Proceedings of the Institute of Acoustics

ULTRASONIC MONITORING OF SLIP-CAST CERAMICS(1)

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1. INTRODUCTION

Slip-casting is a process whereby a ceramic component is formed by water absorption in a mould. The ceramic material is present ususally as a suspension in water (the slip), which may also contain other additives to prevent flocculation and to aid in removal of the compacted material from the mould.

The process of slip-casting has been studied extensively [1, 2] and it is well-established that to a first approximation, the rate of increase of compact ceramic layer thickness during the casting process is given by

$$x^2 = At \qquad (1)$$

where x is the cast wall thickness and A is a constant which depends upon the slip and mould properties. The above is a parabolic relationship, where the square of the wall thickness is proportional to the casting time t. Note that this equation assumes that certain physical conditions, such as the pressure drop across the cast wall and the nature of the slip, remain constant. In practice, deviations may occur, for instance if the mould becomes saturated with water [3]. In addition, the constant A can vary with repeated casts into a single mould and varies between different moulds. In view of the above, it would be highly desirable to have a method for monitoring the thickness and other properties (such as density) of the compacted ceramic layer *during* slip-casting. This is particularly the case since research has shown [4] that the density may be greater closer to the mould wall. Previous work has shown that γ-ray absorption may be used to estimate the thickness of deposited ceramic layers [5]. Here, a more simple and accurate approach is described, using ultrasonic pulses. It will be demonstrated that various physical properties can be monitored in a range of ceramic materials, including alumina (Al_2O_3) and silicon nitride (Si_3N_4).

2. APPARATUS

A schematic diagram of the apparatus used in experiments for the slip-casting of alumina ceramic is shown in Fig. 1. A Parametrics 5055PR pulser/receiver was used to excite a 5 MHz immersion transducer, suspended in the ceramic slip of A-16 Alcoa alumina. This was being cast in a cylindrical plaster of Paris mould, of inside diameter 100 mm and 5 mm mould thickness. Ultrasonic pulses propagated through the slip and echoes were returned to the transducer from both the slip-ceramic and ceramic-mould interfaces. These were then received by the same transducer, in a "pulse-echo" mode and digitized by the Biomation 8100 digitizer at preset time intervals after the initiation of casting. The timing was controlled by the IBM PS/2 Model 30 microcomputer, into which ultrasonic data was transferred for analysis and storage. The result was a series of ultrasonic waveforms, captured at preset intervals in t.

In the case of silicon nitride, slips were prepared from Ube powder with a 5% Y_2O_3 additive. This mixture is used industrially and contains a reproducible oxygen impurity content, with the Y_2O_3 being an additive to aid in the subsequent sintering process. The kinetics of the slip-casting process are known to be a function of the pH of the slip and for the Si_3N_4 experiments the pH was carefully controlled over the range 8.3 to 9.3 using ammonia solution. The same 5 MHz transducer was used to collect data in these experiments, but the mould was now a single slab of plaster of Paris, with the side-walls being of perspex to result in a one-dimensional cast. In addition, a Tektronix 2430A digitial oscilloscope was used in place of the Biomation 6100 transient recorder.

Typical ultrasonic waveforms obtained in an experiment with alumina are presented in Fig. 2. Here, the signal received at the transducer by direct reflection from the slip-ceramic boundary (labelled C) shifted significantly in time, as x increased and hence as the interface moved towards the stationary transducer. The larger echo from the ceramic-mould interface (W) also moved by a small amount, due to the increased thickness of the ceramic layer, as the sound velocity in the layer was greater than that in the slip. A small third signal, R, was due to multiple reflection within the ceramic layer.

By measuring the times of arrival of reflections from the two interfaces (t_1 and t_2, corresponding to propagation times of signals C and W in Fig. 2, respectively), it is possible to estimate the cast thickness x as a function of casting time t. If d is the distance of the transducer face from the mould wall, then t is given by

$$t_1 = \frac{2(d - x)}{v_s} \qquad (2)$$

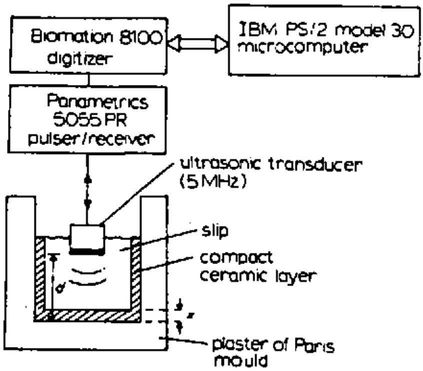

Fig. 1 Schematic diagram of apparatus for monitoring the slip-casting of alumina.

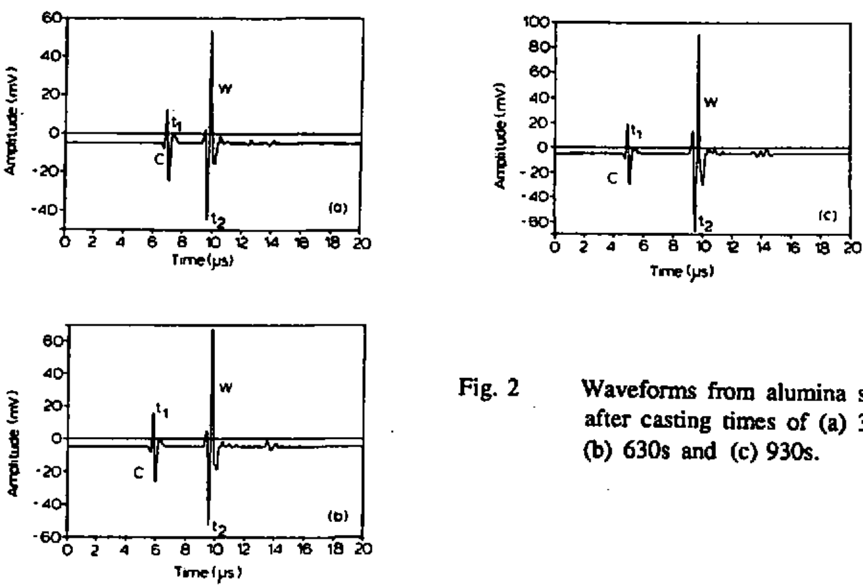

Fig. 2 Waveforms from alumina slips after casting times of (a) 330s, (b) 630s and (c) 930s.

where v_s is the acoustic velocity in the slip.

In addition,

$$t_2 = 2\left[\frac{(d-x)}{v_s} + \frac{x}{v_c}\right] \qquad (3)$$

and

$$t_2 - t_1 = \frac{2x}{v_c} \qquad (4)$$

where v_c is the acoustic velocity in the ceramic layer.

Knowledge of d and prior measurement of v_s, allows x to be monitored at discrete times t into the casting process, using Equation 2. Equations 3 or 4 could then be used to estimate the acoustic velocity in the compact ceramic layer, if desired. The above has been used to monitor the casting of both alumina and silicon nitride slips, as will now be described.

3. CASTING OF ALUMINA SLIPS

A series of experiments have been conducted using the apparatus of Fig. 1. In the first, which resulted in the waveforms of Fig. 2, the thickness x was monitored as a function of time, with successive waveforms being recorded at time intervals of 20 sec after the initiation of casting (up to a total cast time of 20 min). Fig. 3a shows the results obtained, where it is observed that the rate of thickness increase is greater at the start of the cast, as is common practice. If a parabolic increase in x was present, then a linear relationship
would exist between x^2 and t. The values of x^2 derived from the experimental data are plotted against t in Fig. 3b, where it is evident that the expected linear relationship was observed.

It is interesting to note that if the parabolic relationship of Equation 1 is assumed, the unknown constant A may be found from the slop of a graph such as Fig. 3b. This is important, in that as the experimental measurements lead to a linear relationship, the thickness x at any future time may be predicted once A is determined. Hence, a few measurements soon after casting has commenced would allow A to be estimated, from

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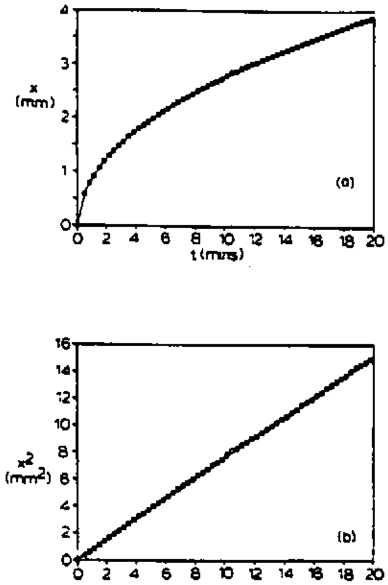

Fig. 3 Dependence of cast alumina thickness against time, showing (a) thickness and (b) thickness squared.

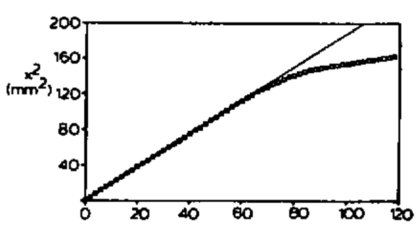

Fig. 4 The square of thickness against time for a long-term cast of alumina, showing deviation from the parabolic law of Equation (1).

which the whole process may then extrapolated. It would thus not be necessary in this case to monitor continuously, in that the rate of the process is determined by the constant A. For a given slip, *A* is determined by the condition of the mould and hence the ultrasonic method may be used to monitor mould degradation, i.e. to measure changes in *A* with repeated casts from the same mould.

As stated earlier, *A* is also affected by other physical parameters, one of which being the decrease in suction caused by liquid saturation of the mould. This is easily monitored using the present technique. As an example, a similar mould as previously was used to cast a clyindrical crucible from A-16 alumina slip, up to a cast time of 120 min. The data obtained, plotted as x^2 against t, is shown in Fig. 4. Note that the linear relationship, in agreement with Equation 1, was obeyed for 60 min of casting time. Thereafter, deviations occurred, indicating that the suction power of the mould had decreased significantly.

The above indicates that in the case of alumina, the kinetics of the casting process can be monitored successfully. The quality of the mould can also be deduced from this data, which should be of interest to the alumina manufacturing industry.

4. CASTING OF SILICON NITRIDE SLIPS

A set of experimental waveforms recorded in the presence of Si_3N_4 slip is presented in Fig. 5, after casting times of 10s, 30s and 50s. It will be noted that the waveforms contain more noise than those of Fig. 2 with lower signal levels. This was due to the greater attenuation in the slip, which was more viscous and which contained a higher solid loading of ceramic particles. However, the parabolic relationship of cast thickness with time was still observed, as the data plotted in Figs. 6(a) and (b) indicates. Note also that the casts were much faster than for alumina, again due to the different nature of the slip.

In the case of alumina, data was taken with the slip having one pH value only. For the silicon nitride, however, casts were performed at various pH values. Relationships such as that shown in Fig. 6(b) were determined for each pH, where the slope of the straight line was used to determine the value of the Constant (*A*) of Equation (1). This gave an indication of the change in casting rate with pH. The results are plotted in Fig. 7, where it is clear that an increased pH leads to a smaller value of A and hence a decrease in the casting rate.

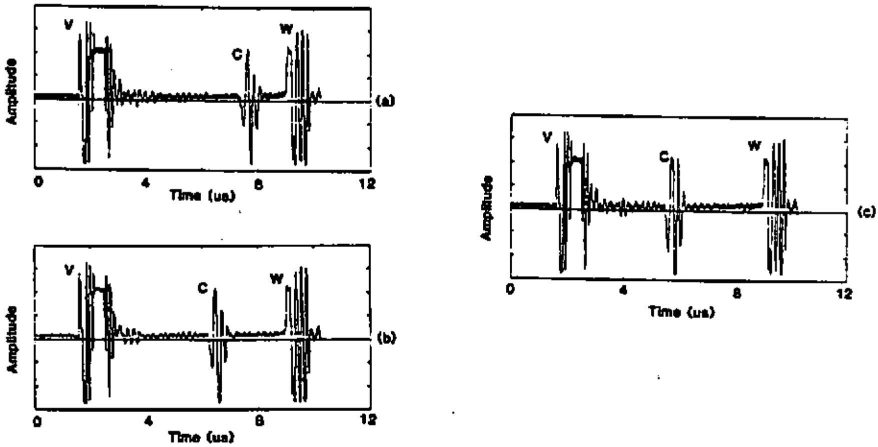

Fig. 5 Experimental waveforms obtained from silicon nitride slips, after casting times of (a) 10s, (b) 30s and (c) 50s.

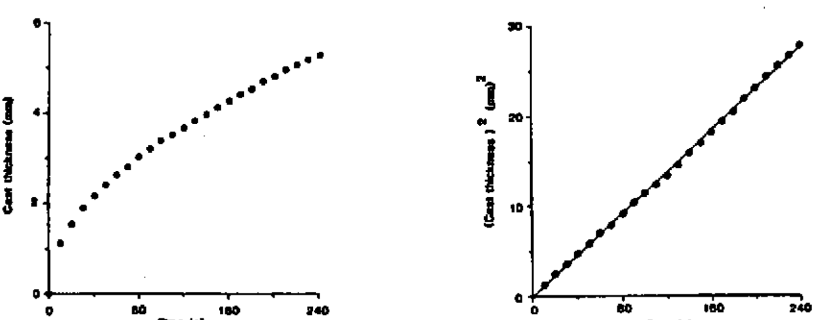

Fig. 6 Thickness and thickness squared of the cast ceramic layer as a function of time, for a silicon nitride slip of pH = 8.49.

5. DISCUSSION AND CONCLUSIONS

The waveforms presented in the above indicate that a reasonable amount of energy is reflected at the interface between the deposited ceramic layer and the slip. This, at first, might seem surprising. However, the ceramic particles used in the slip are typically of the order of a micron in size. At the interface between slip and deposited ceramic, there would be a gradient in solid particle concentration. This would occur over a distance corresponding to many particle diameters. The wavelength in the slip at the 5 MHz frequency used would be of the order of 1 mm. Hence, even if the interface were to extend over many particle diameters, it would still appear to be a sharp interface when compared to the much longer wavelength of the ultrasound.

Present research aims to establish whether this technique can be used to determine the properties of the ceramic layer as it is deposited. Preliminary results indicate that it is possible to monitor the change in acoustic velocity υ_c in this layer during casting. If this is the case, then it might be possible to establish the extent of packing density variations, which in turn lead to unwanted effects in such ceramics on firing to produce the finished product.

6. REFERENCES

[1] E R Herrman & I B Cutler, Trans. Brit. Ceram. Soc. 1, p207 (1962).
[2] D S Adcock & I C McDowall, J. Amer. Ceram. Soc. 40, p355 (1957).
[3] B W Nies & C M Lambe, Ceram. Bull 35, p319 (1956).
[4] F M Tiller & C D Tsai, J. Amer. Ceram. Soc. 69, p882 (1986).
[5] R F Deacon & S F A Miskin, Trans. Brit. Ceram. Soc. 63, p473 (1964).

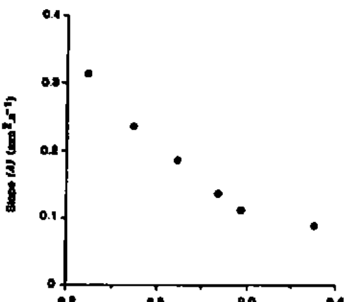

Fig. 7 Variation of the constant *A* in Equation (1) with pH of silicon nitride slips.