# ULTRASONIC MONITORING OF REACTION BONDING SILICON NITRIDE

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## INTRODUCTION

A method is discussed to use ultrasonic techniques to monitor the reaction bonding of silicon nitride. Reaction bonding takes place in a nitrogen atmosphere heated up to 1390°C. As with many sensors used in hostile environments, it is difficult to design the ultrasonic sensor in a way that provides optimal clarity of the signal. The sensing system has to be designed within physical limitations on access to the furnace and it has to satisfy considerations on the design of a cooling system for the ultrasonic transducer. These limiting factors have been overcome so that ultrasonic signals have been obtained during processing, albeit with a significant noise level. Signal processing techniques have been developed which make it possible to obtain information on changes in phase velocity and attenuation during reaction bonding. The signal processing techniques have the potential to be implemented in real time for the monitoring of the progress of the reaction. This information can then be used for process control feedback.

## BACKGROUND AND MOTIVATION

Interest in reaction bonding of silicon nitride is motivated by the useful characteristics of silicon nitride and attraction to the reaction bonding process as a way of producing complex parts efficiently. Silicon nitride shows excellent potential as an engineering ceramic in high temperature applications since it has a low coefficient of thermal expansion and high thermal shock resistance. The reaction bonding process is also ideal for the production of complex parts since the process produces a near net shape part that requires only finish machining. The reaction bonding of silicon nitride takes place through an exothermic reaction between silicon and nitrogen. Reaction bonding takes place at 1150-1400 °C which is near the melting point of silicon. The reaction rate is a function of temperature and the availability or concentration of nitrogen.

In spite of the obvious advantages of using silicon nitride, production problems have limited its commercial use. The difficulty of producing high quality parts with consistency has been one important stumbling block. Production variability in the material preform due to differences in density and impurities in the powder affects the reaction bonding to such a degree that it is unrealistic to expect that a fixed set of processing parameters can be developed. Since the reaction is exothermic and occurs near the melting point of silicon, thermal runaway may occur resulting in melted silicon in the interior of the part. An excessively fast reaction rate on the exterior of the part can also prematurely close off the pore network which is needed for diffusion of nitrogen to the interior so that the reaction bonding can occur in the interior of the part. Areas in the finished product in which unreacted silicon or melted silicon is present, result in a significant degradation in the strength of the finished part. On the other hand, insufficient processing temperature can slow or stop the reaction. Reaction bonding of the material takes at least ten hours at quite high temperatures so an increase in processing time can be expensive and would have a significant impact on the cost of the final part.

The variability encountered in the reaction bonding of silicon nitride can be reduced if a method is available to directly measure the rate at which the reaction bonding is taking place in the material. Using direct measurement of the reaction, the optimum reaction rate can be determined for each part being produced. The optimum reaction rate depends on the particular part being produced since section thickness would be a significant limiting factor. Variation in the material or other process variables could be compensated for by corrections to the processing temperature. The resulting process control would help to guarantee that high quality parts are being produced.

Related research has been done in the use of long buffer rods to transmit ultrasonic signals as a means of making measurements in a hostile environment. In-situ ultrasonic monitoring of sintering was done by Gieske and Frost [1]. This work was with ZnO and superconducting ceramic materials. In the present work, extensions are made in the signal processing to improve the accuracy and increase the understanding of the measurements. In particular the emphasis here is on measurement of the phase velocity in a very dispersive system. Extensive work on the use of acoustic waveguides which serve as buffer rods has been done by Jen and co-authors [2, 3, 4, 5]. Some of this work has been focused on the reduction of multiple mode excitation which is solved here by using a different approach. Some earlier work on less attenuative high temperature materials was done by Papadakis [6]. It is only recently that a set of tools has become available in the field of digital signal processing that make it possible to solve the problems encountered in practical applications. By applying some of these tools, the measurements can be made more easily and with greater accuracy.


PHYSICAL SYSTEM AND PROCESSING

Reaction bonding of the material is carried out using conventional processing techniques. The preform used is a cylinder approximately 3 cm. long and 1 cm. in diameter which has been produced in an isostatic press from commercial silicon powder of nominal 10 $\mu$m diameter. The furnace uses flowing forming gas (90% $N_2$ 10% $H_2$) held slightly above atmospheric pressure for the reaction. Forming gas is used rather than pure nitrogen to increase the percentage of material reacting and to encourage the reaction of

SiO [7]. The reaction is begun during a two hour hold at 1250°C before being brought up to 1350°C for ten hours. Final processing occurs during a one hour hold at 1400°C, very near the melting point of silicon.

The ultrasonic monitoring system consists of 1 MHz. transducers in contact with a buffer rod which transmits the ultrasonic energy from the transducers on the exterior of the furnace to the sample reacting in the high temperature forming gas atmosphere. Six signals are obtained in both pulse-echo and through-transmission modes at each data point during reaction bonding to allow calibration of the signal. Both the change in attenuation and the change in phase velocity of the silicon nitride sample during reaction bonding are measured. Excitation of the ultrasonic transducers is done using a high energy square wave pulser. Coupling of the transducer to the buffer rod is done using a commercial high temperature couplant. The buffer rod to sample interface uses dry coupling. In pulse-echo and through-transmission configurations pre-amplifiers with gains of 34dB and 54dB were used respectively.

Fused quartz buffer rods are used resulting in some compromise in performance but with the advantage of keeping the cost of the probe assembly low. The use of a commercial high temperature transducer necessitated a water cooling system since the transducers are limited to use below 250°C. The cooling system however was sized to keep the transducers well below this temperature since efficiency is degraded at higher temperatures. Motion of the buffer rods is also necessary since acquisition of a full set of reference signals for calibration requires that the buffer rod be withdrawn from the sample to obtain a reflection from the free end of the probe each time that data is acquired during reaction bonding. Because of physical design constraints on the system, the buffer rods cannot be optimized for transmission of the ultrasonic signal. Design of these waveguides for simplicity of the ultrasonic signal would dictate either a larger or a smaller diameter than the buffer rods which were used. Buffer rods with a diameter of less than one-fifth of a wavelength would have simplified the processing of the signals because only one mode excited in the waveguide would have a significant amplitude. However it was found that insufficient energy was transmitted through small diameter buffer rods to penetrate the material. Multiple modes would have been excited in buffer rods which had a diameter
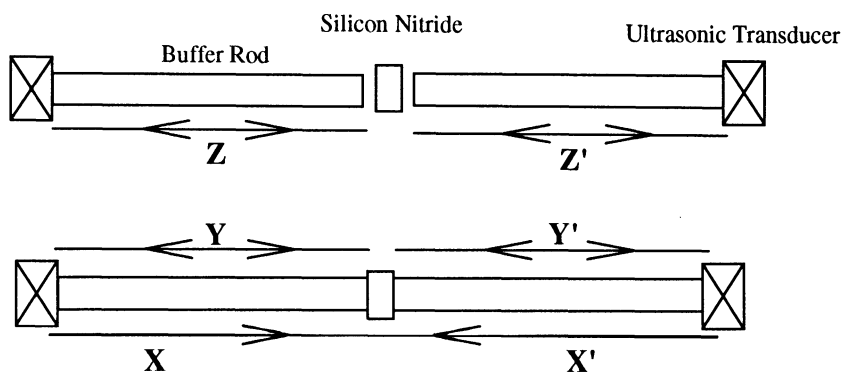


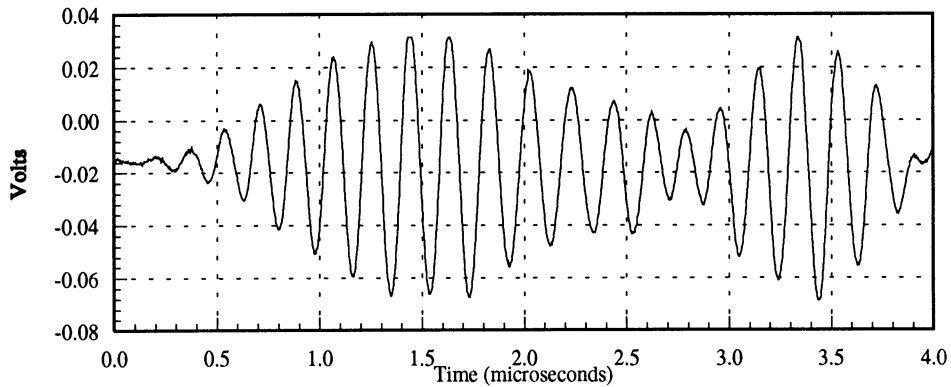Figure 1. Six signals acquired from test system.

Figure 2. Through transmission signal, unreacted material at 1350° C.

greater than those employed (a diameter greater than three wavelengths). However these signals would have a larger separation in time which would have allowed a single mode to be simply windowed out in the time domain. The larger diameter buffer rods would not fit into the small tube furnace used in this work and would in general be a significant thermal load on even a large furnace. The same constraints are seen in the work of other researchers in this area, [1, 2, 5] so a solution to this problem which is presented here is thought to be of general interest.

A through-transmission ultrasonic signal from early in the reaction bonding cycle is shown in figure 2. The signal shown was taken after the sample had been at reaction temperature for 30 minutes. There is approximately 85 dB. of attenuation as a result of the two dry coupling interfaces and from attenuation in the sample. The resulting ultrasonic signal has a significant noise level. The transmitted signal is also quite complicated. The complexity of the signal is due primarily to the multiple and overlapping modes excited in the buffer rods and dispersion in the silicon nitride sample and in the buffer rods. The resulting signal however is of sufficient quality to allow further signal processing to be performed. Two different types of signal processing are considered. The first approach makes use of homomorphic deconvolution to separate modes and then uses a pseudo-Wiener type of deconvolution to obtain the response of the material. The second method makes use of wavelet processing to provide information for additional signal processing.

SIGNAL PROCESSING

In the first signal processing method used, the first step in processing of both the through-transmission and pulse-echo ultrasonic signals is the separation of the multiple modes that are excited in the buffer rods. This process is made more complex by strong attenuation of the higher frequencies in the silicon nitride sample. As a result of this attenuation the spectrum of the through transmission signal is not a known quantity. The classic problem of separation of multiple echoes of an unknown spectrum makes the problem suitable for homomorphic deconvolution (windowing of the cepstrum) of the signal. A particularly difficult aspect of the calculation of the complex cepstrum is the unwrapping of the phase from its principal value. An adaptive algorithm is utilized that makes this step reliable for the calculation of the complex cepstrum [8].

Separation of the modes is then followed by deconvolution of the six signals obtained. The deconvolution results in the response of the material being separated from

other effects that occur during reaction bonding of the material. Examples of the effects that are accounted for is variation in coupling between the buffer rods and the transducer, variation in coupling between the buffer rod and the silicon nitride sample and changes in the response characteristics of the transducer as a result of heating. The deconvolution problem results in the form [11]:

$$Hs(\omega) = \sqrt{\frac{X(\omega)X'(\omega)/Z(\omega)Z'(\omega)}{\left[1-|Y(\omega)/Z(\omega)|^2\right]\left[1-|Y'(\omega)/Z'(\omega)|^2\right]}}$$

(1)

where the X′ designates a signal obtained from the opposite end of the fixture from X (similarly for Z and Z' or Y and Y'). The signals are shown in figure 1. $H_S$ is the response of the ceramic. The amplitude of $H_S$ yields the attenuation by the silicon sample and the phase of $H_S$ yields the corresponding phase velocity. Because of poor signal to noise ratio in combination with the instability of the deconvolution process, the deconvolution is implemented as a pseudo-Wiener deconvolution using a constant in the denominator to avoid the numerical instability that occurs in the normal implementation of deconvolution.

This technique is successful in calculating the amplitude of the deconvolved spectrum. Phase velocity calculations however are complicated by the need to unwrap the principal valued phase after deconvolution. Problems are encountered in spite of using the adaptive phase unwrapping algorithm referenced above. The difficulty is thought to be a result of numerical instabilities in the processing and should be investigated further. Results obtained for the change in attenuation of the silicon nitride using homomorphic deconvolution to separate the modes in the buffer rods are shown in figure 3. A clear trend is seen with the amplitude increasing as reaction bonding progresses. The use of the amplitude of the signal for monitoring the reaction shows promise. However, simpler signal processing would be desirable. Use of phase velocity rather than amplitude may also be preferable since many undesirable factors which influence the amplitude have a smaller effect on the phase velocity.

The second approach to the signal processing of the data obtained during reaction bonding involves use of wavelet decomposition for the identification of the modes and to assist in further processing of the signals. The first step in this processing is the creation of the time-frequency map, commonly called a spectrogram or a running spectra. For this purpose the most common wavelet function, the Morlet wavelet, is employed. The wavelet $\Psi_{a,b}$ is defined as:

$$\Psi_{a,b} = \left(\sqrt{a}\right)^{-1} \Psi[(t-b)/a], \quad \Psi = e^{t^2/2}e^{i\omega t}$$

(2)

The Morlet wavelet is simply a complex exponential windowed by a gaussian function. The gaussian windowed wavelet is chosen since it meets the bound of the time-bandwidth inequality as an equality. The time-bandwidth inequality represents the tradeoff that must be made between resolution in time and resolution in frequency. Since at this point the change in frequency content as a function of time is being investigated, satisfying the uncertainty equation as an equality represents the best compromise that can be made. Wavelet decomposition consists of finding the inner product of the wavelet function at different delay times and frequencies. No discussion of the concept of wavelets will be introduced here since many excellent references exist, see e.g. [9]. It is sufficient for present purposes to note that no fast algorithm has been used, just a direct implementation
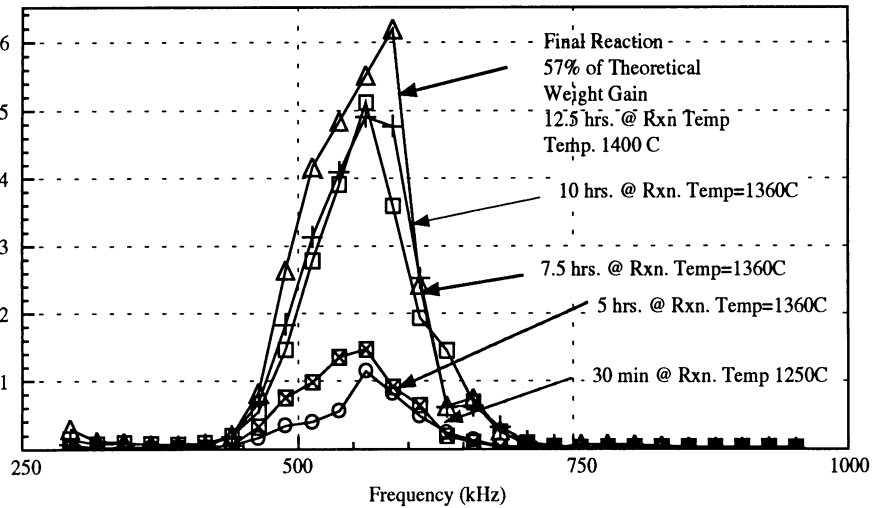
Figure 3. Amplitude of the spectrum of deconvolved signals, modes separated using homomorphic deconvolution.

of the definition and that redundant basis functions are employed since no further processing of the decomposed signal is performed. The time-frequency map (or spectrogram) is developed from the magnitude of the inner product. Figure 4 show the running spectra for two of the through transmission signals acquired during reaction bonding of the material. Neither of these maps have had the influence of other changes in the apparatus removed through deconvolution so only qualitative observations are made. In the spectrogram the axes represent time and frequency respectively while the gray scale represents amplitude of the signal at a particular time and frequency, darker color representing greater amplitude. The first image shown is a through-transmission signal taken after the material has been at reaction temperature for thirty minutes. Both low
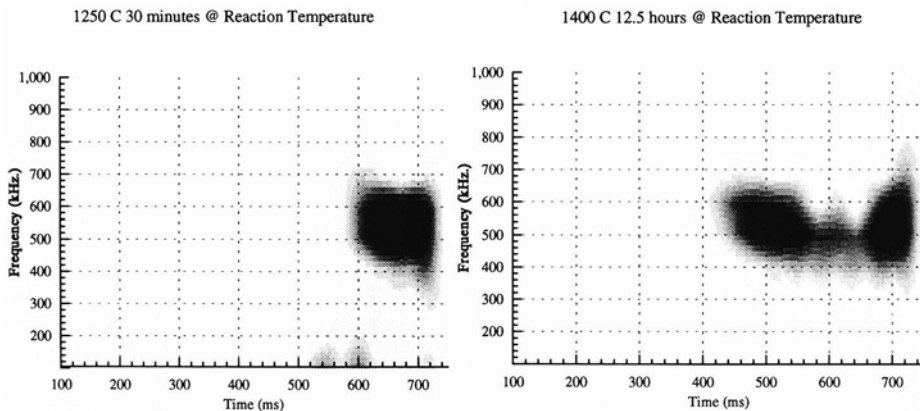


Figure 4. Wavelet spectrograms for through transmission ultrasonic signals.

frequency (100 kHz.) and higher frequency (600 kHz.) signals are evident with the low frequency signal arriving at an earlier time. Later in the reaction bonding, after having been at the reaction temperature for 12.5 hours, the low frequency signal has either disappeared or been lost in the increasing amplitude of the higher frequencies. The most important observation to be made from the running spectra is the obvious increase in velocity during the processing time considered. The emergence of the gray area further to the left of the image corresponds to an increase in the velocity. Some amplitude in the frequency range approaching 1 MHz. is beginning to appear in the second map. It is interesting to note looking at the running spectra, that the center frequency of the transducers being used is 1 MHz. However in looking at the signal map it is clear that low frequencies arrive earlier and have greater amplitude, particularly early in the reaction bonding. From the information in these images it is possible to choose the center frequency of a wavelet to use for further processing of the signals. Further work will consider alternative methods of using the running spectra to obtain as complete information from the signal as possible.

From the running spectra a wavelet center frequency of 550 kHz. is chosen for further processing of the ultrasonic signal. An approximation to the phase velocity and attenuation of the material may be made by using the amplitudes and arrival times from the corresponding peaks of the wavelet domain signals in the deconvolution formula. In this work this method is applied to data from one wavelet center frequency. Since minimization of processing time is important to process monitoring, the simplest processing scheme is employed here. To increase the accuracy for other applications different approaches may be taken. The amplitude of the signal may be more accurately approximated by a summation of the wavelet domain amplitudes up to a designated time point in the wavelet transformed signals. A complete exploration of frequency effects would include all of the frequencies with significant energy. Increased accuracy in the identification of the peak amplitude arrival time can be obtained by fitting a curve to the discrete time points. Since the wavelet is narrow band and the spectrum is known, the shape of the curve between points is known with a high degree of accuracy.
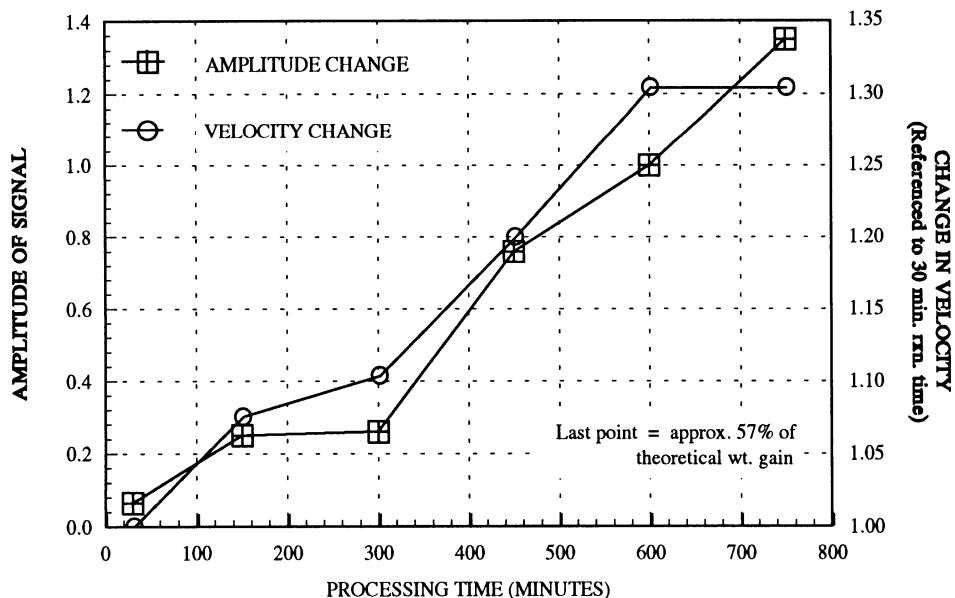


Figure 5. Velocity and amplitude change, wavelet processing.

Figure 5 shows the change in amplitude and velocity of the ultrasonic signal during reaction bonding as described above. The values are plotted on a relative scale. The first values are not taken with the material in a fully unreacted state since the preform is quite brittle. Measurement at high temperatures is difficult until sufficient strength is acquired from partial reaction of the material. The 30% increase in velocity seen in the material is very close to the 34% increase observed during ex-situ testing in [10]. Part of the discrepancy can be attributed to the use of base data for completely unreacted material in the referenced work rather than the partially reacted material used for calibration here.

CONCLUSIONS

It is clear from the results for the velocity and the attenuation of the silicon nitride that a measurable change may be observed during reaction bonding. The change, at least in amplitude, is evident regardless of which method is used to process the signal. Use of the information for process monitoring is in part constrained by the complexity of the signal processing used. Streamlining of the signal processing and implementation of a standardized procedure such as that used in the wavelet processing produce results for a real time monitoring system. It should also be noted that the time limitations on computing attenuation or velocity of the ultrasonic signal for real time control is much less stringent since the reaction bonding takes place on a time scale on the order of ten hours. Simplified processing required for the feedback system could then be implemented using a low cost personal computer. Given the interest in this material for use in large scale production, further development and production hardening of this technique could help speed progress in industrial application of reaction bonding silicon nitride.

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