Filler Content Effects on the Ultrasonic Response of Composite Materials Using the Pulse-Echo Amplitude Technique

Asst. Prof. Awad Hallosh Khidhir
Mechanical Engineering Dept., College of Eng.
Al-Mosul University, Mosul, Iraq

Asst. Lect. Amer Yahya Al-Jarjees
Mechanical Engineering Dept., College of Eng.
Al-Mosul University, Mosul, Iraq

Abstract

The aim of this study is to investigate of the capability of ultrasonic technique in assessment of the filler content effects in polymer matrix composite and to provide additional data concerning the particulars of the use ultrasonic response, velocity and attenuation measurements for quality control and sorting composite materials. For the experiments, filled acrylic (matrix) with different weight percentages of calcium silicate and carbon black powder (fillers) individually were moulded. The pulse-echo amplitude technique and normal probes of (1, 2 & 4 MHz) was used. The directional dependence of velocity, attenuation, hardness and filler content were investigated. Also the ultrasonic measurements were carried out at various testing frequencies to observe the reliability of the technique employed.

Both filler type and filler weight (%) were systematically varied to investigate their effects on the ultrasonic response of thermoplastic acrylic (PMMA). It was observed that, in general, as the filler weight (%) increased, there is decay in the ultrasonic response provided particles dispersion and deagglomeration was maintained. Increase of filler weight (%) also accompanied by increase in the ultrasonic attenuation and reduction in ultrasonic velocity and hardness. Clear correlation between these measurements was obtained beyond (15%) filler weight percent.

It was concluded that ultrasonic technique are sensitive to the filler content in composite materials. Because the ultrasonic response, attenuation and velocity is dependent upon filler content, and can be used to predict the mechanical properties of the filled composite materials. It can be utilized also to sort composite materials with marked differences in one of the above variables. Furthermore the ultrasonic techniques eliminate the need for preparation and destructive testing of products, which can be carried out in a few minutes, and low cost.

الخلاصــــــة

الهدف من هذه الدراسة هو التحري عن قدرة تقنية الموجات فوق الصوتية في تخمين تأثيرات محتوى المادة المالئة (filler) في قاعدة (matrix) بوليميرية وإعطاء بيانات تتعلق بخصوصيات استخدام قياسات،الاستجابة فوق الصوتية (ultrasonic response)، السرعة (velocity) والتوهين (attenuation)لغرض السيطرة النوعية والتصنيف للمواد المركبة (composite materials). لغرض الاختبار تم قولبة الاكريليك (acrylic) الذي يمثل القاعدة مع نسب وزنية مختلفة من مسحوق (powder) المادة المالئة كلا على انفراد وهي سيليكات الكالسيوم (calcium silicate) واسود الكربون (carbon black). تم استخدام تقنية سعة صدى-النبضة (pulse-echo amplitude) ومجسات عمودية (normal probes) بتردد ١ ، ٢ ، ٤ ميكاهيرتز. تم التحري عن الاعتماد ألاتجاهي للسرعة، التوهين، الصلادة (Hardness) مع محتوى المادة المالئة. القياسات فوق الصوتية تم تنفيذها باستخدام ترددات مختلفة لإدراك الثقة بالتقنية المستخدمة وإمكانياتها.

تم تغيير كل من نوع المادة المالئة ونسبها المئوية الوزنية بانتظام للتحري عن تأثيرها على الاستجابة فوق الصوتية للاكرليك اللدن حراريا (Thermoplastic). تبين بشكل عام انه كلما ازدادت النسبة المئوية للمادة المالئة يكون هناك انحدار(decay) للاستجابة فوق الصوتية بشرط ضمان الانتشار لدقائق مسحوق المادة المالئة وعدم حصول تكتل لها . كذلك زيادة النسبة المئوية للمادة المالئة يصاحبه زيادة في التوهين فوق الصوتي ونقصان في السرعة والصلادة. الربط بين هذه القياسات أو المتغيرات واضح في المدى البعيد بعد الإضافة الوزنية ١٥ % من المادة المالئة في المواد المركبة التي تضمنها البحث.

تم الاستنتاج بان التقنية فوق الصوتية حساسة لمحتوى المادة المالئة في المواد المركبة. إن سبب اعتماد الاستجابة فوق الصوتية ، الاضمحلال والسرعة على محتوى المادة المالئة يقود إلى إمكانية استخدامها للتنبوء بالخصائص الميكانيكية للمواد المركبة المملؤة (المحشوة).كذلك يمكن تصنيف المواد المركبة بدرجات متفاوتة اعتمادا على احد تلك المتغيرات. التقنية المستخدمة تلغي الحاجة للتحضير والفحص اللااتلافي للمنتجات، بالإضافة إلى ذلك يمكن انجازها في دقائق قليلة وكونها اقتصادية وغير مكلفة.

1. Introduction

Composites are combinations of two materials in which one of the materials, called the reinforcing phase, is in the form of fibers, sheets, or particles, and are embedded in the other material called the matrix phase. The reinforcing material and the matrix material can be metal, ceramic, or polymer. Typically reinforcing materials are strong with low densities while the matrix is usually ductile or tough material. If the composite is designed and fabricated correctly, it combines the strength of the reinforcement with the toughness of the matrix to achieve a combination of desirable properties not available in any single conventional material [1].

The design and fabrication of composite materials optimized for a special application has been the subject of extensive research recently. The application range from mechanical structure to electronic devices. In designing composite materials, primary important is given to the proper choice of component phases and the way in which they are interconnected to maximize a predefined figure of merit for the application envisaged [2].

It is now well-established that if ultrasound can be propagated in given medium than significant information about that medium can be deciphered [3].

Ultrasonic attenuation measurements are a standard method used to assess the effect of materials degradation [4].

Ultrasonic technique is one of widely used techniques for nondestructive testing (NDT) of materials. In ultrasonic testing useful information about integrity or geometry of the object under a test is obtained. Measurement configuration often encountered in NDT includes pulse-echo reflection technique [5].

Ultrasonic response of filled polymers depends strongly on the concentration, particle size and dispersion of fillers [6].

Interrogation of a specimen by ultrasonic pulses yields information relating to material acoustic properties and specimen dimensions. Dimensions can be obtained from the signals reflected from the front and back faces of the specimen [7].

The prime aim of the presently reported work was thus to explore the effect of filler content on the ultrasonic response of composites using the pulse-echo amplitude technique.

2. Materials and Experimental Techniques

2.1 Materials

Materials and Experimental Techniques were carried out at Mosul University in the materials testing laboratory. Acrylic moulding powder Ref. No.B121 was used, which characterized by the following properties according to the manufacturer specifications:

Matrix: Acrylic (PMMA), 100% polymer, no filler, Thermoplastic.

Colour: Transparent (water – white).

Moulding temperature: 145 – 170 ^{o}C.

Moulding pressure: 150 – 250 bar.

Mount properties: shrinkage: 0.5 %; Relative density: 1.18, Softening point: 110 ^{o}C.

Fillers - Calcium silicate of particle sizes in the range 48-52 μm.

- Carbon blacks of particle size 150 μm.

For reliable screen analysis these fillers particle sizes are obtained using a mechanical sieve shaker and the recommended sieve sizes ASTE11: 70, 8 in diameter, full depth 2 in.

2.2 Experimental Techniques

2.2.1 Sample Preparation

Acrylic moulding powder (PMMA) which represent the matrix of the composites involved were filled with different weight percent (%) of two different type of filler particles. i.e. 0% (pure acrylic), 5%, 10%, 15%, 20%, 25%, 30%, 35% and 40%. The filler are carbon black and calcium silicate particles were added individually to represent the two sets of samples involved in this investigation. This gradual increase of filler content is to reveal their effect on the ultrasonic response and the capability of the ultrasonic technique in assessment of the composites constituents. The filler particles were added to the acrylic moulding powder and mechanical blending was employed to uniformly mix and disperse the particles. The material blends were moulded using automatic press shown in **Fig.(1)**. All samples were moulded using 4 bar pressure and temperature (150 – 200 ^{o}C) for 7 minute and then tap water cooling for 4 minutes according to the manufacturer moulding specification. For each filler weight percent of the two sets, 5 mm, 10 mm and 15 mm sample thickness were moulded. All samples are round of 25 mm diameter (i.e. total number of samples of the two sets are 54).

Figure (1) Automatic moulding press

All samples are grinded and polished to obtain the same surface condition and accurate thickness. Samples were photographed using optical microscope and digital camera. Five photo microstructures from each set of different filler percentage and their related ultrasonic screen trace are shown in **Figs.(2)** and **(3)**.

The filler content and its type could be identified from the reflection pattern. This identification was based on the appearance of the screen trace and their related gain setting in (dB) as shown on it. The identification also involves the measurement of the amplitude of the first signal, attenuation and the estimated average sound wave velocity.

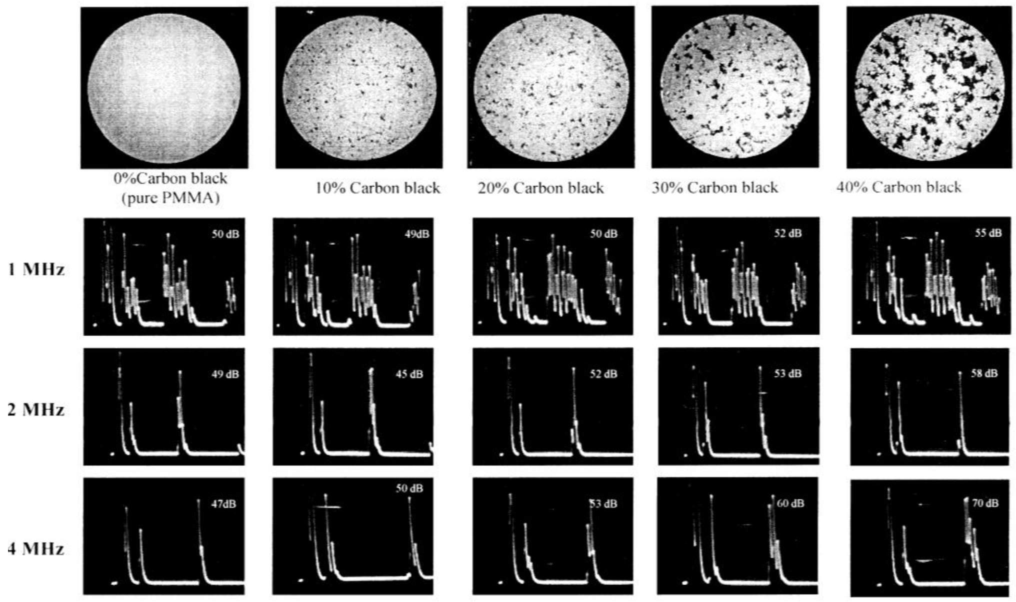

Figure (2) Microstructures of PMMA/Carbon black, selected samples (X40), of different filler weight % and their related oscilloscope screen trace, using normal probes of different frequencies

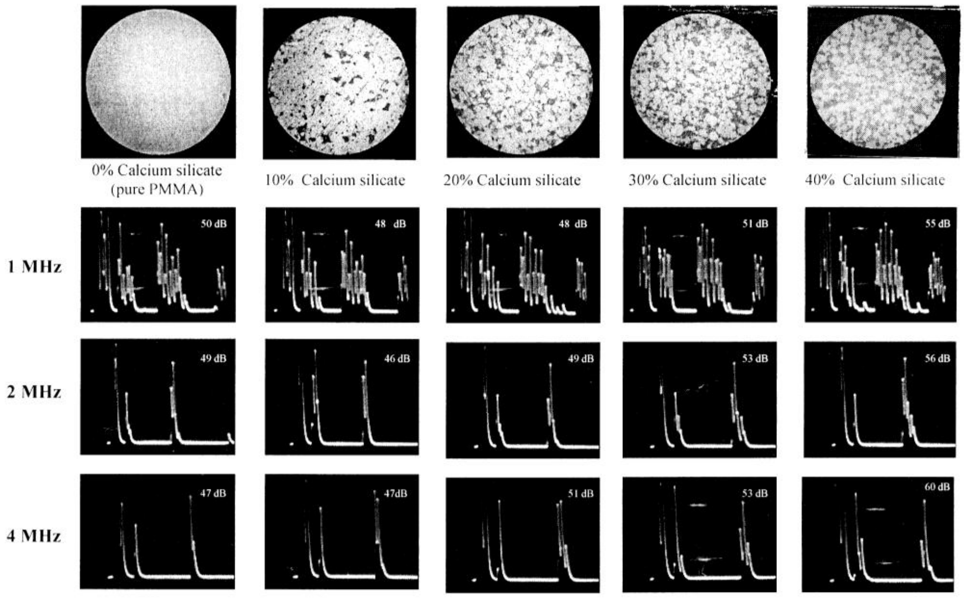

Figure (3) Microstructures of PMMA/Calcium Silicate, selected samples (X40), of different filler weight % and their related oscilloscope screen trace, using normal probes of different frequencies

2.2.2 Hardness Tests

The hardness tests were performed by hardness tester. Rockwell hardness was taken (HRR) that it was a ball of 12.7 mm diameter and load of 60 kg , which is suitable for soft thermoplastics hardness measurements. For the sake of systematic study, five identifications for hardness have been applied on each specimen and the averages of these values were calculated.

Hardness measurements are sensitive to changes in filler weight percentages especially at higher filler content.

2.2.3 Ultrasonic Measurements

Pulse-echo amplitude technique with a scan presentation also used in the measurements. The longitudinal ultrasonic waves were generated and applied by Krautkramer probs. The wave was transmitted into the specimen through oil-base couplant. The signals were amplified and displayed on the screen, type Krautkramer USM2 flaw detector as shown in **Fig.(4)**. A constant pressing force on the probe was applied to maintain a constant thickness of couplant films at the probe-specimen interface. Time-base calibration was performed using (V_1) block.

Figure (4) Krautkramer ultrasonic flaw detector USM2 type and test samples

The propagation velocity of the sound was determined using the formula given in equation (1) [8];

$$V = tV_{ref} / (KT) \quad \ldots\ldots (1)$$

where:

t: is the thickness of the specimen.

V_{ref}: is the velocity of sound in the calibration block.

K: is a measure in mm representing on scale on the time-base range of the screen.

T: is the scale value of the peak from back wall. Measurements were repeated three times on each surface and then, the mean value of the velocity was calculated.

Attenuation measurements were performed in accordance with ASTM E214 and ASTM E664. The first back wall echo was considered as the reference peak and its amplitude was adjusted to a constant value (80% FSH), to allow other peaks to be compared with. Three independent measurements were done on each surface and the average value was calculated. The apparent attenuation is calculated according to equation (2) [8];

$$\text{Apparent Attenuation} = \frac{20\log_{10}\dfrac{A_m}{A_n}}{2(n-m)t} \quad \ldots\ldots (2)$$

where: (A_m) and (A_n) are the amplitude of the (m^{th}) and (n^{th}) back reflections in decibel (dB) where **n** > **m** and (t) is the thickness of the specimen. In this study, (m) is chosen as first back wall echo height and (n) is chosen as second back wall echo height.

The echo amplitude in (dB) on the oscilloscope screen traces was shown in **Fig.(2)** and **(3)** and it represent the number of (dB) required to bring the first back wall echo to (80%) Full Screen Height (FSH).

3. Results and Discussion

Carbon black filler has been used in both thermoplastic and thermosetting polymers, to increase its heat-resistance, dimensional stability, stiffness, electrical and thermal conductivity and processability. Calcium silicate has been used in thermosetting polymer, to increase heat resistance dimensional stability, stiffness and hardness [1].

The first series of test was carried out on these two sets of samples, using normal probes of 1, 2 & 4 MHz to establish the effectiveness of the technique employed in studying the effect of the filler content on the ultrasonic echo amplitude response. For this establishment purpose primary sample thickness of 10 mm was considered.

The test results of five selected samples from each set are presented in **Figs.(2)** and **(3)**. **Figure (5)** demonstrates the capability of the technique employed to differentiate between the ultrasonic echo amplitude response of these composites and in assessments their filler contents effect especially beyond the (15%) filler content zone. The microstructures show an increase in the agglomeration of fillers as their weight % increased. The superiority of the 2 MHz probe over the 1 MHz and 4 MHz was demonstrated during the test. On using the 1 MHz probe signal envelopes was obtained and that could be related to its wider beam spread, low resolution and less sensitivity to the composites texture. The 2 MHz enables to improve back wall echo (BWE) reflection in composites. In using 1 MHz probe filler particles produces broadband noise of large amplitude of BWE reflection which masks useful signals. Although the 4 MHz probe has the highest sensitivity among the probe used to the composite texture it shows the lowest ultrasonic echo amplitude response and that possibly related to its higher attenuation characteristics among the probes used. Krautkramer [9] has shown that higher frequency results higher ultrasonic attenuation.

The unclarity of the main back wall reflected signal (low signal to noise ratio) and the bad resolution observed, when using the 1 MHz probe make it unsuitable for test measurements.

Further measurements was carried out on another two sets of filled PMMA of the same filler types and of the same filler weight % but the sample thickness increased to 15 mm. This increase in sample thickness shows the thickness factor effect on the ultrasonic response, as accumulated effect of the filler content on the ultrasonic response.

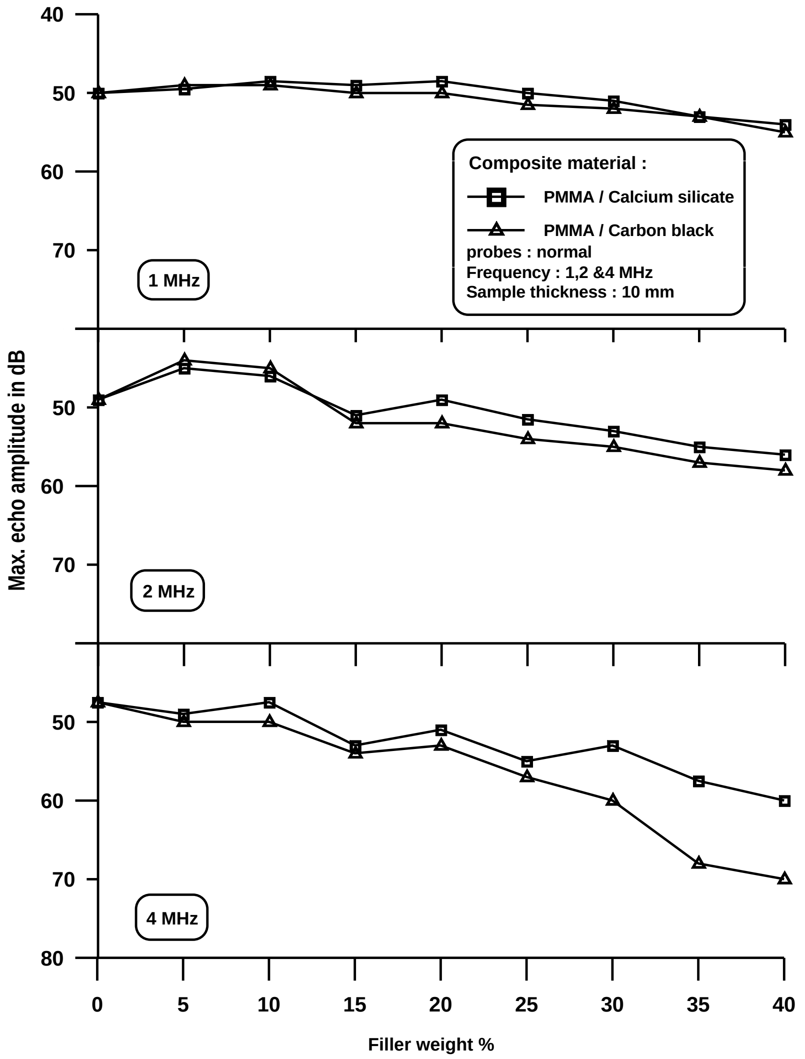

Figure (5) The effect of filler weight % in moulded acrylic (PMMA) on the ultrasonic response using normal probes of 1,2 and 4 MHz frequency at temperature 25 °C

The results shown in **Fig.(6)** confirm the results shown in **Fig.(5)**, that the filled acrylic PMMA with calcium silicate shows higher ultrasonic response than PMMA with carbon black. The lower ultrasonic response when using carbon black filler could be related to the filler particles size factor. The accumulation of the filler effect as a result of increase in thickness, results in a wider distribution of the ultrasonic response. **Figure (6)** also shows that probe 4 MHz with full attenuator capacity (80 dB), can not bring the first signal to the same reference (80% FSH). This possibly related to the higher attenuation and the severity of energy loss when acrylic filled with carbon black of the larger particle size (150 µm).

Figure (7) confirmed this fact and shows also that it is impossible to obtain two consecutive back wall echoes in order to calculate the ultrasonic attenuation for the 40% carbon black sample in spite of using smaller thickness of 5 mm for attenuation measurements purposes. The higher attenuation could be related to higher scattering of the wave on carbon black filler particles when using 4 MHz probe of the lowest wavelength among the involved probes.

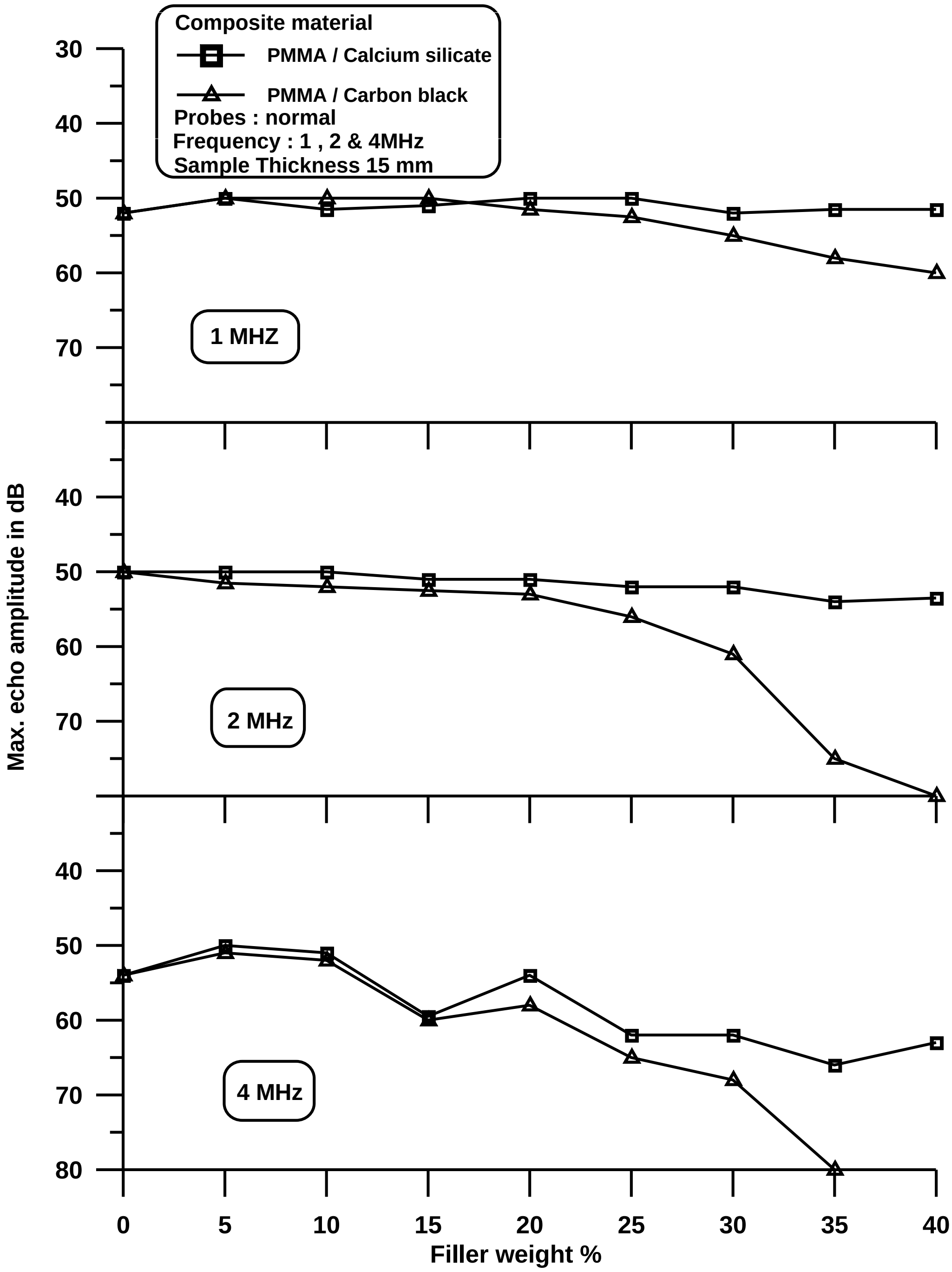

Figure (6) The effect of filler weight % in moulded acrylic (PMMA) on the ultrasonic response using normal probes of 1,2 and 4 MHz frequency at temperature 25 °C

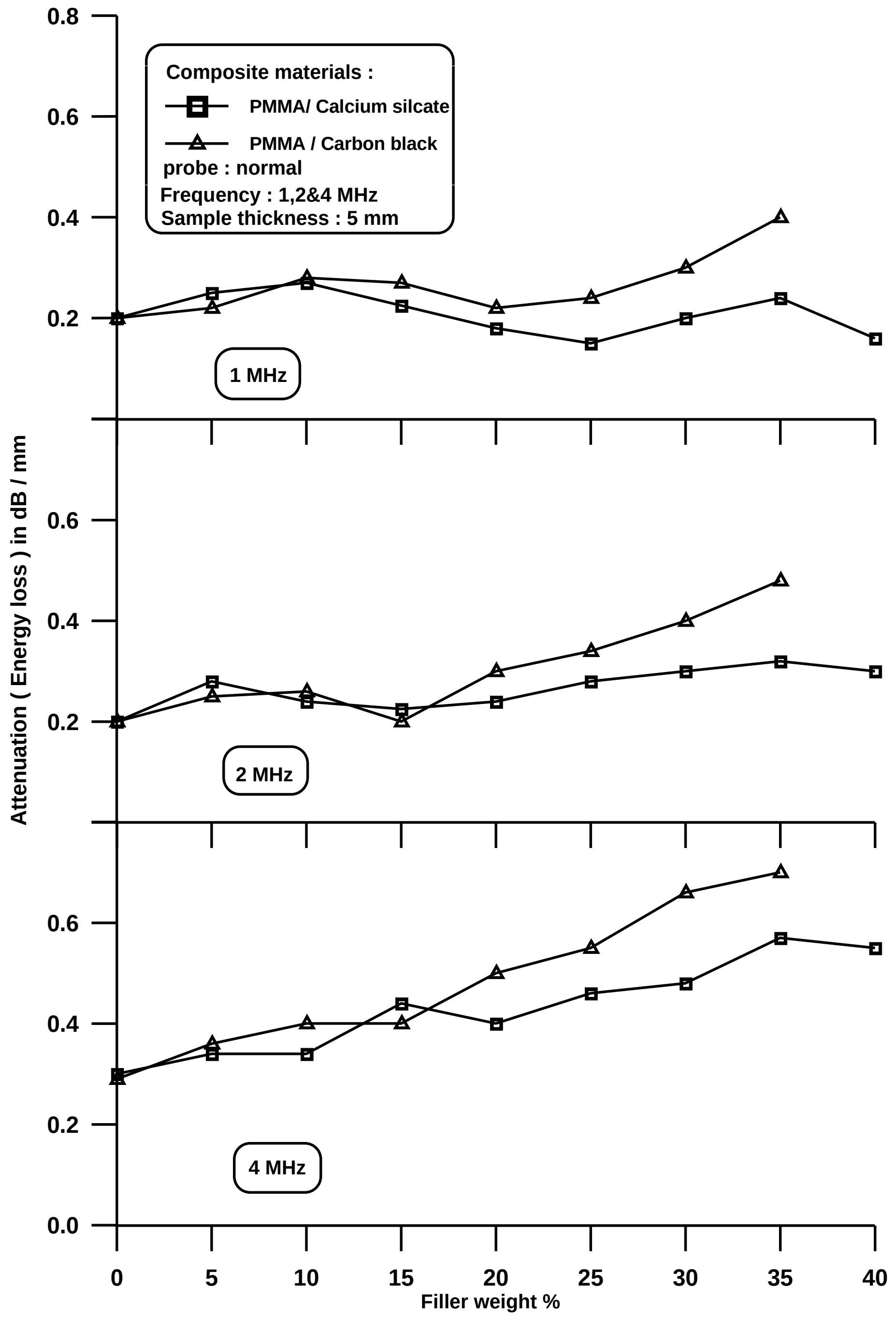

Figure (7) The effect of filler weight % on the ultrasonic attenuation (energy loss) of filled moulded acrylic (PMMA) using normal probes of 1,2 and 4 MHz frequency at temperature 25 °C

Figures (8) and **(9)** show the ultrasonic echo amplitude response from samples of 10 mm and 15 mm thickness. 1 MHz probe gave the highest ultrasonic response, and the 4 MHz gave the lower ultrasonic response and most sensitive to the filler content and that possibly related again to its higher ultrasonic attenuation characteristics. Again the most suitable probe for inspection was the 2 MHz and that demonstrated during the test. **Figure (10)** confirm these results and show the higher ultrasonic attenuation of the 4 MHz probe. Clear correlation was seen between filler content and ultrasonic attenuation beyond (15%) filler weight percentage. **Figure (11)** shows a correlation between the acoustic properties and the hardness properties using the 2 MHz after it has been established as the superior probe among the probes involved. The clear correlation was also shown beyond (15%) filler content also, that as attenuation increases the velocity decreases and the hardness take a similar trend. The reduction in hardness possibly related to the agglomeration of the tough filler materials that impart the Acrylic matrix PMMA and make it ductile, i.e. increasing filler content beyond (15%) accompanied by reduction in the hardness and increase in ductility and that possibly related to imparting the composite matrix by the soft fillers contents beyond this percent (%). Because of the increase in ductility, the damping capacity of the composites increases. Hence it is able to absorb a greater amount of sonic energy and increase in ultrasonic attenuation and gradual reduction in ultrasonic velocity [10].

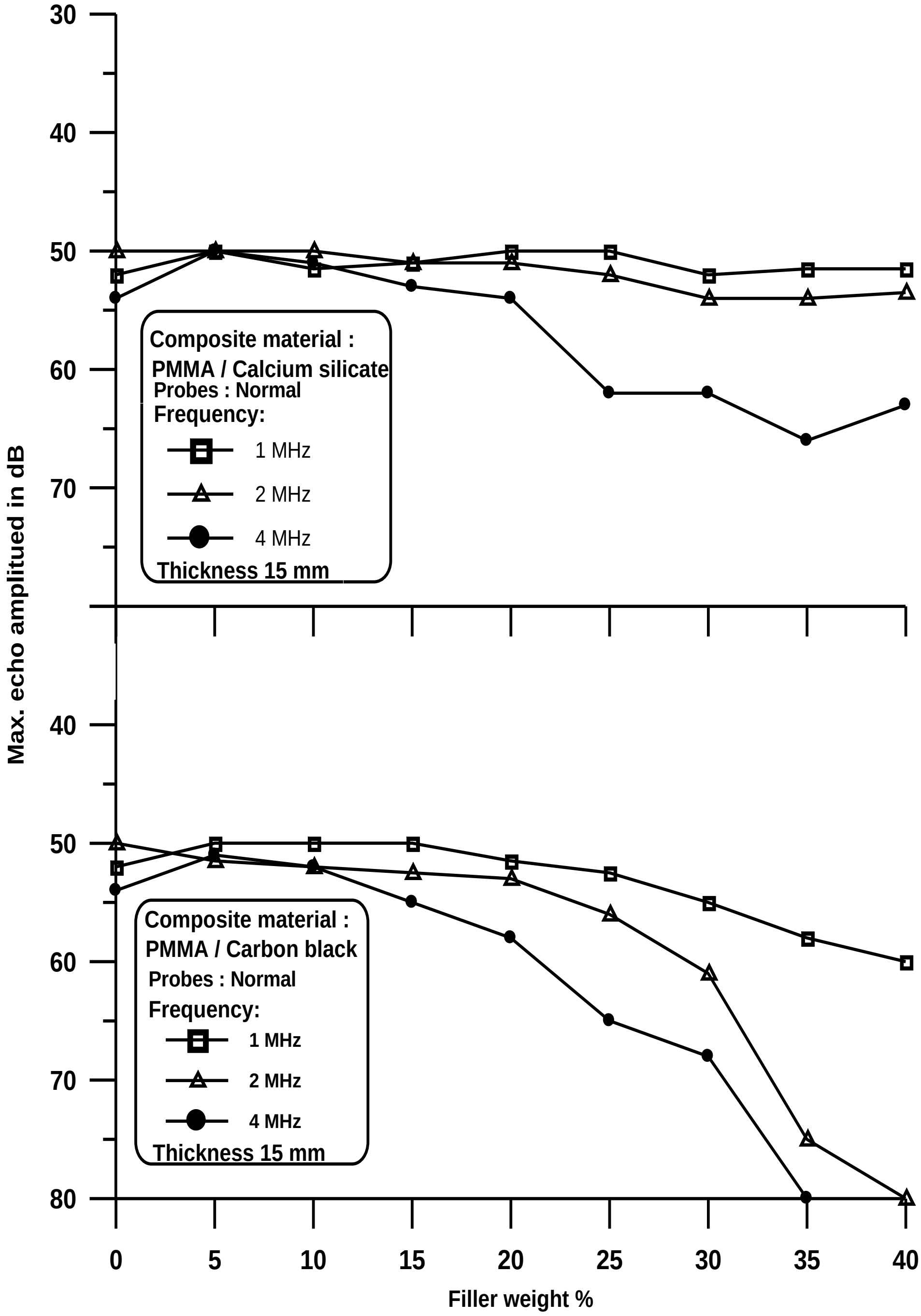

Figure (8) The effect of filler weight % in moulded acrylic (PMMA) on the ultrasonic response using normal probes of 1,2 and 4 MHz frequency at temperature 25 °C

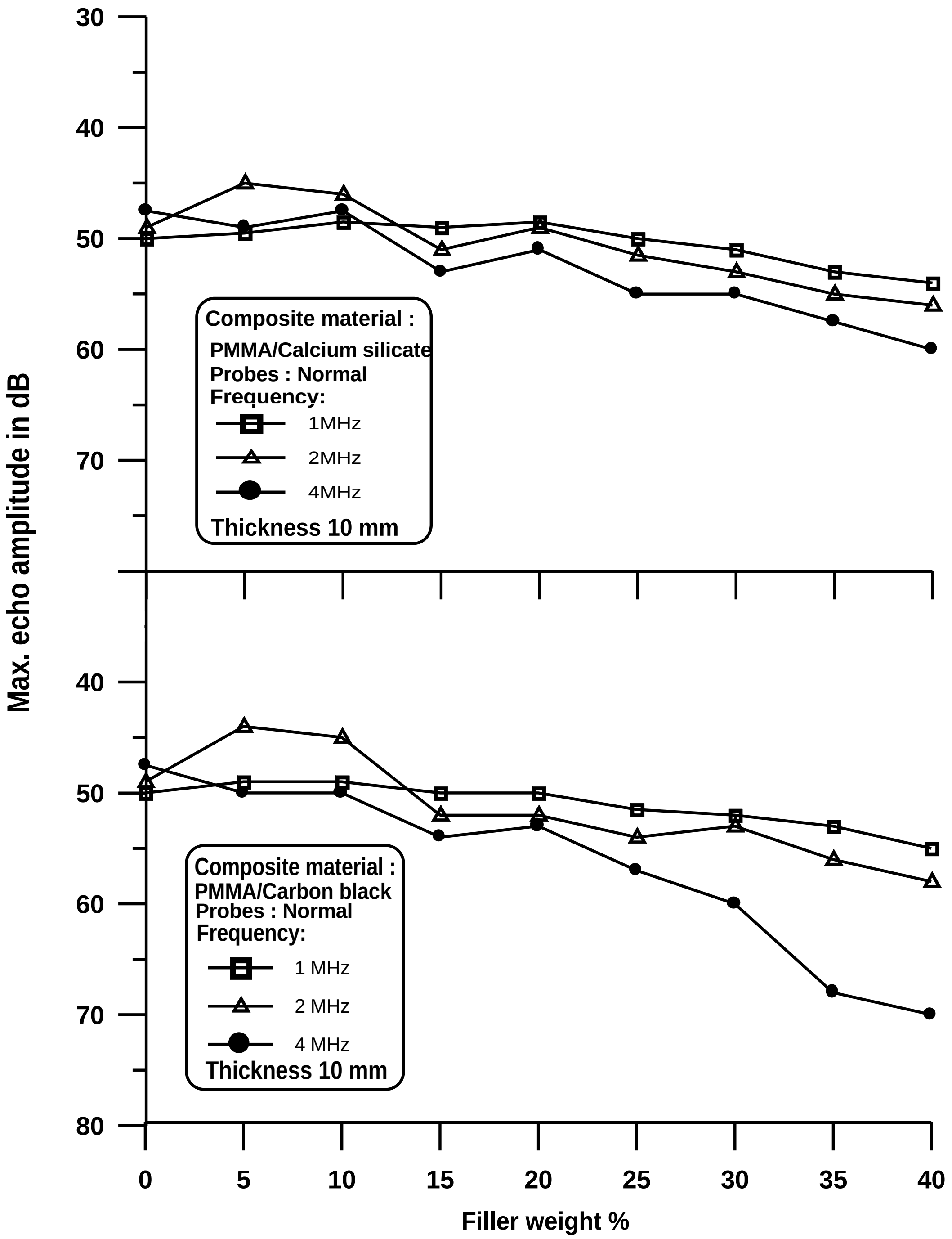

Figure (9) The effect of filler weight % in moulded acrylic (PMMA) on the ultrasonic response using normal probes of 1,2 and 4 MHz frequency at temperature 25 °C

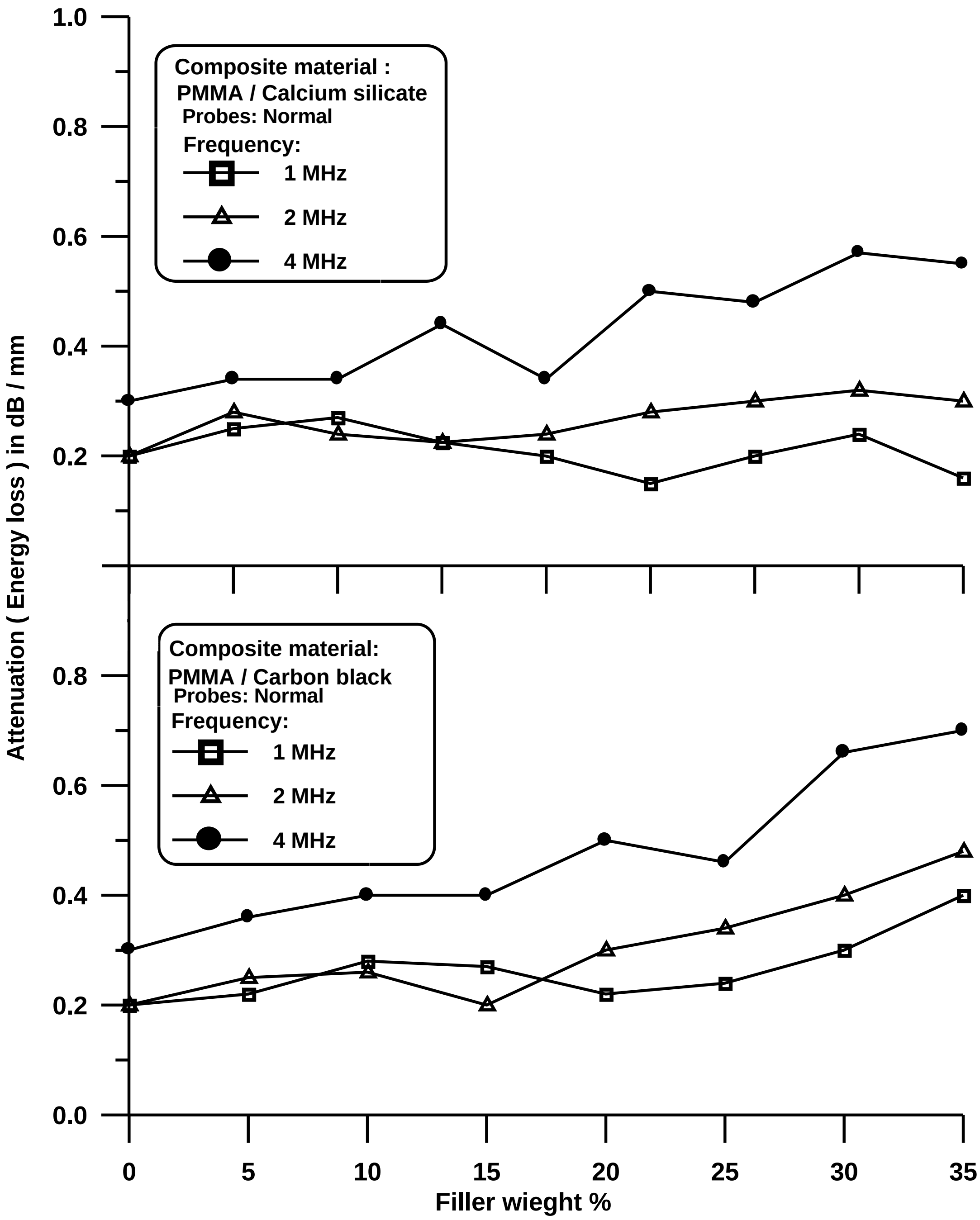

Figure (10) The effect of filler weight % on the ultrasonic attenuation (energy loss) of filled moulded acrylic (PMMA) using normal probes of 1,2 and 4 MHz frequency at temperature 25 °C

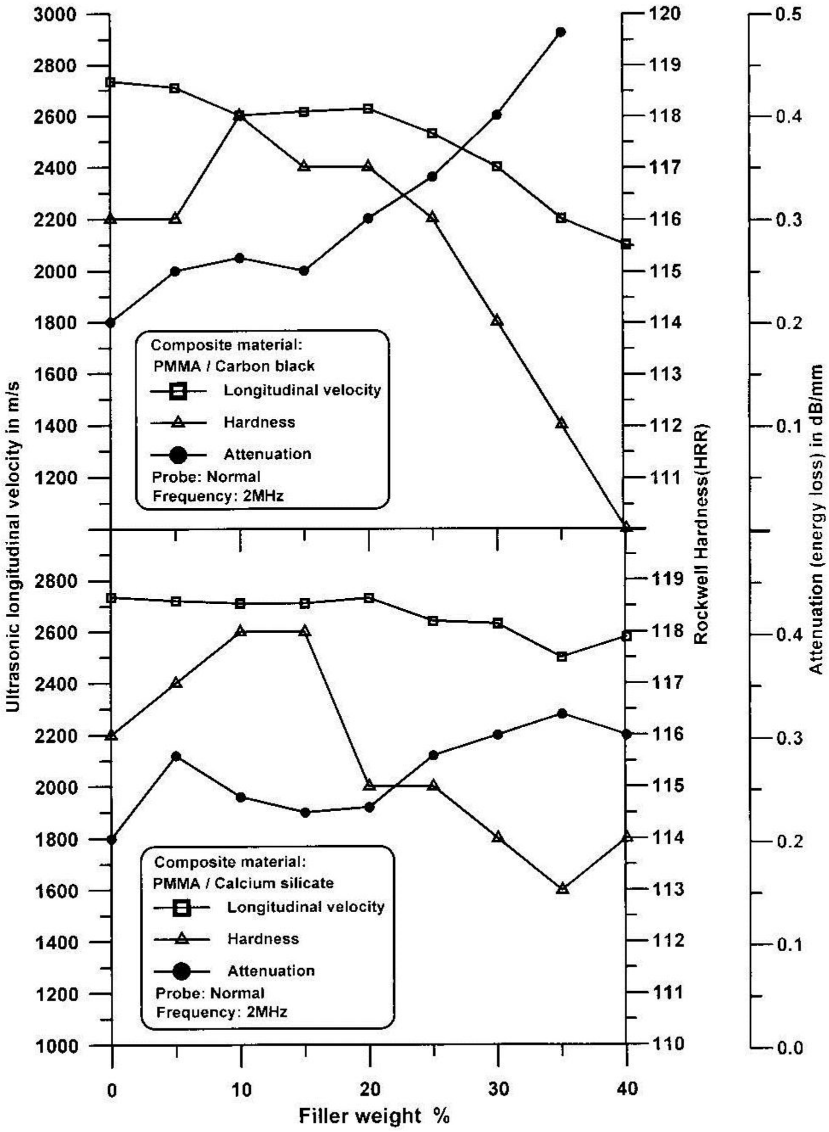

Figure (11) Ultrasonic longitudinal velocity, hardness and attenuation correlation of filled moulded acrylic (PMMA) with different filler weight % at temperature 25 °C

4. Conclusions

1. Better investigation on the effect of filler content in composites could be achieved using the pulse-echo amplitude technique.
2. Sample thickness (sound path) of 10 mm is most suitable for ultrasonic investigation of the filled polymer.
3. The filler type has considerable effects on the ultrasonic response.
4. Probe 1 MHz is not suitable for test and not sensitive to filler content variation and composites texture.
5. For clear correlation and systematic investigation, all measurements should be carried out in the zone (15 – 35) filler weight %.
6. Best response and acceptable sensitivity to the composite material texture were obtained using the 2 MHz probe that is superior to 1 & 4 MHz probes.

5. References

1. James P. Schaffer, Ashoke Saxena, Stephen D. Antolovich, Thomas H. Sanders, Jr., Aqnd Steven B. Warner, ***"Composite Material"***, The Science and Design of Engineering Material, Richard D. Irwin, Inc., 1995 Chicago, VSA, pp. 595-630.

2. Gururaja T. R., Walter A. Schulze, Leslie E. Cross, Robert E. Newnham, Bertram A. Auld, and Yuzhong J. Wang, ***"Piezoelectric Composite Material for Ultrasonic Transducers Applications, Part I: Resonant Modes of Vibration of PZT Rod-Polymer Composites"***, IEEE Transactions on Sonics and Ultrasonics, Vol. 50-32, No.4, July 1985.

3. Bhardwaj M. C., Neeson I., and Stead G., ***"Introduction to Contact-Free Ultrasonic Characterization and Analysis of Consolidated Materials"***, Application of NDE in Power Metals Seminar, Iowa State University, Ames, IA, USA, Aprile 25, 2000.

4. Eric I. Madaras, ***"Ultrasonic Attenuation Results of Thermoplastic Resin Composites Undergoing Thermal and Fatigue Loading"***, NASA Langley Research Center, MS 231, Hampton, VA23681-0001.

5. Dagodinas D., ***"Ultrasonic Signal Processing Methods for Detection of Defects in Composite Material"***, NDT material, July 2003, Vol. 8, No.7.

6. Cheng-kuei Jen, and Ngoc Huynh, ***"On-line Ultrasonic Technology for Monitoring of Polymer Processing"***, Industrial Materials Institute-National Research Council Canada, April 23, 2003.

7. I. G., Scott, and C. M., Scale, ***"A Review of Non-Destructive Testing of Composite Materials"***, NDT International, Published by Butter Worths, Vol. 16, No. 3, June 1983, pp. 137-192.

8. C. H., Gür, and B. Aydinmakina, ***"Ultrasonic Investigation of Graphite Modularity in Ductile Case Irons"***, German Society for Material Testing, NDT Annual Conference, Proceeding 75-CD, Berlin, 21-23 May 2001.

9. Krautkarammer, J., and Krautkarammer, H., ***"Ultrasonic Testing of Materials"***, 2nd Eddition, Springer-Verlag, Berlin, Heidelberg, New York , 1977.

10. Prasad, R., and Kumar, S., ***"An Investigation into the Ultrasonic Behaviour of Cast and Heat-Treated Structures in Steel"***, British Journal of NDT, Vol. 33, No. 10, October 1991, pp. 506-508.