SOME PHYSICAL PROPERTIES OF THE TWO PHASE COMPOSITE MATERIAL: POLYMETHYL METHACRYLATE WITH METAL FILLERS

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ABSTRACT

The effect of adding copper and aluminium powders, with different volume fractions ranging from 0 to 10 %, to polymethyl methacrylate was studied. The thermal conductivity was measured using a steady state method and the experimental results were compared with a theoretical model. The effect of adding the metallic phase on the ultimate compressive strength and the Charpy impact energy was also investigated. The thermal conductivity and the Charpy impact energy showed reasonable increase with increasing the volume fraction of the metallic phase. A similar behaviour was noticed with respect to the ultimate compressive strength, but the increase was not significant.

INTRODUCTION

The polymethyl methacrylate (PMMA) is a sort of polymers. It is considered to be the most satisfactory denture base material. It is important to improve its low thermal conductivity for better appreciation of taste as well as reducing the foreign body feeling of dentures [1]. Also, it is important to increase its ultimate compressive strength and its impact energy in order to elongate its life time and protect it against easy fracture during the usual use.

Recently, studies were made to improve the physical properties of (PMMA) by adding fillers to it in different forms [2-4].

In this work, copper and aluminium in the form of fine powders were added with different volume fractions to (PMMA). The effect of the metallic phase on the thermal conductivity ,ultimate compressive strength and the impact energy was investigated.

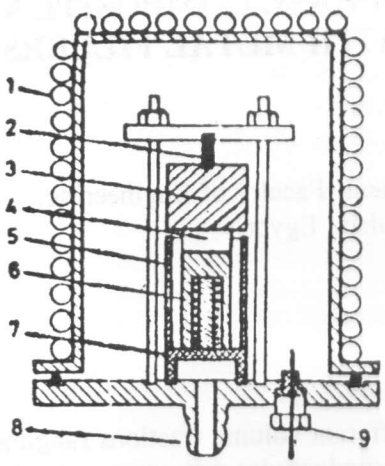
EXPERIMENTAL

Copper and aluminium in the form of a very fine powders, with purity 99.9 % from Koch-Light Laboratories Ltd England with a particle diameter of about 15 μm, were individually added in different ratios by weight to the (PMMA) powder (Resin for dentures, Superacryl, SPOFA-DENTAL, PRAHA). The mixture was mixed in a morter until a homogenous colour was observed. The obtained PMMA-metal mixture was then mixed with monomer (methyl methacrylate acid) in the ratio 2 : 1 by volume [5]. After about 20 minutes at room temperature, a plastic mass was formed which was no longer sticky and could be packed into the prepared moulds. The moulds were designed in different shapes to suit the experimental work.

The samples were kept in a water bath at a constant temperature 75 °C for 16 hours. This process is called polymerization [6]. The samples were removed from the moulds, then sanded and polished.

The percentage volume fraction of the metal phase was calculated by the use of the predetermined densities of the metal phase and the (PMMA) phase.

For the thermal conductivity measurements, the samples were made in the form of disks of 25 mm diameter and about 2 mm thickness. For the ultimate compressive strength measurements, the samples were made in the form of cubes of 10 mm side length. For the impact energy, they were made in the form of bars with a square cross section 10x10 mm and length 75 mm. The bars were notched in the middle (V-shape notch with depth 2 mm and angle 45° for each side).

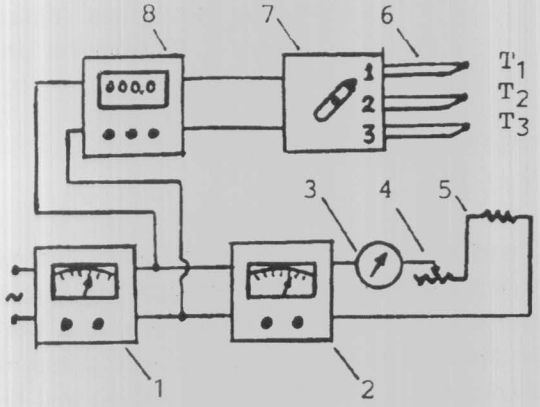


1.Water tubing. 2.Glass rod. 3.Heac sink 4.Sample. 5.Radiation shield. 6.Heater. 7.Asbestos base. 8.To vacuum system. 9.Teflon insulated electrodes.

Figure 1. A schematic diagram for the thermal condutivity apparatus.

The thermal conductivity of each sample was measured by a steady state method using an apparatus whose schematic description is shown in Figure (1). The vacuum chamber is made of copper with soldered copper tube on its outside surface for water circulation. A stainless steel base is sealed to the chamber with the aid of a rubber O-ring. The base connects the chamber to the vacuum system and to the electrical circuit shown in Figure (2) through eight teflon insulated electrodes.

The sample was mounted between the heat sink (cylindrical black copper block) and the heat source with highly polished flat surfaces. A very thin layer of silicon grease was used between the contact surfaces in order to improve thermal conduction by eliminating the small air gaps which may be found. A constant pressure was applied on the heat sink by using a clamping system. A cylinder made of highly polished stainless steel was used as a radiation shield in order to minimize the heat lost by radiation from the small lateral area of the sample. Three calibrated copper-constantan thermocouples were attached by silver soldering to the upper part of the heat source, lower part of the heat sink and the inner surface of the vacuum chamber. These thermocouples were connected through a multiterminals connecting box to a precise digital thermometer (Aoip TNC 20) with a precision $\pm$ 0.1 °C .



1.Voltage stabilizer. 2.Low voltage power supply. 3.Ammeter. 4.Variable resistance. 5.Heater. 6.Thermocouples. 7.Multi-terminals connecting box. 8.Digital thermometer.

Figure 2. The electrical circuit.

After the apparatus was assembled, it was evacuated to 10^{-3} mmHg and water was allowed to flow from a thermostat through the copper tubing surrounding the vacuum chamber at a constant rate and temperature. The nichrome heater wire was supplied with a constant current using a stabilized direct current low voltage power supply. The current was adjusted and maintained constant by the use of a variable resistance.

The approach to the steady state was checked every half an hour. The thermal equilibrium was attained after about 4 hours. The final temperatures T_1, T_2, T_3, of the bottom surface of the sample, heat sink and the surrounding vacuum chamber respectively, were recorded. Neglecting the radiation heat loss from the lateral area of the sample, the thermal conductivity (K) of the sample could be calculated from the following equation :

$$K = \frac{\sigma \varepsilon S d (T_2^4 - T_3^4)}{A(T_1 - T_2)} \qquad (1)$$

where (A) is the sample surface area, (d) is the sample

thickness, (σ) is the Stefan's Boltzmann constant, (S) is the surface area of the heat sink and (ε) is the emissivity of the heat sink surface which was determined using a sample of known thermal conductivity.

The ultimate strength and the impact energy were determined in the laboratory of materials testing in the faculty. The ultimate compressive strength was determined by a universal testing machine from the original cross sectional area of the sample and the maximum applied force shown on the machine scale. The impact energy was determined by the Charpy method using an impact testing machine where a pendulum is released to swing down to fructure the sample at the notch. The energy lost by the pendulum during the fructure of the sample can be determined directly from the machine scale[7].

RESULTS AND DISCUSSION

Figure (3) shows the measured thermal conductivity of the two systems, PMMA-Al and PMMA-Cu , versus the volume fraction of the metallic phase. It is clear that, for a certain volume fraction, the thermal conductivity of PMMA-Cu is higher than that of PMMA-Al and this was expected.

In spite of the higher thermal conductivity obtained with copper, aluminium is preferable because it has two advantages, its very little cytotoxic effect and it is an inexpensive metal. In general the physical properties of the (PMMA) with metal powder depend on the particle size and shape of the metal particles. For the same volume fraction of a certain metal with different particle shapes, the particles that overlap and form conductive pathways within (PMMA) will show higher thermal conductivity than the particles which are separated by the (PMMA) matrix. The ideal form is reported to be elongated particles of metals with a length to diameter ratio from 75 to 125 [8].

A theoretical technique to predict the thermal conductivity of a two phase solid mixtures has been developed by Cheng and Vachon [9]. They modified and extended Tsao's model [10] and predicted the effective thermal conductivity (K_E) of the two phase system as:

$$K_E^{-1}=D+\frac{1-B}{K_c} \qquad (2)$$

with:

$$D=\frac{1}{\sqrt{C\Delta K(K_c+B\Delta K)}}\ln\frac{\sqrt{K_c+B\Delta K}+\frac{B}{2}\sqrt{C\Delta K}}{\sqrt{K_c+B\Delta K}-\frac{B}{2}\sqrt{C\Delta K}} \qquad (3)$$

$$B=\sqrt{\frac{3\phi}{2}} \qquad (4)$$

$$C=4\sqrt{\frac{2\phi}{3}} \qquad (5)$$

$$\Delta K=K_d-K_c \qquad (6)$$

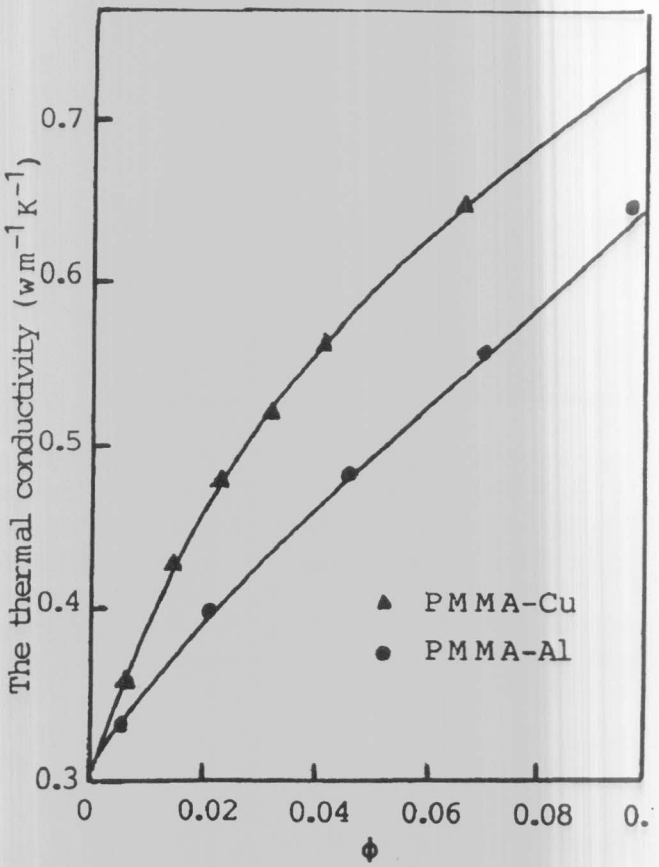


Figure 3. The thermal conductivity vs the volume fraction of the metallic phase.

were (K_c) is the thermal conductivity of the continuous phase (here, the PMMA phase), and (K_d), (ϕ) are the thermal conductivity and the volume fraction of the

discontinuous phase (here, the metallic phase) respectively.

The effective thermal conductivity for both systems PMMA-Cu and PMMA-Al was calculated according to equation (2) and plotted for comparison with the obtained experimental results in Figures (4) and (5). It is noticed that there are large differences between the measured values and the predicted theoretical values from equation (2). To explain this behaviour, the calculated thermal conductivity for PMMA-Cu, ($K_{E,Cu}$), and PMMA-Al,($K_{E,AL}$), are given in Table(1) for the volume fractions up to 0.2 .

Table 1. The calculated thermal conductivity using equations (2) and (7).

ϕ	$K_{E,Cu}$	$K_{E,Al}$	K_E
0.02	0.3734	0.3732	0.3738
0.04	0.4088	0.4085	0.4092
0.06	0.4410	0.4406	0.4415
0.08	0.4722	0.4718	0.4728
0.10	0.5037	0.5033	0.5044
0.12	0.5360	0.5355	0.5368
0.14	0.5696	0.5690	0.5705
0.16	0.6048	0.6042	0.6058
0.18	0.6421	0.6414	0.6433
0.20	0.6820	0.6811	0.6832

It is clear that there are very small differences between $K_{E,Cu}$ and $K_{E,Al}$. It can be concluded that for the small volume fractions in the case of $K_d >> K_c$, the first term (D) in equation (2) has a very small effect on the calculated value of the effective thermal conductivity and it can be approximated to :

$$K_E = \frac{K_c}{1-B} \qquad (7)$$

The effective thermal conductivity (K_E) as calculated from equation (7) is given also in Table (1) . From the given data in the table, it can be seen that the maximum difference between (K_E) when calculated from equations (2) and (7) is 0.0021 $Wm^{-1}K^{-1}$. It is concluded from the above discussion that, for the small range of volume fractions in the case of $K_d >> K_c$, equation (2) cannot distinguish between the effect of the various discontinuous phases. As an attempt to improve this behaviour, another term depending on K_d is proposed to be added to equation (7) and the following equation is proposed to calculate the effective thermal conductivity :

$$K_E = \frac{K_c}{1-B} + 0.005 n \Phi K_d \qquad (8)$$

where (n) is a factor depends on the shape of the discontinuous phase particles and is taken in the present work 1.038 for Cu and Al particles used.

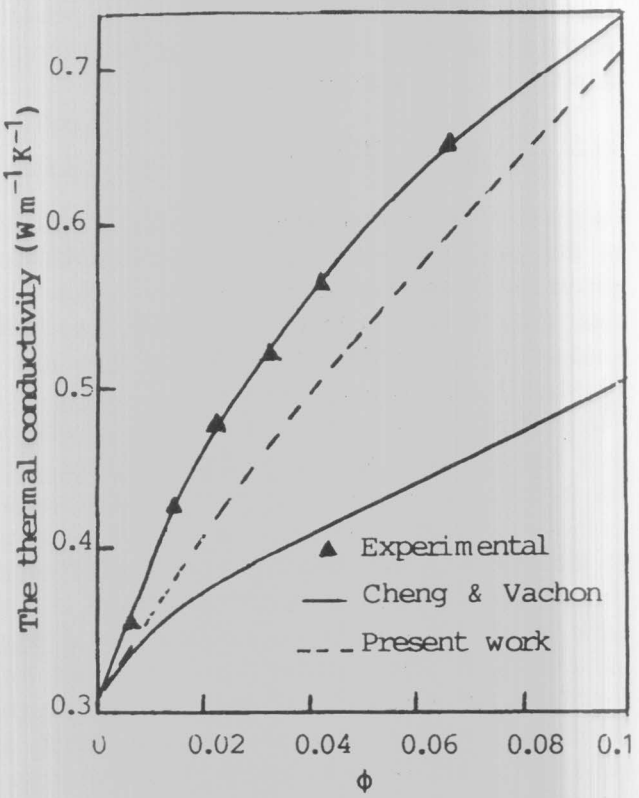


Figure 4. Experimental and theoretical thermal conductivity of PMMA-Cu vs. the volume fraction of the metallic phase.

The calculated values for both systems using equation (8) are plotted versus volume fraction and shown in Figures (4) and (5). It is clear that equation (8) is more convenient to predict the effective thermal conductivity if $K_d >> K_c$ in the small range of volume fractions only where the first term of equation (2) has a very small effect on the value of (K_E).

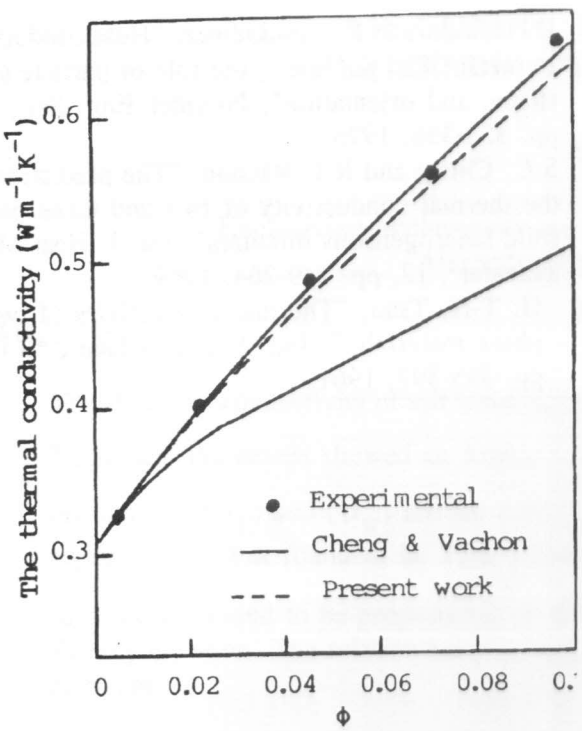


Figure 5. Experimental and theoretical thermal conductivity of PMMA-A1 vs. the volume fraction of the metallic phase.

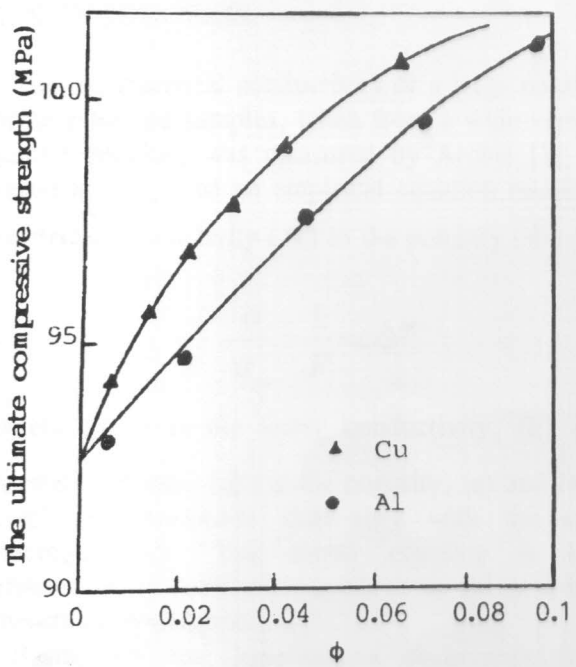


Figure 6. The ultimate compressive strength of PMMA-Cu and PMMA-A1 vs. the volume fraction of cu and A1.

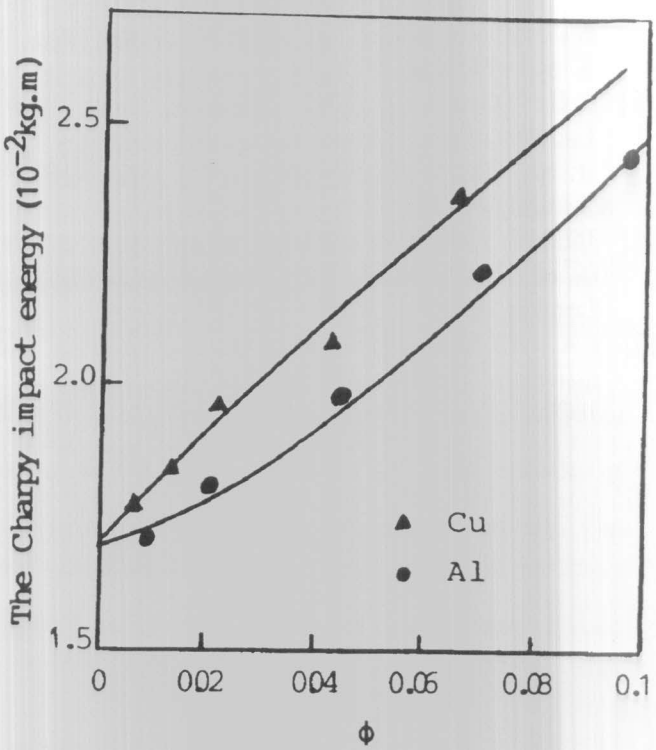


Figure 7. The charpy impact energy of PMMA-Cu and PMMA-A1 vs. the volume fraction of Cu and A1.

The obtained results for the ultimate compressive strength and the Charpy impact energy versus the volume fraction of the metallic phase are shown in Figures (6) and (7) respectively. In general, there are improvements in the two mentioned properties with increasing the volume fraction of the metallic phase. It is noticed that the addition of the metallic powder has a little effect on the compressive strength while it has a reasonable effect on the Charpy impact energy.

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