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Non-collinear wave mixing for non-linear ultrasonic detection of physical ageing in PVC

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ABSTRACT

This work considers the characterization of linear PVC acoustic properties using a linear ultrasonic measurement technique and the non-collinear ultrasonic wave mixing technique for measurement of the physical ageing state in PVC. The immersion pulse-echo measurements were used to evaluate phase velocity dispersion and attenuation of longitudinal waves in PVC test specimens. The suggested non-collinear ultrasonic wave mixing technique measurement technique was verified on measurements of laboratory and field PVC test specimens. The measurement results confirm that the ultrasonic wave mixing technique is suitable to estimate the physical ageing state of PVC.

1. Introduction

Polyvinylchloride (PVC) is an amorphous polymer offering good performance at low cost. As such, it is widely used (about one quarter of the global annual polymer production is PVC), especially in infrastructural and building applications, e.g., in water and gas distribution networks. From the moment when an amorphous polymer is cooled down through the glass transition region, it undergoes a process that is called physical ageing or structural relaxation [1], in which the polymer chains slowly move towards their thermodynamically favoured positions. This process is characterized by time-dependent changes of bulk properties such as specific volume, enthalpy, entropy, mechanical and dielectric response, which attempt to establish thermodynamic equilibrium. More specifically, the yield stress increases over time, accompanied by a decrease of fracture toughness due to localization of the plastic deformations [2]. This implies that PVC structures such as supply systems for gas and water will get more susceptible to third party damage over time. The degraded parts of these supply systems need to be replaced in a timely fashion to prevent the risks involved with pipe failure. However, up to now there is no non-destructive inspection system available to identify the necessity of replacement or the residual lifetime of (parts of) these pipeline networks.

Measurements of the ultrasonic wave velocity and attenuation are widely used for characterization of acoustic properties of polymers [3]. In this study an ultrasonic longitudinal wave was used for the initial characterization of PVC test samples in terms of phase velocity dispersion and attenuation of the wave.

The ageing-induced changes in the linear elastic properties of PVC are small and it is hard to detect them in situ when the plastic construction is embedded or/and when only single-sided access to the structure is possible. The (linear) elastic polymer properties of PVC have been studied widely using ultrasonic measurement techniques [4–11], but none of these references consider the process of physical ageing of PVC. Therefore the measurement results may vary slightly when repeating the measurement. Physical ageing was, however, investigated with other measurement techniques for a wide range of glassy polymers. In particular, physical ageing of PVC was investigated by means of:

- measurement of the creep compliance [12–17];
- measurement of the heat distortion temperature [18];
- differential scanning calorimetry [19–22];
- dynamic mechanical spectroscopy (dynamic mechanical analysis) [23];
- tensile test measurements [2,24,25];
- micro-indentation tests [26].

Obviously, most of the listed measurement techniques are hardly suited for non-destructive field inspection of physical ageing in polymers, or the measurement technique is limited when the critical spots can be in the center of the material. For example, micro-indentation tests correlate with the physical state ageing in PVC, but this measurement technique is limited to the

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surface properties of the polymer structure. The literature indicates that the measured tensile compliance loss as a function of frequency has a peak that is almost unaffected by the PVC physical ageing over a wide range of ageing time (10^{-8}–10^{6} s) at 23 °C [12]. The reported investigations were performed over a wide frequency range, from 0.01 Hz to 5 MHz and a part of the measurements was carried out using ultrasonic waves. The tensile compliance loss peak was obtained around 30 kHz. The reported low frequency of the peak and low sensitivity of acoustic waves to physical ageing of PVC show that linear acoustic measurements are not suitable for determination of the physical ageing state in PVC. There is too little information in the open literature about the possibilities to estimate the physical ageing state in various polymers using an ultrasonic measurement technique.

It is well known that non-linear ultrasonic measurements are more sensitive to certain material properties in comparison with linear ultrasonic measurements. Three different "classic" non-linear wave approaches are used in characterization of material properties: (a) acousto-elasticity, (b) wave mixing and (c) resonance frequency shift and modification of the quality factor of vibrating bars, when the acoustic amplitude increases [27]. The second approach was used here, and will be discussed in more detail.

Two different approaches can be distinguished in wave mixing: collinear wave mixing, better known as harmonic generation, and non-collinear wave mixing, when two waves intersect at a certain angle and a third wave is generated in the intersection volume. More than two waves can be mixed in the general case. Previous work shows that the measurements based on harmonic generation are able to detect fatigue [28–31] and physical ageing [32–36] in solids. Most of the reported studies show that most of the harmonic generation investigations were carried out with metallic alloys, except the investigation of hydrothermal ageing of metal-epoxy bonds [36]. Notably, no change in nonlinearity of the metal-epoxy bond was observed due to hydrothermal ageing.

Experimental results of non-destructive testing and material evaluation using non-collinear wave mixing are reported only for metallic alloys, see e.g., Refs. [37–39]. Recently, the non-collinear wave mixing technique was assessed for fatigue detection purposes [39].

Here a non-collinear wave mixing method was selected for the investigation of physical ageing in PVC. The non-collinear wave mixing technique has the following advantages over the harmonic generation technique (or collinear wave mixing):

- possibility of wave mode separation;
- possibility of frequency separation;
- elimination of the surrounding medium influence;
- possibility to steer a scattered wave (spatial separation);
- possibility to control the location of the intersection volume;
- possibility to use a low power ultrasonic measurement technique;
- possibility of single-sided or double-sided access.

However, the non-collinear wave mixing technique is more complex compared to the harmonic generation technique. The technique requires special conditions such that resonance between the interacting waves will occur in order to generate the interaction wave.

2. Measurement of phase velocity and attenuation of longitudinal waves

An immersion pulse-echo measurement setup was used to measure longitudinal wave phase velocity dispersion and attenuation. A broadband transducer of 2.25 MHz center frequency with an active area diameter of 10 mm was used to generate and to receive longitudinal waves. The transducer was positioned perpendicularly to the surface of the test specimen maintaining 30 mm distance between the transducer and the test specimen. The transducer was excited using a single rectangular pulse of 2 MHz.

The phase velocity of the longitudinal wave was derived from the measurement data employing a phase spectrum method [40]. The attenuation of the longitudinal wave in the test specimen was estimated using a spectral approach as presented in [41]. Influences of diffraction and mismatch of acoustic impedances between the test specimens and water were compensated in the measured attenuation data [41].

3. Non-collinear wave mixing measurement method

The synchronism conditions for two interacting phonons $\mathbf{k}_1$ and $\mathbf{k}_2$ can be written in terms of conservation laws for quasi-momentum and energy of interacting phonons [37,42,43]:

$$\mathbf{k}_1 \pm \mathbf{k}_2 = \mathbf{k}_3, \tag{1}$$

$$\omega_1 \pm \omega_2 = \omega_3 \tag{2}$$

where $\mathbf{k_i}$ (i=1,2,3) is the wave-vector of the phonon, ω_i is the angular frequency of the phonon and $\omega_i = 2\pi f_i$. f_i is the frequency. The synchronism conditions are necessary, but not sufficient. The allowed interaction cases and their conditions are presented in literature [42,44], and will not be repeated here. The interaction between shear and longitudinal waves was found most suitable to evaluate PVC when using immersion transduction for the measurements. The wave interaction process is denoted by $S(\omega_1)+L(\omega_2)\rightarrow L(\omega_1+\omega_2)$, where $S(\omega_1)$ and $L(\omega_2)$ are the shear and longitudinal pump waves, respectively, and $L(\omega_1+\omega_2)$ is the longitudinal response wave. The frequencies of the two pump waves were selected such that the response wave would strike the PVC/water interface perpendicularly. Fig. 1 presents the arrangement of the transducers in the measurement set-up for a test specimen of 25 mm thickness. Two broadband transducers of 2.25 MHz center frequency with an active area diameter of 10 mm were used to generate the pump waves. The shear wave pump transducer was inclined at an angle $\theta_S = 53°$ and driven by a burst of 30 cycles of rectangular pulses of $f_1 = 1.8$ MHz and 0.2 V amplitude. The longitudinal wave pump transmitter was inclined at an angle $\theta_L = 32°$ and driven by a burst of 30 cycles rectangular pulses of $f_2 = 2.7$ MHz and 0.2 V amplitude. Both pump wave signals were amplified using broadband RF amplifiers. A gain of

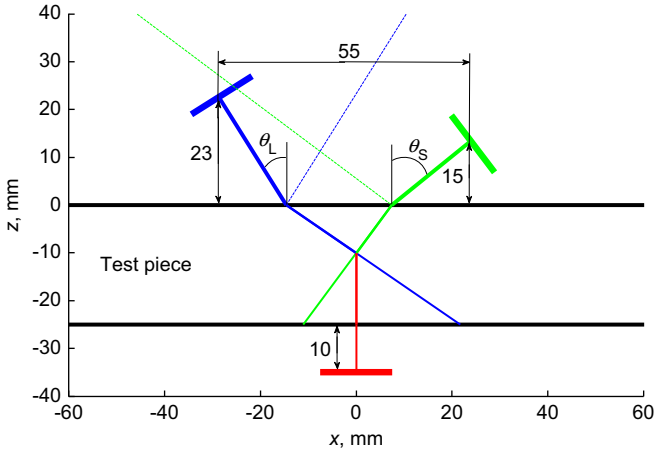

Fig. 1. Arrangement of transducers for the non-collinear wave mixing process $S(\omega_1)+L(\omega_2)\rightarrow L(\omega_1+\omega_2)$ in PVC. The dashed lines indicate the specularly reflected pump waves.

40 dB was used in the experiments. The values of angles θ_S and θ_L are approximate, because fine adjustment of the angles was performed manually using rotary stages with a tuning resolution of 0.08°. The generated wave of 4.5 MHz was received by the broadband 5 MHz center frequency with an active area diameter of 10 mm receiver on the opposite side of the test specimen. A home-made preamplifier of 20 dB was used for amplification of ultrasonic signals. A through-transmission mode was used in order to achieve a better signal to noise ratio, as the energy reflection coefficient from the PVC and water interface is low. The received signals were filtered using a narrowband FIR filter with a Kaiser window and stored on a personal computer for further analysis.

4. Test specimens

Two sets of PVC test specimens were prepared for measurements with the non-collinear wave mixing technique: the "laboratory" and the "field" specimens. The laboratory PVC test specimens were prepared from a commercially available extruded flat plate of 25 mm thickness. The new plate was without induced fatigue and should have uniform elastic properties and little thickness variation. This was not to be expected in the used PVC specimens. The dimensions of the prepared samples were 25 mm × 25 mm × 200 mm. The test specimens were heated above the glass transition temperature of PVC at 110 °C for one hour to erase the previous thermal history of PVC. After heating, the samples were quenched immediately in antifreeze fluid of −30 °C, thus completing the rejuvenation process of the samples. Knowing that the glass transition temperature is about 80 °C or more in PVC [45], the test specimens were annealed at a temperature of 60 °C for a range of time intervals. Annealing at higher temperatures (but below the glass transition temperature) leads to accelerated physical ageing. The time intervals are listed in Table 1.

The second set of PVC test specimens was prepared from an old PVC pipe that had been in use for drinking water supply service for several decades. The pipe had an external diameter of 600 mm and a wall thickness of nominally 18 mm. 16 bar shaped test specimens were cut from the pipe in circumferential direction, approximately covering one quarter of the circumference (see Fig. 2). In order to maximize the amplitude of the received signal, the vertical distance between the transmitters and the top surface of test specimens was increased by 7 mm (compare Fig. 1) and the incident angles of the pump waves were changed to $\theta_S = 50°$ and $\theta_L = 31°$. Thus, the interaction volume of the pump waves was steered to the middle of the test specimens, and a difference in acoustical properties was accounted for, as will be discussed later. As such, the need for this modification already clearly indicates a difference between the elastic properties of the laboratory and the field PVC test specimens.

5. Measurement results

Knowing that the elastic properties of polymers are strongly dependent on temperature [46], the temperature of the immersion fluid was monitored during the experiments using a digital thermometer with a thermocouple. The temperature of the water was 20.0 °C ± 0.1 °C and 20.9 °C ± 0.3 °C in all pulse-echo measurements and non-collinear wave mixing experiments, respectively. To minimize random noise errors in a spectral analysis of ultrasonic signals of pulse-echo measurements, the ultrasonic signals were averaged from 256 signals. There was no averaging applied during capturing and processing of the ultrasonic signals in the presented results of the non-collinear wave mixing measurements.

Averaged values of the phase velocity measurement in both groups (laboratory and field) of test samples are presented in Fig. 3. A difference is observed between the phase velocities of the longitudinal waves in the laboratory and field PVC test samples. The error bars show significant scatter of the phase velocity in the field PVC test specimens. This higher scattering is caused by a non-uniformity of thickness and surface properties of the PVC test specimens. The mean standard deviations are ± 4 m/s and ± 7 m/s for the laboratory and field test samples, respectively. The difference between the velocities confirms the need of fine readjustment of incident angles in non-collinear wave mixing experiments for evaluation of the laboratory and field PVC test specimens.

The absolute values of the measured phase velocity are listed together with the attenuation in the laboratory samples at the central frequency of 2 MHz in Table 1. The cross-correlation coefficient between the phase velocity and attenuation is 0.45. This low correlation confirms the earlier reported low sensitivity of acoustic waves to physical ageing of PVC [12], clearly showing that the linear ultrasonic waves are not sufficiently sensitive to physical ageing of the polymer.

The measured attenuation of the longitudinal waves shows more attenuation in the field test samples of PVC (see Fig. 4). Standard deviations are 8.5 dB/m and 17 dB/m in the laboratory and field test specimens of PVC, respectively.

The measured attenuation was fitted by a linear polynomial using a least-squares method. The following equations are obtained for laboratory and field test specimens of PVC, respectively:

$$\alpha_l = 228.97[\mathrm{dB}/m/\mathrm{MHz}]f, \tag{3}$$

$$\alpha_f = 247.60[\mathrm{dB}/m/\mathrm{MHz}]f, \tag{4}$$

where f is the frequency. Evaluated R squared values (R^2) are 0.9875 and 0.9867 for Eqs. 3 and 4, respectively.

Various measurements were made to assess the repeatability of the non-collinear wave mixing experiments, including complete

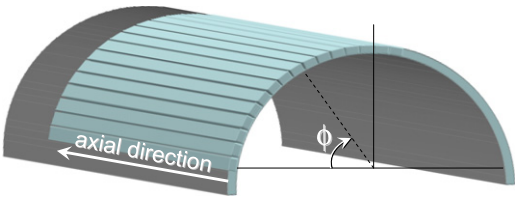

Fig. 2. PVC field test specimens obtained from a pipe segment of 600 mm diameter.

Table 1
Annealing time intervals of the PVC test specimens for non-collinear wave mixing experiments and measured phase velocity and attenuation of longitudinal wave at 2 MHz.

t (h)	3	6	10	14	24	48	96	192	672	1008	1680	2352	2976
c, m/s	2315	2313	2312	2314	2310	2314	2313	2315	2319	2319	2325	2322	2322
α, dB/m	443	455	456	441	452	451	455	457	453	451	443	447	449

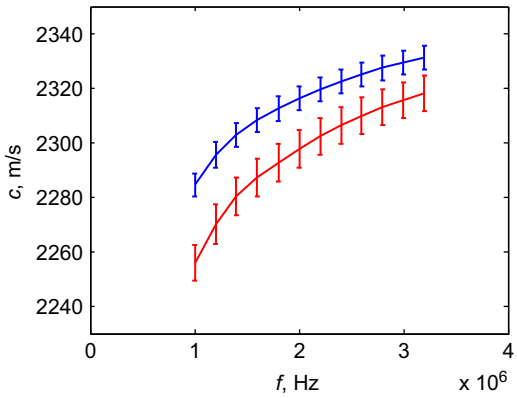

Fig. 3. Averaged phase velocity dispersion curves of longitudinal wave in the laboratory (blue curve) and field (red curve) PVC samples, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

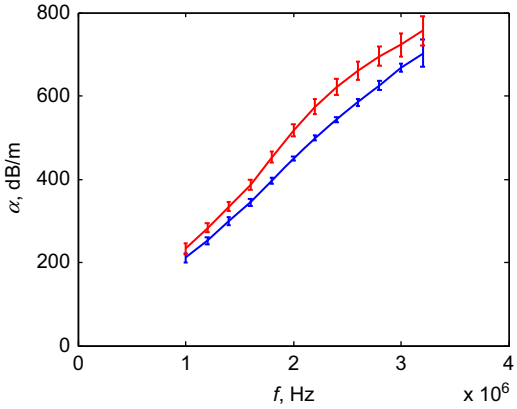

Fig. 4. Averaged attenuation of longitudinal wave in the laboratory (blue curve) and field (red curve) PVC samples, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

disassembly and re-assembly of the measurement set-up. The repeatability was found to be good, with standard deviations of the measured peak-to-peak amplitudes within 1.33%. for measurements on 'the same spot', for both laboratory and field specimens. The measurement results of the laboratory PVC test specimens obtained using the non-collinear wave mixing measurement method are presented in Fig. 5. These results show the relative changes in the peak-to-peak amplitude (see Fig. 6(d)) of the generated wave versus the annealing time of the PVC test specimens, $\Delta A=(A_{pp}-A_{0pp})/A_{0pp}$. The reference amplitude A_{0pp} was measured in the rejuvenated sample 6 h after extraction of the rejuvenated sample from the quenching liquid and A_{pp} was measured in the annealed test specimens. The measured peak-to-peak amplitude A_{0pp} was 41.68 mV. The amplitude of the mixed wave is strongly affected by the annealing time (see Fig. 5). Up to 85% difference in ultrasonic signal amplitudes was obtained between the rejuvenated and strongly physically aged PVC samples. A 30% difference was observed between the slightly (a few hours) and strongly annealed (thousands of hours) PVC samples. Moreover, the measurement results show that the wave mixing technique becomes insensitive or saturated to the physical ageing state of PVC for the long annealing times more than 10^3 h at 60 °C.

Fig. 6(a) shows a raw time-domain ultrasonic signal when PVC test piece of 192 h annealing time was used in the non-collinear wave mixing experiments. Filtered components of two pump waves are presented in Fig. 6(b) and (c). One can see that amplitudes of the pump wave signals are low in a comparison with the non-linear

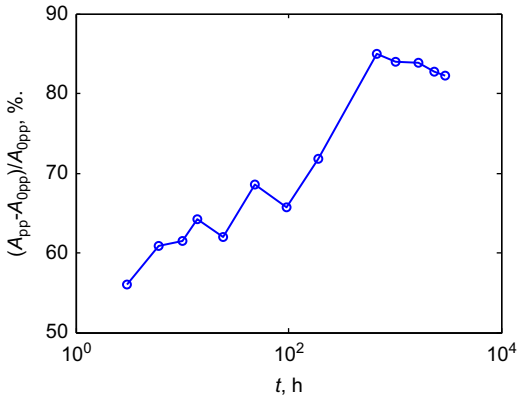

Fig. 5. Dependence of change in peak-to-peak amplitude of the nonlinearly generated wave on the annealing time of PVC laboratory test specimens.

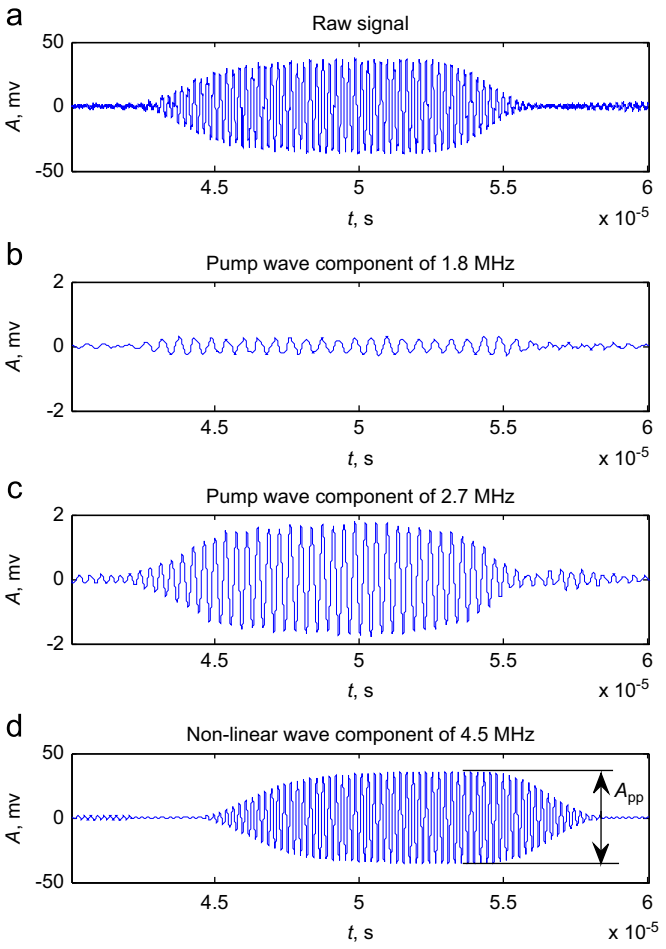

Fig. 6. Raw time-domain signal (a) and its components: pump wave of 1.8 MHz (b), pump wave of 2.7 MHz (c), non-linear wave of 4.5 MHz (d).

wave signal presented in Fig. 6(d). This difference (low amplitude) is caused by the following reasons: (a) the ray-paths of the pump waves do not strike the receiver surface perpendicularly and the central beams deviate from the receiver (see Fig. 1); (b) the sensitivity of the receiver is lower to the 1.8 MHz and 2.7 MHz pump waves than to the 4.5 MHz non-linear wave; (c) an amplification flatness of the preamplifier. A comparison of the signals presented in Fig. 6(a) and (d) shows that the non-linear wave component dominates in the raw ultrasonic signal. It means that the non-collinear measurements can be performed without a signal filtering when two conditions are satisfied: (a) there is a good spatial

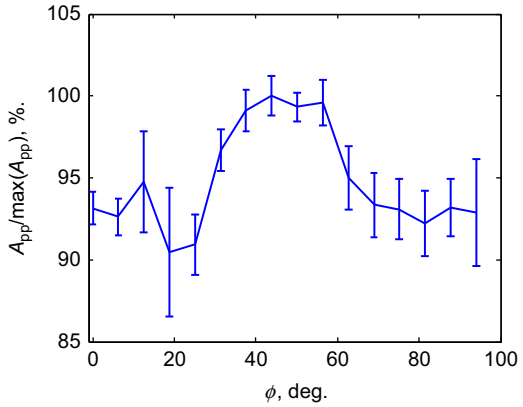

Fig. 7. Variations of peak-to-peak amplitude in the PVC field test samples, where ϕ is the angle in the circumferential direction of the pipe. The error bars indicate the standard deviation per specimen in axial direction. The amplitudes are scaled to the maximum amplitude at $\phi=40^\circ$.

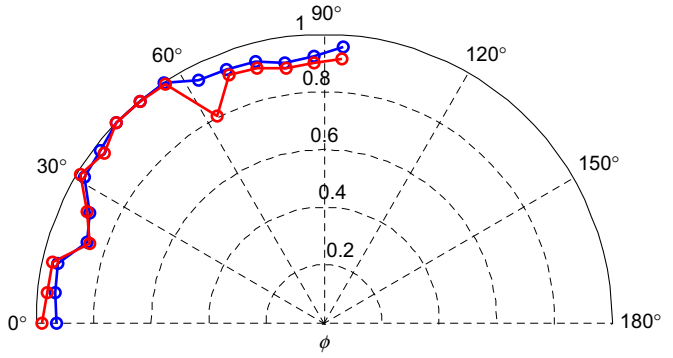

Fig. 8. Distribution of the peak-to-peak amplitude in the circumferential direction of the PVC field test specimens. The blue and red curves denote the normalized measurement results before and after rejuvenation, respectively, each scaled to the maximum amplitude of the series. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

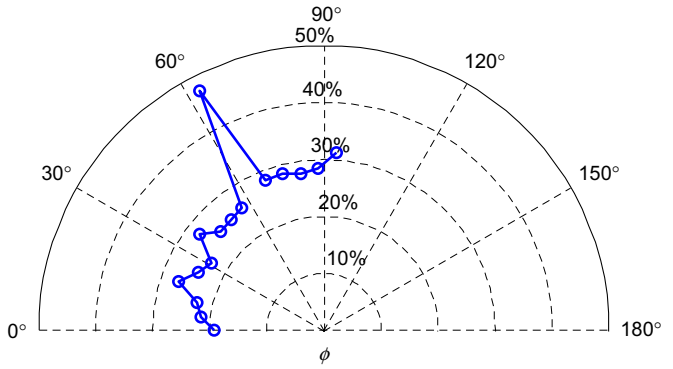

Fig. 9. Relative decrease in peak-to-peak amplitude of the generated wave after rejuvenation of the PVC field test samples.

separation of the informative signal and (b) a non-linear wave component dominates in the informative signal.

The PVC field test specimens were measured initially in the axial direction of the pipe (Fig. 2) to check homogeneity of the elastic properties. The nonlinear measurements were carried out at 14 discrete points with a step of 5 mm in all 16 samples.

Fig. 7 shows the standard deviations of the measured peak-to-peak amplitude (scaled to the maximum amplitude at $\phi=43.87^\circ$) in the axial direction and the distribution of these measurement results in the circumferential direction (Fig. 2). The results show that the maximum standard deviation in the extrusion direction is 3%. The maximum measured standard deviation in the axial direction is significantly smaller than the peak-to-peak amplitude maximum change of 10% in the circumferential direction. The variation in circumferential direction is attributed to thickness variation and weld lines (or 'spiderlines') induced during manufacturing by the extrusion process, which will lead to a variation in morphology and material properties.

The measurement results of the field test specimens before and after rejuvenation are presented in Fig. 8. The results were scaled by the maximum peak-to-peak amplitude of each measurement series separately. The blue and red curves mark the normalized measurement results before and after rejuvenation, respectively. In this case, 5:30 h passed after extraction of the rejuvenated samples from the quenching liquid. Good agreement was found between the repetition and distributions of the measurement results, with one possible outlier at 64°. Effects other than rejuvenation are apparently absent, such as variation in the set-up or specimen geometry.

Although the distributions in Fig. 8 are very similar, the absolute values of the amplitudes of the response decrease strongly due to rejuvenation. The relative change of amplitude, $\Delta A=(A_{pp}-A_{0pp})/A_{0pp}\times 100\%$, in the PVC field test specimens is presented in Fig. 9, again as a function of the circumferential position. The measurement results show that this relative change is in the range of 20–30% for most of the test specimens. The measurements confirm that the PVC pipe was physically aged during its prior service in water supply.

Previously it was shown that hardness measurement results correlate with the yield strength observed in tensile testing [26]. Therefore the non-collinear wave mixing results were verified partially employing micro-indentation experiments. The hardness of a laboratory specimen of the longest annealing time was measured. Sequentially, the PVC test specimen was rejuvenated and its hardness measured. A 30% difference was obtained between the measured hardness in the sample. A similar experiment was repeated with a field test sample of PVC. In this case a 5% difference in the hardness of the sample was estimated. These changes are proportional to the measurement results obtained by non-collinear wave mixing experiments when 85% and 20–30% were estimated in the laboratory and field test specimens, respectively (see Table 2). A cross-correlation coefficient between the ultrasonic and hardness measurement results varies in the range of 60–70% (depending on the measurement position) when the measurements are carried out in the unprocessed field test samples. The experimental procedure and results of the micro-indentation experiments will be reported more in detail in the near future.

Table 2
Measured changes in the PVC specimens.

	Non-collinear ultrasonics	Hardness
Laboratory	85%	30%
Field	20–30%	5%

6. Conclusions

Linear ultrasonic measurements and non-collinear wave mixing ultrasonic measurements have been performed for contactless measurement of the state of physical ageing of PVC. The measurements of longitudinal wave phase velocity dispersion and attenuation can be used to identify differences between linear properties of

plastics of the same polymer, but these measurements are insensitive to the physical ageing state of PVC (see Table 1).

The non-collinear ultrasonic wave mixing process $S(\omega_1)+L(\omega_2)\rightarrow L(\omega_1+\omega_2)$ does lead to significant sensitivity to physical ageing. The suggested method has been verified experimentally on two different groups of PVC test specimens: laboratory and field. It was shown that the ultrasonic signal amplitudes can change up to 85% from rejuvenated to artificially aged PVC test specimens in the laboratory test specimens. A difference up to 30% was obtained measuring the field test samples.

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