POLYMER-POLYMER MELTED STRUCTURE: EXPERIMENTAL STUDY OF THE NONLINEAR ACOUSTIC BEHAVIOR

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Duflo Hugues[1]; Izbicki Jean Louis[1] ; Creton Costantino[2]

1 Laboratoire LAUE
Place Robert Schuman - BP 4006 - 76600 Le Havre - France
Tel : +33 2 32 74 47 38
E-mail : hugues.duflo@iut.univ-lehavre.fr

2 Laboratoire PCSM-ESPCI
10, Rue Vauquelin - 75231 Paris Cédex 05 - France
Tel: +33 1 40 79 46 83
Fax: +33 1 40 79 46 86
E-mail : Costantino.Creton@espci.fr

ABSTRACT Two plates of an identical glassy polymer are heated together at a temperature above their glass transition temperature, during different times (td). An acoustic method is used to show the nonlinear effects occuring when acoustic waves are transmitted through the polymer-polymer structure. First, the resonance of the structure are obtained. Then a signal composed of two different harmonic waves (none of them corresponding to a resonance frequency) is synthesized : resonances are obtained because of the intermodulation phenomenon. First results show that density power spectrum of the intermodulation varies with td.

INTRODUCTION

The goal of this paper is to investigate the nonlinear behavior of a polymer-polymer melted structure. Preliminary experimental results are shown. The process of polymer-polymer interdiffusion is studied with an acoustic method. When both plates of polymer are heated together at a temperature above their glass transition temperature, polymer chains start to diffuse[1] in a zone we call interphase. The density of molecular chains increases with the time of heating in this zone[2]. Previous acoustic characterization of the adhesion[3] shown that a correlation exists between annealing time, thickness of the interphase and width of the resonance.

We studied here the nonlinear acoustic behavior of a transmitted wave for two different angles. We present firstly the system of the two plates and its acoustic response. Then the experimental setup is presented and the nonlinear effect of the acoustic wave transmission is shown.

EXPERIMENTAL SETUP

For this study, the polymer is a linear polystyrene (average molecular mass: 250 Kg/mole). Its elastic tensile modulus E at 1 Hz was measured at 3.3 GPa and its glass transition temperature was measured by a differential calorimetric method (DSC) at 98°C (Its tradename provided by Elf-Atochem is Lacqtene). The two plates are melted during different times. We present results for two different systems of plate with first heating time: t_1=7mn, and the second time t_2=17mn.

First we present the experiment which determine the angular transmission with a pulse excitation. Then a new experimental setup is described to identify the nonlinear effects.

Angular transmission with pulse excitation

The bistatic method used two broadband transducers. One acts as transmitter and the other as a receiver (Fig. 1)

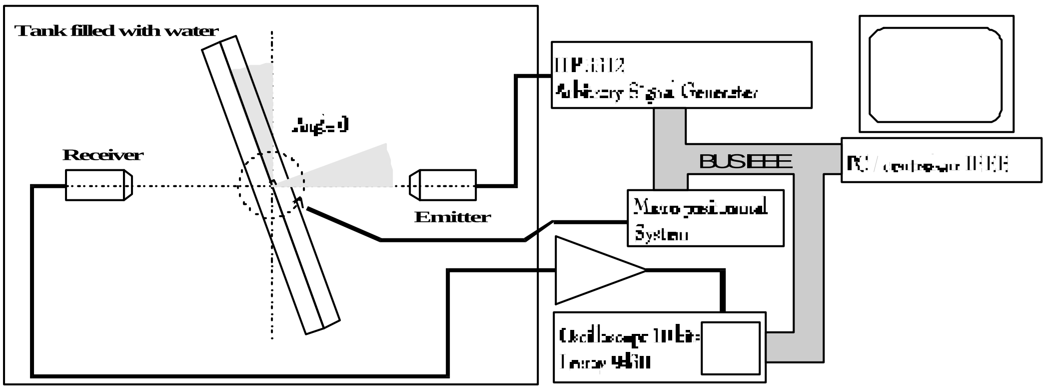

Figure 1: Experimental setup: bistatic method.

An angular response is obtained for the system 1 (t_1=7mn). The angle varies from 0 to 40 degrees (by 0.1 degree step) and the electric signal sent to the emitter is a pulse with 300V amplitude. A FFT is applied on each signal (for the different angular position). On the figure 2, we see that a critical angle exists (near 39 degrees). For our following experiments, we choose two angles, one near 39 degrees and one inferior: 35 and 20 Degrees.

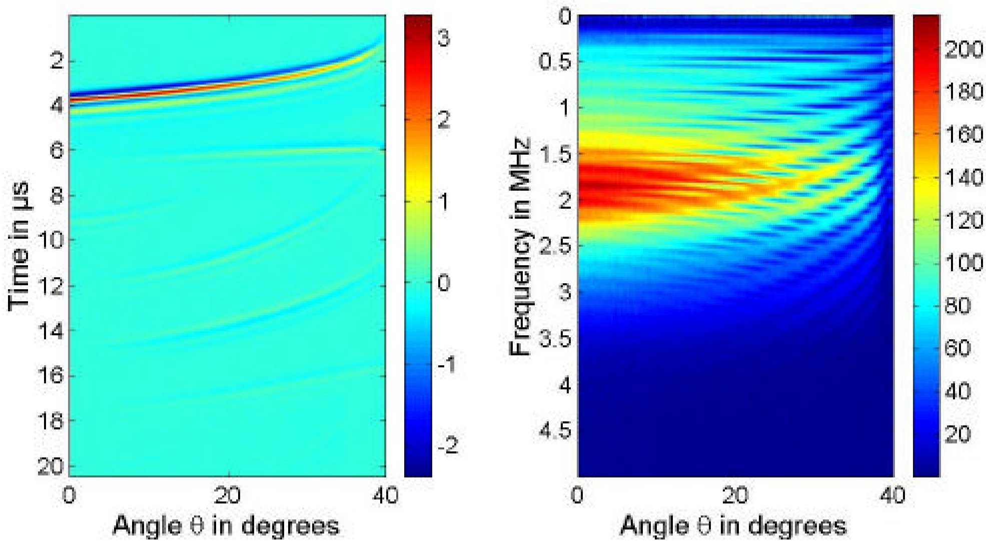

Figure 2: Signal acquisition and spectral analysis for different angles.

Nonlinear effects

To detect the nonlinear behavior of the transmission wave, a signal is built with an arbitrary wave generator which contains two frequencies. This burst signal (Fig. 3) is sent to the emitting transducer. The duration time is 100µs. To have a complete description, we have automated the experiment to program the generator for f_2=1kHz to 400kHz by 1kHz step. For each frequency f_2, we store the received signal (by the oscilloscope) in the computer. The f_1 frequency is chosen close to 1MHz, and corresponds to a minima for the transmission in the spectrum for the angle teta.
The results (Fig. 4) of an experimentation for θ=35 degrees for the system t_1=7mn give first an image (Fig. 4a) where the horizontal axis is f_2 (0 to 400 KHz) and the vertical one is the time

(with the delay due to the propagation of the wave in water). After processing the FFT for each angle we obtain a second image (Fig. 4b). The figure shows the two principal components of the signal, a horizontal line at $f=f_1=1.19MHz$ and an oblique line $f=f_2$. To observe nonlinear effects without perturbation, we made a zoom of a region between $f=2.f_1$- 400KHz and $2.f_1$+400KHz.

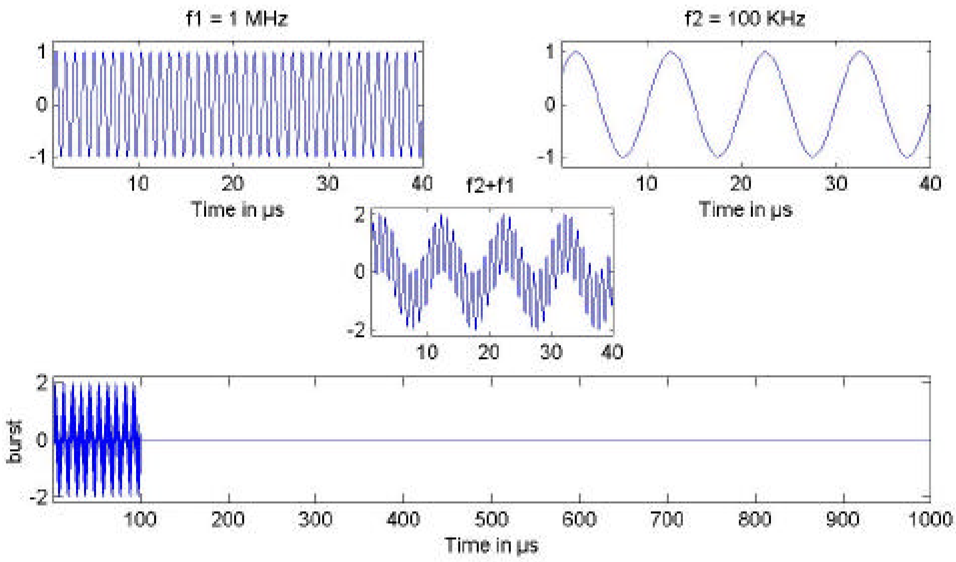

Figure 3: Building of the emitting signal

The third figure shows us two oblique lines which corresponding to the relation $f=2.f_1+f_2$ (between 2.4 and 2.8 MHz) and $f=2.f_1-f_2$ (between 2 and 2.4 MHz). There are more oblique lines which are due to the signal at $f=f_1+m.f_2$ with m=1,2,3,4,…. All these oblique lines are the proof that intermodulation and nonlinear effects exist.

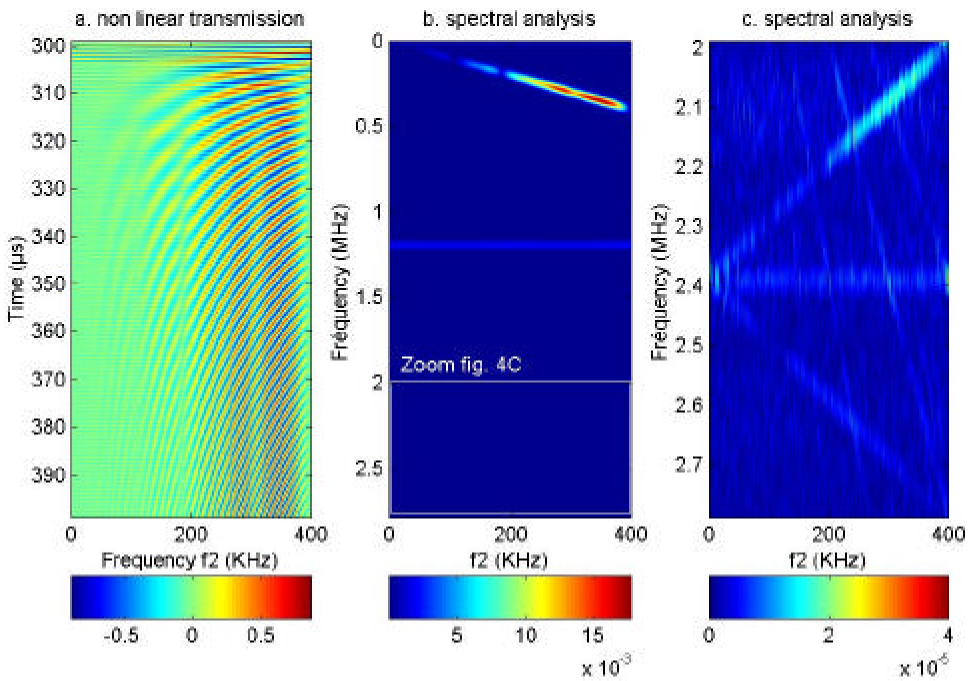

Figure 4: Plate 1 (t1=7mn, f1=1.19MHz), temporal and spectral results, transmission for θ=35degrees

ANALYSIS

To study the nonlinear interaction, we have chosen to take the part of the spectral results around $f=2.f_1$. Then we can measure the amplitude of the signal on the oblique lines. Two different lines with equation $f=2.f_1-f_2$ and $f=2.f_1+f_2$ are interesting. We have also plotted on Figure 5 the amplitude on the line of equation f=f2 (Oblique line on Fig. 4b between 0 and 0.4MHz). Next (Fig. 5b) we have drawn the amplitude on $f=2f_1-f_2$. The signal is very noisy, then we have performed a summed average (Fig. 5c). To have a normalized response, we have finally divided the average signal by the free response of the signal at $f=f_2$. The last figure is the normalized amplitude that we are going to compare for different angles and for the two systems of plate (All the normalized amplitude are multiplied by 10000).

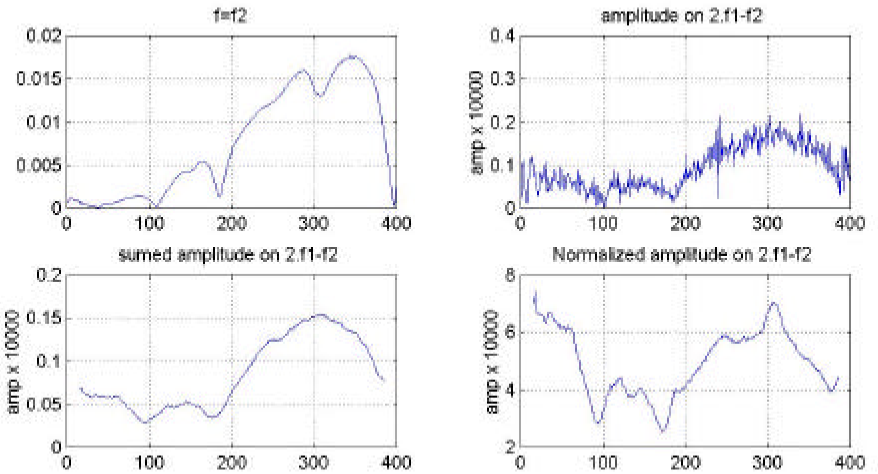

Figure 5: Amplitude measurement on oblique lines of the spectral results, (Plate 1: t1=7mn, f_1=1.19MHz, θ=35degrees).

<u>Numerical results</u>

In all cases, angle equal to 20 or 35 degrees and time=7 or 17 mn of heating, we present the numerical analysis made on the computer. For the four case, we present the zoomed zone of the spectral analysis with f between $2f_1$-0.4 and $2f_1$+0.4MHz and the normalized amplitude defined as previous part for the two oblique lines $f=2f_1+/- f_2$.

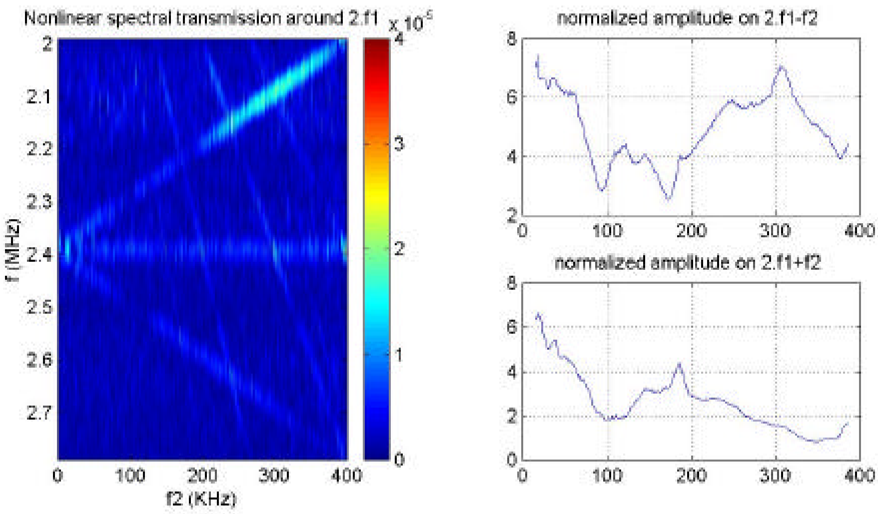

Figure 6: Results for plate 1 (t1=7 mn) spectral results for θ=35 f_1=1.19MHz

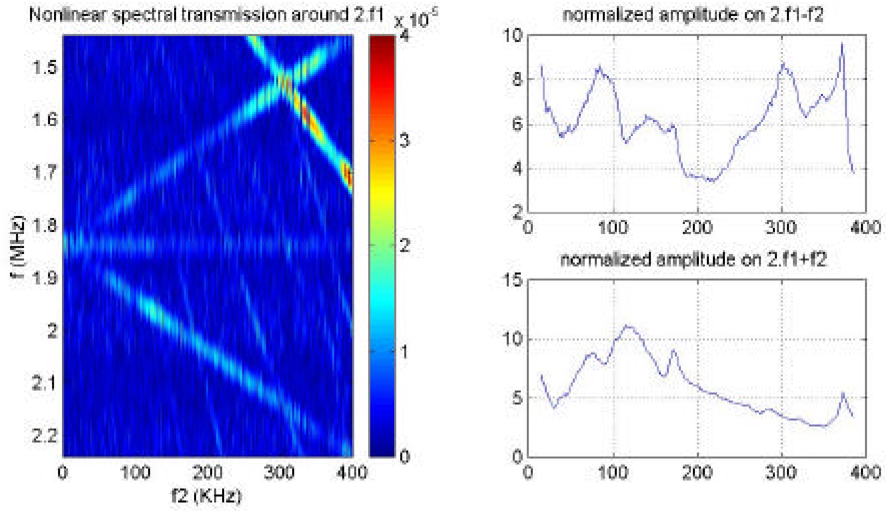

Figure 7: Results for plate 2 (t1=17 mn) spectral results for θ=35 f_1=0.92MHz

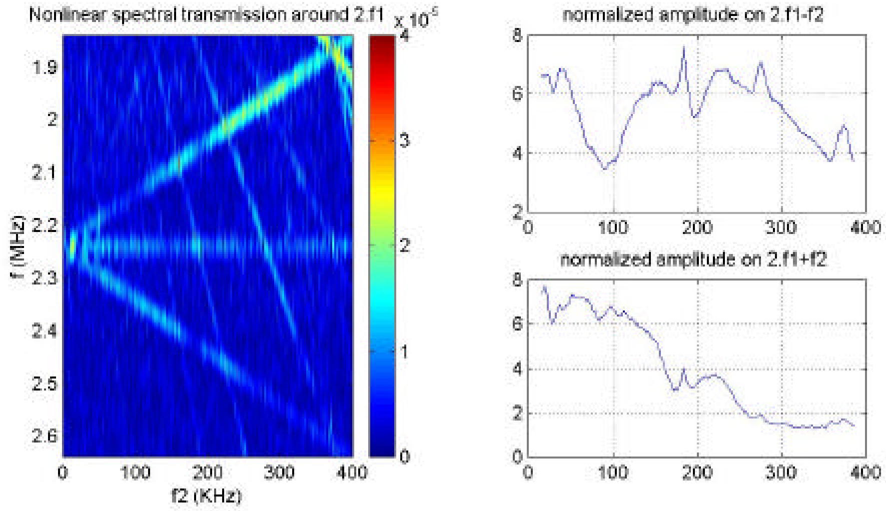

Figure 8: Results for plate 1 (t1=7 mn) spectral results for θ=20 f_1=1.12MHz

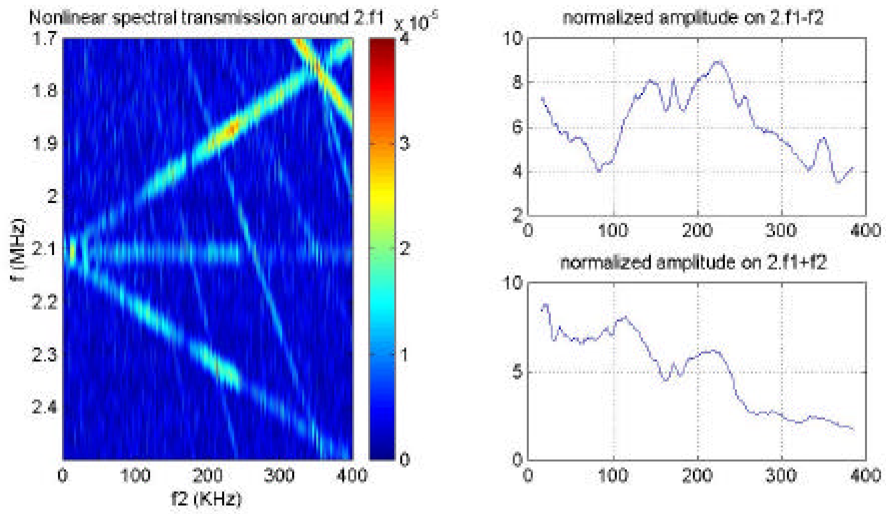

Figure 9: Results for plate 2 (t1=17 mn) spectral results for θ=20 f_1=1.05MHz

The results on all figures seem to be different. On the other hand, for a given angle, curves are similar for two different annealing time. All these experiments have been made in the same conditions. For the four experimental configurations (Fig. 10) the blue curves (o) are nearly always higher than the red curves (+).

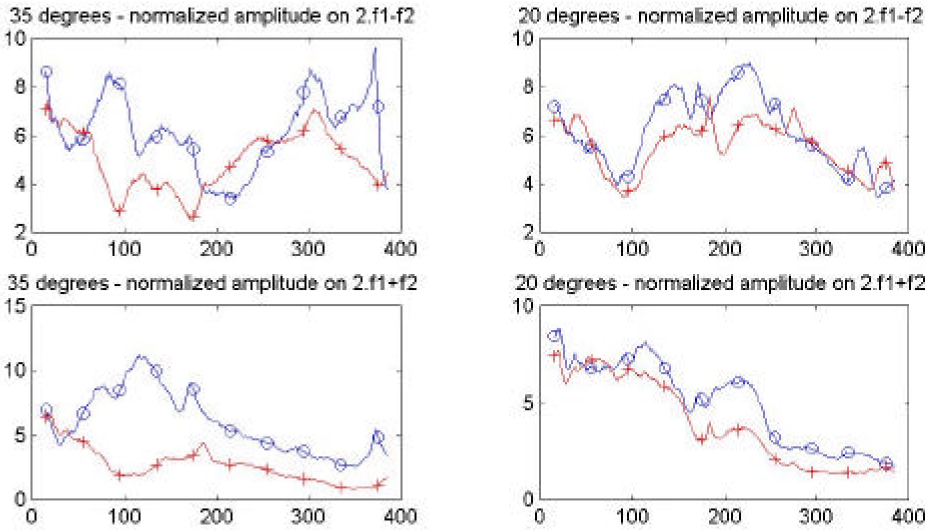

Figure 10: Global results of the normalized amplitude for the two angles and the two duration time of annealing (+) for t_1=7 mn and (o) for t_2=17mn

CONCLUSION

The acoustic method permits us to show nonlinear effects. The system of two plates of polymer is sensible in transmission to a particular signal with two harmonic waves at frequencies f_1 & f_2. The nonlinear effects are proved by observing signal with frequency $f=m.f_1+n.f_2$ ($m,n \in$ □). The numerical analysis of the acquired temporal signals shows clearly these oblique lines. The normalization of the amplitude on these lines have been plotted. Globally, the nonlinear effects increase with the time of heating as same as the diffusion of polymer chains.

The experiments must be continued. The making of new system of plate with more different time of heating is necessary to have intermediate results. We hope so determine more fine correlation between the heating time and the nonlinear effects in order to determine relation with some adhesion coefficients.

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