ACOUSTIC EMISSION DURING STRUCTURE CHANGES IN SEMI-CRYSTALLINE POLYMERS

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Abstract

Tensile tests of polypropylene and its blends with polyethylene and other additives have been recorded by digital camcorder. Acoustic emission (AE) was monitored during tests by two transducers attached to test samples, allowing linear location of AE sources. Tested materials exhibit large localized plastic deformation or necking, reaching hundreds of percent before failure. Image analysis procedure based on displacement of contrast markers on the surface has been used to quantitatively evaluate axial and lateral strains and strain rates along the sample. Correlation of AE activity with a set of plastic deformation, neck formation and defect development enables us to evaluate influence of additives on the whole stretching process.

Keywords: Acoustic emission, strain visualization, polymer drawing and necking

1. Introduction

The behavior of polymer materials beyond their elasticity limit is important for many technological operations, such as a production of reinforced fibers, sheets, plates and so on [1]. A plastic deformation of semi-crystalline thermoplastics is characterized by the creation and development of a neck. In such cases, the localized plastic deformation can reach hundreds or thousands of percents. The mechanisms of plastic or visco-plastic deformation of these polymers are strongly dependent on supra-molecular structure (content of the amorphous phase, the size and configuration of crystallites, etc.). The attention of plastic processing industry is in recent years focused on cost reduction and improvements of mechanical properties of plastics. It can be achieved by the usage of the controlled co-polymerization or various blending. Typical example of the newly developed material is the blend of polypropylene (PP) and polyethylene (PE). Both materials have different molecular and supra-molecular structure and as a result they also have different mechanical behavior. The main task is how to interconnect these two structures so that the resulting blend should have the best mechanical properties (the problem of blend compatibilization). Special compatibilizers have been developed for this purpose (copolymer additives). In this study, e.g., we used a modifier, VISTALON, with variable content of ethylene units.

The influence of compatibilizers or other additives (rubber particles, etc) is relatively small in the range of elastic (or linear visco-elastic) deformation. But it becomes important when the plastic deformation becomes large near the neck formation and with neck elongation, as well as in the case of defect (crack) initiation and growth. The creation and development of a localized plastic deformation under different strain rates can be well documented using video-recording of a sample during the standard tension test. Then it is possible to analyze the distribution of longitudinal and transverse deformation of the specimen in each moment of the video image record (it enables to localize the place of a neck formation and its continuous elongation). However, the
video-recording gives no local information about micro-mechanisms and structural defects related to the neck formation and elongation. Such information may be obtained by the acoustic emission (AE) method [2].

2. Tension Tests of Polymer Blends

Polymer specimens for standard tension test were prepared; a set was made of pure isotactic PP and further sets of PP and PE (HDPE) blends with various additives and modifiers. During tension tests under different strain rates (5 to 200 mm/min.), AE signals were detected by two sensors placed at the specimen ends. The goal was to monitor changes of plastic deformation mechanism and beginning of damage and fracture, dependent on the composition of the material and the strain rate. Many tests were performed with the following specimens: pure PP, PP+PE (50+50%) blend without compatibilizer, blend of PP+PE (47+47%) + 6% compatibilizer VISTALON 407 (i.e. copolymer PP+PE with 40% of ethylene units), blend PPR (PP+22.5% modifier ROYALEN), blend PPK (PP+22.5% amorphous rubber KELTON) and blend PPB (PP+22.5% semi-crystalline rubber BUNA).

Some experimental and procedural problems were encountered during the first tests: The first question concerned with the reproducibility of acoustic coupling of broadband AE sensors placed on a glassy surface of the specimen subjected to large plastic elongation and neck development. Next problem was related to the linear localization of AE sources during the continuous increase of the distance between both sensors. Also a relatively high attenuation of tested polymer samples (more than 1 dB/cm) and its growth during the test causes AE detection and evaluation difficult. The detailed analysis of material structure changes requires correlation of AE sources with the localized plastic deformation. Considering inhomogeneous distribution of plastic deformation along the sample (neck formation and elongation), and the fact that the total elongation of the specimens to the fracture can achieve tens to hundreds of mm (deformation is hundreds of %), it is evident that only a non-contact (optical) strain measurement method can be used. Equidistant marks were drawn on the specimen surface, and their changes were scanned by digital camcorder. Camera also scanned the position of the AE sensors. So the instantaneous distance between sensors, necessary for successful AE source location, was obtained from video-records. The video-record is synchronized with AE registration by the time code and also (for the fast check) by synchronous video-recording of the AE-analyzer audio-monitor output. Due to the high material attenuation and relatively noiseless deformation mechanisms, the AE analyzer had to be set on high total amplification (of about 85 dB). Also the threshold level for AE counting was set just above the background noise.

The experimental setup used in tension tests is shown in Fig. 1. Specimen is loaded using the computer-controlled electromechanical tensile testing machine TIRATEST 2300 with attached dynamometer of 10 kN capacity. Signals from AE transducers were parallel-processed by two 2-channel AE analyzers: The analog D/E 3000 analyzer served as a "stereo" audio-monitor and linear locator. Signal waveforms were recorded by the digital analyzer DAKEL-XEDO equipped with signal processors. It allows registration of global parameters, as well as digital recording and frequency analysis of sampled AE events. Low-frequency audio-monitor output was recorded by the digital camera (48 kHz/16 bit) at the same time with movies. After the test, the video and sound data from the camera are transferred through the interface IEEE1394 (firewire) into a PC. The changes of distances between calibration marks (initial spacing was 5 mm), changes of specimen widths (lateral deformation) and also changes of distance between AE sensors during the whole test are then analyzed by the special software designed for this purpose.
Analyzed details of specimen evolution during the test are shown in Fig. 2 (snapshots by steps of 200 s). Video-recording is synchronized with presence of emissive event detected by AE sensors with the accuracy of 40 ms. It makes it possible to correlate diagrams of heterogeneously distributed instantaneous specimen deformation (strain rates) with that of AE event occurrence.

Fig. 1. Schematic drawing of experimental setup used in tension tests of polymer samples.

Fig. 2. Samples of videorecords during test of pure PP; the creation and elongation of the neck (5 mm spaced markers are connected by white lines and neck ends by black lines).
3. Evaluation of Video-Recordings

We used a digital camcorder that scans 25 frames/s, with resolution 720 x 576 pixels, and color depth 24 bit. Video and sound information are encoded using camera firmware (codec DV video). Resulting data flow recorded by the camera (DIGITAL-8 format) amounts to 3.6 MB/s. After the test, all data are transferred to a PC hard disk. One-minute recording matches AVI file with size of 216 MB. Total time of tension test until fracture of a specimen was as much as 30 minutes, which results in AVI-file size of about 7 GB. With respect to the main research objectives (deformation behavior of tested material, continuous plastic deformation and neck formation) the information content is inefficiently huge. Only selected frames were analyzed from video files (50 to 500 frames at equidistant time intervals of 3 to 30 s). Distribution of longitudinal and lateral strain along the sample was automatically analyzed in selected frames, showing the neck formation. At very high elongation rates (200 mm/min.) the test duration is only a few s, hence all saved frames were analyzed.

Algorithm used for automatic evaluation of deformation characteristics from video-records must take into account various problems associated with image recognition:

a) Non-uniform scene illumination during the test (designed algorithm was sufficiently robust to eliminate that influence, so the contrast and brightness corrections were not necessary)

b) Analyzed scene is distorted due to the short focal distance. Quite sufficient solution was based on a simple linear transform (rhomboid transformed to rectangle).

c) During the sample stretching it is difficult to separate specimen from surroundings, and moreover, both sample heads are moving (constant position have only static parts, e.g. upper clamp). The problem is solved by suitable editing of the first and last frames (easily detectable crosses are added to the last markings on a sample). The relative movement of enlarged sample heads is influenced only by pure elastic deformation proportional to the strain rate and thereby the sample position in all other scenes can be well localized.

d) The markers drawn on the specimen change not only their lengths (lateral contraction), but their widths (differently in various specimen parts). Variable width of markers disallows the use of classical line detection algorithm, and it must be replaced by the edge detection (left and right edges are detected and the line position is identified as their arithmetic mean). The edge detection is performed by the convolution of analyzed scene with a suitable mask.

e) The light source is not placed in the camera axis, which results in shadowing of thickness changes (false lines has sometimes more contrast than detected lines). Presence of false lines requires to detect real edges only around their expected position. The prediction is realized using the edge position in previous scene and knowledge of exact strain rate.

Designed automatic algorithm consists of following steps:

1. Detection of the auxiliary crosses in the first and the last analyzed frame (i = 1, N).
2. Detection and localization of the left and right edges in the first frame.
3. For i = 2 to N:
   Detection of the left and right edges in frame (i) using the knowledge about detected edges in figure (i-1). Determination of marker positions in frame (i).
4. For i = 1 to N:
   Detection of top and bottom sample edges at places of detected markers in frame (i). Determination of specimens width in places of detected markers.
The accuracy of evaluated strain distribution depends primarily on a pixel resolution of camera used. Further errors are caused by the algorithm of video-recording compression, and by the precision of drawn markings.

4. Discussion of Results

Results of AE analysis revealed that AE activity during the heterogeneous plastic deformation is predominantly concentrated in areas of necking (neck boundaries). After the further neck drawing, especially if another deformation mechanisms start to develop, the other areas become also active. Intensity of AE increases rapidly when defects are formed in plastically exhausted zones. Development of crack-like defects lead to specimen rupture. The typical AE activity distribution along the sample of PP+PE blend is illustrated in Fig. 3a, where the AE activity is characterized by the number of events Ne (3 decades in logarithmic scale) emitted between two subsequent time snapshots taken at 30-s interval. The variation of loading rate from 0.1 to 50 mm/min has no substantial influence on the test history.

Fig. 3. AE activity during the test of blend PP (50%) + PE (50%).
a. Number of AE events Ne (log. scale) distributed along the sample snapshots.
Fig. 3. AE activity during the test of blend PP (50%) + PE (50%). b. (top) Loading force vs. time. c. (middle) AE activity expressed by RMS values of both AE channels, A and B (upper and middle curves), and RMS recorded by camcorder (channel A). d. (bottom) Number of counts Nc1 in channel A.

Samples made of pure PP exhibited very low AE activity, with the maximum in transition to plastic deformation, when the neck with sharp boundaries begins to form. When the neck elongates, the AE activity is rapidly reduced. It re-appears again just before the specimens rupture, when a fibrous structure forms and breaks (axial fibers are drawn out). If the displacement rate is higher than 100 mm/min, the deformation behavior of the PP specimen becomes completely different, and specimen breaks in a quasi-brittle manner.
Fig. 4. Test of the blend PP (77.5%) + additive ROYALEN (22.5%)
a. Time dependence of the loading force F.
b. AE count rate dNc1/dt.
c. Time development of axial distribution of AE events Ne.
d. Strain rates $d\varepsilon/dt$ vs. $t$ and $x$. Distributions along the sample axis $x$ are related to the initial markers spaced by 5 mm (sectors).
Blends made of PPR, PPK and PPB exhibited similar deformation behavior, but the formed neck had no sharp boundaries. They are also characterized by the relatively small AE activity as it is illustrated in Fig. 4.

![Graph](image1)

Fig. 5. Test of the pure blend PP (50%) + PE (50%) (for other characteristics, see Fig. 3). Top: Time development of axial distributions of AE events Ne. Bottom: Time development of strain rate $\frac{d\varepsilon}{dt}$ distribution along sample axis. Distributions are related to initial markers spaced by 5 mm (sectors).

Quite dissimilar AE activity was detected for PP+PE blends. Already initial tests indicated differences in deformation behavior of blends with added compatibilizers. Test examples are shown in Fig. 5 (PP+PE without additives, see also Fig. 3) and in Fig. 6 (mixture with 6% of compatibilizer VISTALON 407).
Fig. 6. Test of blend PP (47.5%) + PE (47.5%) + VISTALON (5%). Top: Time dependence of the loading force F. Middle: AE count rate dNc1/dt. Middle-lower: Time development of axial distributions of AE events Ne. Bottom: Strain rates dε/dt, vs. t and x. Distributions are related to initial markers spaced by 5 mm (sectors).
Detailed analysis of considerably different AE activities at distinct periods of drawing process can help explain structural mechanism of compatibilizer effect. AE activity of samples without modifier markedly increases at the beginning of neck formation without sharp boundaries. This activity remains very high until the specimens rupture, and can be compared with AE of polymer composites. To explain AE activity, we must consider that PP and PE have different crystallite sizes and the structure with different crosslinks (tie molecules). In a blend, there are two different networks mechanically joined together, which must be rearranged and uncoupled during their deep drawing. In contrast to previous pure polymer, the blend PP+PE with added compatibilizer VISTALON 407, which is completely amorphous copolymer, had in the first deformation phase the behavior similar to the pure blend. When the neck started to draw, the AE activity became very low, similar to the pure PP. It may be caused by the presence of compatibilizer, which allows better sliding of networks through the amorphous phase. Nevertheless, after reaching a certain drawing limit, the AE suddenly starts to grow to the level of the blend without modifier. This relatively complicated AE behavior reflects very sensitively changes of structural mechanism during various deformation periods in the materials with complicated molecular network structure. Synchronous video-recording with the AE monitoring shows the instantaneous correlation of the heterogeneously distributed plastic strain rates with the occurrence of active AE sources.

Recorded AE signals were also subjected to frequency analysis and their evaluated parameters in time and frequency domain were used as input data for the source type classification. However, no sharply different classes have been recognized. It is probably caused by many geometrical factors affecting the way of elastic wave propagation:

a) Dimensions of the active part of test specimen (100 x 10 x 4 mm, initially) are comparable to the length of detected elastic waves spreading from AE source to a sensor (2 to 20 mm in hundreds kHz frequency range). It results in strong wave dispersion and guided waves are mainly detected, which substantially modify the spectral and other characteristics of received signal (filtration effects) and partially implicate loss of information about the sources.

b) Geometrical form and dispersion behavior of the specimen pass through considerable changes during the course of very large deformations (hundreds of percent). Moreover, the distance between AE sensors changes. It complicates the attempt of getting precise linear location of AE sources.

c) Tested polymer materials exhibit relatively high attenuation in the used AE frequency range (100 kHz to 1 MHz). Only the AE events with higher amplitudes were detected by both transducers. This allowed localization of only about 30% of all registered events. To obtain correct interpretation of structural changes, it is therefore necessary to evaluate global AE characteristics in both channels separately, and make corrections to them for the attenuation changes.

5. Conclusions

Deformation behavior of semi-crystalline polymer materials, PP and PE, and their blends with various additives, was investigated by means of video-recording and AE monitoring during the tensile tests. The video-records were used to evaluate the localized plastic deformation distributed along the sample. Comparison of plastic zone expansion (necking) and spatial distribution of AE events shows that primary sources of AE are places, where the plastic necking starts to develop, areas of neck drawing, and finally locations where neck stopped its elongation and crack defects are created. It has been detected that deformation and AE behavior of pure PP and PP with small addition of rubber-like fillers (very low AE activity) markedly differs from the
behavior of polymer blends of PP with PE, whose plastic deformation is accompanied by very high AE activity. Tests also helped to clarify influence of blending compatibilizers on a plastic zone development: Compatibilizers prolong the first stage of plastic deformation with low AE activity (comparable with the pure PP), but after reaching some deformation limit, their AE behavior is similar to blends without additives.

Using the low-cost digital camera for video-recording of mechanical tests together with proper evaluation algorithms allows detailed analysis of AE source distribution connected with structural changes during the localized plastic deformation.

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References