the technology is that end-users can reduce their energy consumption due to kiln operations by up to 50% and potentially increase their throughput by up to 34%. By incorporating this new MAT heating technology, these customers can reap the benefits while spending a fraction of the capital costs for modification compared to the investment required in a new modern furnace design with only incrementally better efficiency.

Market Barriers

MAT technology has some market barriers to entry that will continue to be minimized through continued research, pilot, and demonstration-scale technology investments being pursued over the next several years. Within the lime calcination and cement industries, significant capital infrastructure is in place and resistance to completely scrap the current decades-old calcining technology is high. Ceralink has structured its research, development, and deployment plan to focus on rather low impact retrofits to the current post-heating zone of the process that still result in impressive potential energy savings.

Additionally, lime companies have slimed staffs and do not have the technical capacity or available technical staffs to support a large technical project like designing, building and demonstrating a new technology like MAT. There may be significant resistance for companies to take the cost-risk of retrofitting a MAT section themselves. Thus a grant to pay for the modifications or a significant portion of it will help spur implementation.

Competition

In considering the competitive landscape for the MAT technology, there is relatively little in the area of applying the same type of specialized direct thermal heating to the product that the MAT technology offers. There have been significant advances in the adoption of fluidized bed technology for lime calcination and cement production, as well as in the other target industries. In this technology, the most compelling facet has been in the superior combustion efficiency owing to the intimate mixing with the moving bed of material. This has allowed the use of cheaper grades of coal and other low-cost fuel sources such as biomass to become attractive.

Fluidized-bed systems are estimated to have capital costs equivalent to 88% of the capital costs of a modern cement facility and operating costs equivalent to 75% of a modern cement facility's operating costs. However, their operating and capital costs are not less than the operating costs of older, fully capitalized kiln-based plants, slowing their adoption, since they are likely to be considered only for future capacity expansion. Another barrier to adoption is the natural reluctance to invest in the large capital expenditures associated with new plant construction. The risks associated with building a new technology commercial-scale plant, based on the successful demonstration of a one-tenth scale facility, are considered high [17]. Thus, the chances of industry adoption of MAT, which offers a low impact retrofit to a small footprint area of the overall process, are significantly higher than for fluidized bed technology.

Marketing and Commercialization Approach

The approach for commercialization of this technology will require 1) continued pilot scale calcining demonstrations, 2) involvement of lime kiln companies, and 3) involvement of an industrial microwave equipment provider. Ceralink identified two builders of lime kilns whom could be approached to partner with for continued technology and equipment development. Eventually, Ceralink plans to license and transfer the technology to the kiln builders once the technical aspects are finalized. The lime company could then implement the MAT post heating sections in new builds and pursue retrofit of existing lime kilns in order to improve their efficiency. Retrofits would likely be attractive to lime manufacturers, as the purchase of a new lime kiln is on the order of a $30 million dollar investment, where as a MAT retrofit would be on the order of $1 million. Ceralink would receive a royalty from the kiln builder.

Current scale-up designs indicate that four 100kW microwave generators (915 MHz frequency) will be required to provide sufficient energy to a chute measuring 6ft in diameter and 4 ft. long to finish calcining the limestone before the limestone is cooled. This is sufficient scale for a production size rotary calciner with 600 ton/day output. Ceralink has identified a microwave equipment supplier that could easily supply the required microwave equipment and engineering work to assist with integration. The cost of the microwave equipment is on the order of $600k (Appendix B).

Market Impact

As identified in the most recent comprehensive analysis of the lime and cement calcination industry commissioned by the Department of Energy's Industrial Technologies Program in 2003 [17]:

"The greatest opportunities in reducing energy consumption and lowering emissions associated with cement/concrete will be obtained with improvements in cement pyroprocessing. On average, pyroprocessing systems in the United States operate at about 34% thermal efficiency. This low efficiency implies that there are potentially significant process and system improvements to improve energy and environmental performance. These process improvements will come from better energy management, upgrading existing equipment (e.g., replacing wet kilns, upgrading to preheater and precalciners), adopting new pyroprocessing technologies (e.g., fluidized bed systems) and, in the longer term, performing the R&D necessary to develop completely new concepts for the cement manufacturing processes."

Thus, Ceralink 's MAT technology fits within the ITP cross-cutting technology mission and has a high chance of both technical and commercial success, as there is already a market pull for this type of critically needed technology.

Ceralink has 11 years of experience in microwave processing of materials and commercializing systems and processes. A Microwave Technology Center was established to facilitate the uptake of microwave processing in a wide range of industries that require the heating of ceramics, glass, metals, and polymers. Ceralink engineers have direct experience with using microwaves for drying, curing, chemical processing, calcining, binder removal, and sintering. In addition to process development, relevant research includes the development of modeling software that uses thermal and dielectric properties to simulate microwave heating. Relevant commercial efforts include the in-licensing of patented Microwave Assist Technology (MAT) from the UK, demonstrating and building MAT furnaces, and successfully licensing to a US kiln manufacturer. Ceralink assembled a team to build a scaled up industrial MAT furnace for a customer, and continues to grow this business. Ceralink also established a joint venture company to market research microwave systems.

Given this experience, the marketing and sales channels to be utilized include directly licensing to OEM kiln and industry plant providers, as well as to aftermarket custom kiln designers and re-designers utilizing the MAT technology. Is it estimated that the investment to bring this technology to the commercial level is on the order of $3 million. Table 6 shows the proposed investment timeline over the next 5 years.

Table 6. Estimated funding required to commercialize MAT for lime calcining

Funding Profile				
	FY11	FY12	FY13	FY14
DOE Office Investment	220,000	800,000	500,000	500,000
Cost Share / Other	60,000	100,000	150,000	150,000
NYSERDA		200,000	200,000	
Project Total	280,000	$1.1 MM	$850,000	650,000

Accomplishments

Ceralink has accomplished the following in line with the project objectives:

- 70 laboratory calcining experiments were conducted (both MAT and conventional) where the energy consumption was monitored and comparisons made
- Processing parameters were developed to obtain >43% weight loss (both MAT and conventional)
- 4 scale-up runs were conducted to test the theory that MAT efficiency increases with larger load sizes
- A new module was built to incorporate radiant heat into a microwave modeling program
- Simulations were conducted to determine the uniformity of heating within the limestone and compare microwave only, radiant only, and MAT heating modes.
- Microwave post heating was identified as the most feasible method to incorporate microwave into production scale lime processing
- A design concept was developed and budgetary costs estimated

In addition, Ceralink presented project progress and results at the following conferences:

- 2011 ACEEE Summer Study on Energy Efficiency in Industry. 2011. Niagara Falls, NY: American Council for and Energy-Efficient Economy. [19]
- Materials Science and Technology 2011 Conference and Exhibition 2011: Columbus, OH. [20]
- 35th International Conference and Expo on Advanced Ceramics and Composites 2011: Daytona Beach, Fl.

Conclusions

In this study, the microwave materials interactions were studied through dielectric property measurements, process modeling, and lab scale microwave hybrid calcination tests. Characterization and analysis were performed to evaluate material reactions and energy usage. Processing parameters for laboratory scale and larger scale calcining experiments were developed for MAT limestone calcination. Early stage equipment design concepts were developed, with a focus on microwave post heating treatment. The retrofitting of existing rotary calcine equipment in the lime industry was assessed and found to be feasible.

Ceralink sought to address some of the major barriers to the uptake of MAT identified as the need for 1) team approach with end users, technology partners, and equipment manufacturers, 2) modeling that incorporates kiln materials and variations to the design of industrial microwave equipment. This project has furthered the commercialization effort of MAT by working closely with an industrial lime manufacturer to educate them regarding MAT, identifying equipment manufacturer to supply microwave equipment, and developing a sophisticated MAT modeling with WPI, the university partner.

MAT was shown to enhance calcining through lower energy consumption and faster reaction rates compared to conventional processing. Laboratory testing concluded that a 23% reduction in energy was possible for calcining small batches (5kg). Scale-up testing indicated that the energy savings increased as a function of load size and 36% energy savings was demonstrated (22 kg). A sophisticated model was developed which combines simultaneous microwave and conventional heating. Continued development of this modeling software could be used for larger scale calcining simulations, which would be a beneficial low-cost tool for exploring equipment design prior to actual building. Based on these findings, estimates for production scale MAT calcining benefits were calculated, assuming uptake of MAT in the US lime industry. This estimate showed that 7.3 TBTU/year could be saved, with reduction of 270 MMlbs of CO_2 emissions, and $29 MM/year in economic savings. Taking into account estimates for MAT implementation in the US cement industry, an additional 39 TBTU/year, 3 Blbs of CO_2 and $155 MM/year could be saved.

One of the main remaining barriers to commercialization of MAT for the lime and cement industries is the sheer size of production. Through this project, it was realized that a production size MAT rotary calciner was not feasible, and a different approach was adapted. The concept of a microwave post heat section located in the upper portion of the cooler was devised and appears to be a more realistic approach for MAT implementation.

Commercialization of this technology will require 1) continued pilot scale calcining demonstrations, 2) involvement of lime kiln companies, and 3) involvement of an industrial microwave equipment provider. An initial design concept for a MAT post-heat treatment section was conceived as a retrofit into the cooler sections of existing lime rotary calciners with a 1.4 year payback. Retrofitting will help spur implementation of this technology, as the capital investment will be minimal for enhancing the efficiency of current rotary lime kilns. Retrofits would likely be attractive to lime manufacturers, as the purchase of a new lime kiln is on the order of a $30 million dollar investment, where as a MAT retrofit is estimated on the order of $1 million. The path for commercialization lies in partnering with existing lime kiln companies, who will be able to implement the microwave post heat sections in existing and new build kilns. A microwave equipment provider has been identified, who would make up part of the continued development and commercialization team.

Recommendations

Based on the results of this demonstration project, Ceralink recommends that additional large scale calcining trials be conducted to further the understanding of microwave heating uniformity as the load size is increased, how impurity levels effect results, and to raise the interest of lime companies. It would also help refine the initial design concept into something that could be built on a pilot scale. Subsequently, pilot scale equipment with a small scale rotary calciner could be used to feed a microwave post heat unit. The energy consumption, cycle times and post heat design concept could be thoroughly tested, optimized and serve as a demonstration for industry acceptance.

Additional MAT modeling work would be useful to answer some of the questions pertaining to uniformity of microwave energy throughout post-heater section, effect of stone size, optimum positioning of microwave waveguide feeds and power/volume heating rate simulations. It was also suggested that the laboratory results could be more easily related to production rotary calciner projections through heat and mass transfer modeling, by monitoring the calcination times versus available calcium oxide at multiple calcining temperatures with and without MAT.

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Appendices

Appendix A- Summary of MAT and Conventional Calcining Runs

MLC 1 Data

Run number	MLC number	Dwell Temp (°C)	Dwell Time (min)	Microwave Percent	Initial Mass (grams)	Final Mass (grams)	Weight Loss %	MW Energy (kWh)	Electrical Energy (kWh)	Total Energy (kWh)
MRF 074	1	1350	10	50	1050	670	41.9	2.31	26.21	28.52
MRF 076	1	1200	10	50	1005	585	41.8	2.08	23.25	25.33
MRF 079	1	1200	10	0	1025	655	36.1	0	24.7	24.7
MRF 082	1	1200	20	50	1000	565	43.5	2.23	24.8	27.03
MRF 085	1	1200	0	75	1000	590	41	1.93	20.9	22.83
MRF 088	1	1200	0	0	999	641.42	35.8	0	23.3	23.3
MRF 092	1	1200	60	0	1004	567.13	43.5	0	31.7	31.7
MRF 093	1	1200	60	0	2006.31	1142.8	43	0	33.74	33.74
MRF 096	1	1200	0	100	1005	572.03	43.08	3.87	20.9	24.77
MRF 099	1	1200	30	0	1009.21	579.49	42.6	0	28.1	28.1
MRF 102	1	1200	45	0	1002.17	566.81	43.40	0.00	no data	no data
MRF 104	1	1200	10	100	1001.98	567.59	43.40	4.16	22.63	26.79
MRF 107	1	1200	10	100	2014.28	1188.89	40.97	4.18	23.69	27.87
MRF 110	1	1200	45	0	2009.36	1139.36	43.30	0.00	32.06	32.06
MRF 113	1	1100	90	0	5000	3843	23.14	0.00	37.55	37.55
MRF114	1	1100	90	100	5000	3109	37.8	6.13	32.24	38.37
MRF 115	1	1100	240	0	5000	2.95	41	0	55.05	55.05
MRF 116	1	1100	120	100	5000	2883.5	42.33	7.04	35.22	42.26
MRF 120	1	1100	120	0	2585	1.725	33.27	0	38.3	38.3

MLC 2 Data

Run number	MLC number	Dwell Temp (°C)	Dwell Time (min)	Microwave Percent	Initial Mass (grams)	Final Mass (grams)	Weight Loss %	MW Energy (kWh)	Electrical Energy (kWh)	Total Energy (kWh)
MRF 073	2	1350	10	50	1005	575	42.79	2.31	26.52	28.83
MRF 077	2	1200	10	50	1000	575	42.5	2.08	22.98	25.06
MRF 080	2	1200	10	0	1005	650	35.32	0	24.49	24.49
MRF 083	2	1200	20	50	1000	585	41.5	2.23	24.14	26.37
MRF 086	2	1200	0	75	1000.33	640	36	1.93	20.76	22.69
MRF 089	2	1200	0	0	1002	677.2	32.42	0	23.07	23.07
MRF 094	2	1200	60	0	1997.55	1133.12	43.3	0	N/A	N/A
MRF 097	2	1200	0	100	1039.89	645.31	37.94	3.86	20.5	24.36
MRF 100	2	1200	30	0	1024.27	585.3	42	0	27.72	27.72
MRF 103	2	1200	45	0	1016.86	579.38	43.02	0.00	29.78	29.78
MRF 105	2	1200	10	100	1012.2	582.89	42.40	4.16	22.09	26.25
MRF 108	2	1200	10	100	2008.67	1263.77	37.08	4.17	22.8	26.97
MRF 111	2	1200	45	0	2012.62	1170.39	41.20	0.00	31.68	31.68
MRF 117	2	1100	120	100	5000	3005	39.9	7.04	35.62	42.66
MRF 119	2	1100	240	0	5000	2985	40.3	0	55.37	55.37
MRF 121	2	1100	120	0	5000	3355	32.9	0	52.1	52.1
MRF 139	2	1200	10	50	1007.7	613.16	39.2	2.08	23.51	25.59
MRF 140	2	1200	10	50	1009.76	614.82	39.11	2.08	23.37	25.45
MRF 141	2	1200	10	50	1003.44	601.98	39.2	2.08	23.14	25.22

MLC 3 DATA

Run number	MLC number	Dwell Temp (°C)	Dwell Time (min)	Microwave Percent	Initial Mass (grams)	Final Mass (grams)	Weight Loss %	MW Energy (kWh)	Electrical Energy (kWh)	Total Energy (kWh)
MRF 075	3	1350	10	50	1010	560	44.55	2.3	26.35	28.65
MRF 078	3	1200	10	50	1000	575	42.5	2.07	23.15	25.22
MRF 081	3	1200	10	0	1000	585	41.5	0	24.8	24.8
MRF 084	3	1200	20	50	1000	565	43.5	2.23	24.62	26.85
MRF 087	3	1200	0	75	1000	593	40.7	1.93	20.89	22.82
MRF 090	3	1200	0	0	1014.54	688.04	32.18	0	23.08	23.08
MRF 091	3	1200	10	0	1007.86	620.09	38.47	0	24.83	24.83
MRF 095	3	1200	60	0	2007.3	1133.42	43.5	0	33.61	33.61
MRF 098	3	1200	0	100	1000.82	580.43	42	3.87	20.9	24.77
MRF 101	3	1200	30	0	1001.51	563.83	43.7	0	28.04	28.04
MRF 106	3	1200	10	100	1012.39	578.77	42.80	4.16	22.64	26.80
MRF 109	3	1200	10	100	2001.38	1269.83	36.50	4.16	23.08	27.24
MRF 112	3	1200	45	0	2001.76	1148.08	42.65	0.00	31.73	31.73
MRF 118	3	1100	120	100	5000	3235	35.3	7.04	35.54	42.58
MRF 122	3	1100	240	0	5000	3240	35.2	0	55.12	55.12
MRF 127	3	1200	20	50	1006.2	569.42	43.4	2.23	24.73	26.96
MRF 128	3	1200	20	50	1007.54	593.7	41.1	2.23	24.88	27.11
MRF 129	3	1200	20	50	1006.26	569.19	43.4	2.23	24.92	27.15
MRF 130	3	1200	20	50	1006.8	569.32	43.5	2.23	25.16	27.39
MRF 142	3	1200	20	50	1002.12	569.75	43.15	1.07	25.99	27.06
MRF 143	3	1200	20	50	1000.32	567.56	43.26	1.07	25.78	26.85
MRF 144	3	1200	20	0	1000.94	568.44	43.21	0.00	26.49	26.49
MRF 145	3	1200	0	0	1000.82	646.04	35.45	0.00	23.41	23.41
MRF 146	3	1200	0	50	1001.18	610.46	39.03	1.93	22.16	24.09
MRF 147	3	1200	0	50	1001.9	632.58	36.86	1.06	22.52	23.58
MRF 148	3	1200	0	50	1000.92	610.74	38.98	0.57	23.04	23.61
MRF 149	3	1200	5	50	1000.08	577.85	42.22	0.64	23.95	24.59
MRF 150	3	1200	5	50	2000	1448.1	27.60	0.63	25.11	25.74

Appendix B Calculation of limestone in cooler section

The mass of limestone that the given amount of energy can calcine was calculated using the equation below.

$$Q = mc\Delta T + \Delta H$$

Q=energy
m=mass
c=specific heat (0.97 which was experimentally determined)
ΔT=temperature change (assume 400°C)
ΔH=enthalpy of the reaction

Assuming a 30% uncalcined stone in the post-heat section, before the cooling section, the total mass of stones in the chamber was calculated. Using a density of 2.04 g/cm^3, which was determined through testing, and the mass calculated earlier, the volume of the chute was calculated.

$$density = \frac{mass}{volume}$$

The size of the chute was calculated using a packing factor of 7.2%. The packing factor was determined from the number of tons per lime that is in the cooling section (9tons/1960ft^3). From there, the chute's measurements were determined.

Appendix C- Calculations of Benefits for Rotary and Shaft Kilns

Rotary Kiln

Energy consumption for calcining:		4.5 MMBTU/ton
Disassociation energy		2.73 MMBTU/ton
Max potential energy reduction	4.5e^6 BTU/ton -2.73e^6 BTU/ton =	1.77 MMBTU/ton
25% reduction	1.77e^6 BTU/ton x 0.25 =	0.44 MMBTU/ton
Average fuel cost		$4.00/MMBTU
Fuel savings	0.44 e^6 BTU/ton x $4 e^{-6} BTU =	$1.76/ton
Energy/ton coal		17.6 MMBTU
CO_2/ton coal		650 lbs

Annual average US production per kiln	400,000 ton	
Fuel savings per kiln	$1.76/ton x 4e^{5}tons =	$704,000
Energy savings per kiln	0.44e^6 BTU/ton x 4e^5 tons =	176 BBTU
CO_2 reduction	176e^9 BTU /17.6e^6 BTU/ton x 650lbs/ton	6.5 MMlbs

Total annual US production	15.8 MM tons	
Total annual fuel savings	$1.76/ton x 15.8e^6 tons =	$28 MM
Total annual energy savings	0.44 e^6 BTU/ton x 15.8e^6 tons =	7 TBTU
CO_2 reduction	7e^{12}BTU /17.6e^6 BTU/ton x 650lbs/ton	259 MMlbs

Vertical Shaft Kiln

Energy consumption for calcining:		3.02 MMBTU/ton
Disassociation energy		2.73 MMBTU/ton
Max potential energy reduction	$3.02\ e^6$ BTU -2.73 e^6 BTU=	0.29 MMBTU/ton
25% reduction	$0.29\ e^6$ BTU x 0.25 =	0.07 MMBTU/ton
Average fuel cost		$4.00/MMBTU
Fuel savings	$0.07e^6$ BTU x 4e^{-6}$BTU =	$0.28/ton
Energy/ton coal burned		17.6 MMBTU
CO_2/ton coal burned		650 lbs

Annual average US production per kiln		200,000 ton
Fuel savings per kiln	$0.28/ton x 2 e^5tons =	$56,000
Energy savings per kiln	$0.07e^6$ BTU/ton x 2 e^5tons =	14 BBTU
CO_2 reduction	$14e^9$BTU /17.6e^6 BTU/ton x 650lbs/ton =	517,000 lbs

Total annual US production		4 MM tons
Total annual fuel savings	$0.28/ton x $4e^6$ tons =	$1.1 MM
Total annual energy savings	$0.07e^6$ BTU/ton x $4e^6$tons =	280 BBTU
CO_2 reduction	$280e^9$ BTU /17.6e^6 BTU/ton x 650lbs/ton =	10.3 MMlbs

Reduction of CO_2 emissions was calculated based on the reduced energy consumption for calcining. CO_2 reduction will be realized through the decreased amount of coal required for heat generation. The reduced coal consumption was calculated by dividing the energy savings by the amount of energy released from burning 1 short ton of coal (17.6 Mil BTU). The tons of coal saved per year were multiplied by the amount of CO_2 released per ton coal (650 lbs/ton coal[21], to yield total CO_2 saved per year.

Appendix D- Microwave equipment costing

Equipment	cost	quantity	total
100kW 915MHz MW generator	$135,000	4	$540,000
Y splitter	$3,300	4	$13,200
E-plane bend	$435	16	$6,960
H-plane bend	$435	16	$6,960
straight (<2ft)	$285	4	$1,140
straight additional ft	$65	32	$2,080
total			$570,340