A heatable stretching device for dynamic X-ray studies

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A Heatable Stretching Device for Dynamic X-ray Studies

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Abstract

In this paper a heatable stretching device is presented which enables the performance of real-time X-ray experiments during deformation. The construction of the oven makes it possible to perform experiments with detectable X-ray diffraction angles (2θ) of up to 30°. It is shown that the apparatus can be used to study morphological changes occurring during the deformation/drawing of polymer materials, using the benefits of synchrotron facilities. A series of small-angle (SAXS) and wide-angle (WAXS) X-ray scattering patterns obtained *during* the drawing of melt-crystallized ultra-high-molecular-weight polyethylene (UHMW-PE) at 373 K is presented.

1. Introduction

In order to gain insight into the molecular basis of the mechanical properties resulting from uniaxial drawing of polymer materials, extensive X-ray studies have been performed in the past. In most cases, however, the samples were drawn under conditions quite different from the measuring conditions (*i.e.* applied drawing temperature and drawing force), possibly giving rise to certain artefacts. Such artefacts can be excluded by performing X-ray studies *during* the deformation process.

Up to now most dynamic measurements *during* a stretching process have been performed whilst drawing a polymer at room temperature (*e.g.* Koch, Bordas, Schöla & Broecker, 1979; Holland-Moritz & van Werden, 1981; Stach, 1986). The first X-ray results (*i.e.* SAXS) of dynamic studies *during* stretching at elevated temperature were recently published by Fronk (1984) and by Reck, Schenk & Wilke (1985).

As far as we know, no devices have been described in the literature which enable successfully reproducible performance of real-time WAXS *and* SAXS drawing studies at elevated temperatures. This paper describes a heatable stretching device of a rather straightforward design which has been designed and built to enable synchronous monitoring of stress, clamp displacement, oven temperature and SAXS or WAXS patterns *during* a stretching process at elevated temperatures.

Table 1. *Technical data of the stretching device described in the text*

	Minimum	Maximum
Clamp displacement (mm)	0	480
Clamp velocity (mm s^{-1})	0·09	86
Drawing velocity (s^{-1})	0·00016	0·86
Sample length (mm)	100	<580
Sample thickness (mm)	—	0·5
Drawing temperature (K)	room temp.	<623
Detectable 2θ region (°)	0	30

2. Experimental arrangement and apparatus

In Fig. 1(*a*) an overview of the stretching device is given. To avoid serious problems with sample fracture at the clamps, the clamps (*A*) are positioned outside the oven (*B*). The device is constructed in such a way that the displacement of the clamps is always symmetrical with respect to the oven, enabling the study of samples with necking behaviour in a well defined manner. For the limitations and possibilities with respect to sample length, clamp displacement velocity and drawing velocity the reader is referred to Table 1.

The applied load during the stretching can be measured by a Gould UC3 stress transducer (*C*), whose output voltage is proportional to the applied force. The stress transducer is mounted in series with one of the clamps in such a way that force measurements are not influenced by any friction of the apparatus itself. If necessary, it is possible to determine the relationship between engineering stress and the actual draw ratio by using calibration curves as will be described in a following section. Such stress–strain relations are frequently used to characterize the drawing behaviour of polymers.

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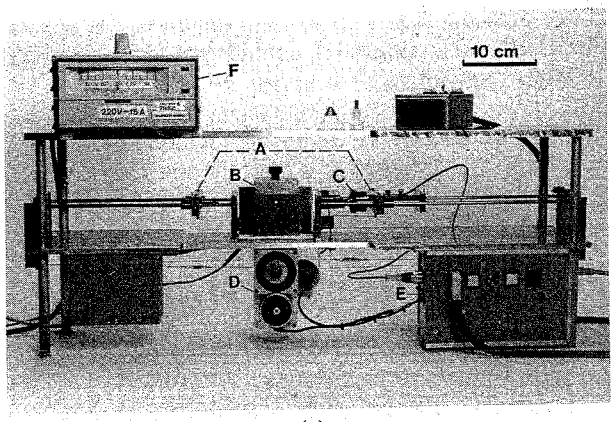
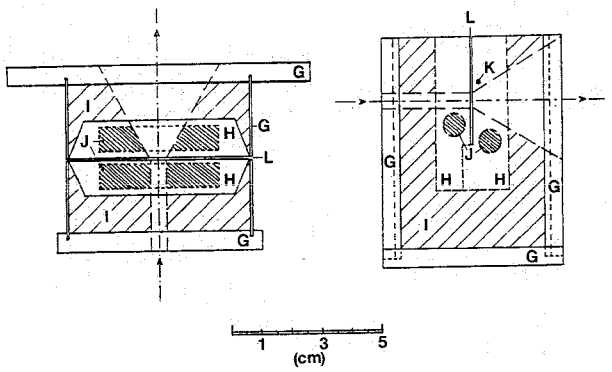


(*a*)



(*b*)

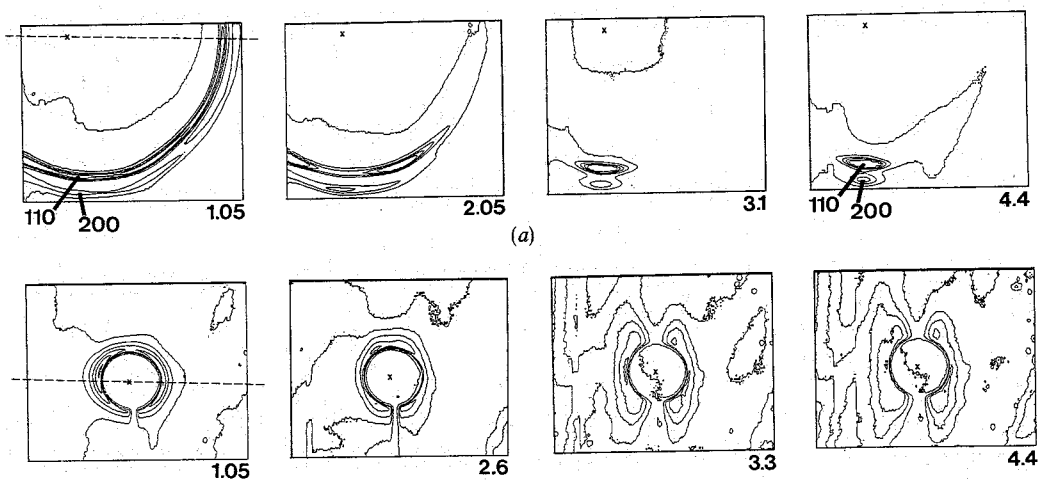
Fig. 1. (*a*) Stretching apparatus, used for real-time X-ray drawing studies at elevated temperatures and (*b*) schematic drawing of top view (left) and side view (right) of the oven used. The arrows indicate the direction of the primary beam. *A*: sample clamps; *B*: oven; *C*: stress transducer; *D*: motor + gearbox; *E*: stretching control unit; *F*: temperature control unit; *G*: aluminium; *H*: brass; *I*: fiberfrax; *J*: Heizpatronen; *K*: chromel–alumel thermocouple; *L*: space for sample.

To enable deformation studies *via* WAXS and SAXS measurements, circular holes are introduced in the oven. Disturbing X-ray scattering effects are avoided by adjusting the holes to the beam size of the X-ray source used. The diameter of the cylindrical hole for the incoming beam is chosen to be 5 mm. For SAXS experiments the hole for the diffracted beam is equivalent to this. For the WAXS studies, however, a conical hole is required for the diffracted beam, to allow measurements of reflections with 2θ up to 30° (see Fig. 1*b*).

The oven can be heated by two HPS-Hochleistung-Heizpatronen devices of 220V/100W each (*J*), installed as close to the holes as possible. The temperature is controlled by a chromel–alumel thermocouple (*K*) combined with a Eurotherm (*F*). To secure good heat conduction between the different metal parts of the oven a heat-transfer agent (WATLUBE) was used. Test experiments revealed only a slight temperature drop (less than 3 K) of the samples close to the oven holes, small enough to avoid inhomogeneity in drawing.

Many polymer materials exhibit necking during drawing, leading to inhomogeneous drawing. A reliable way to relate the draw ratio to the clamp displacement is to use an empirically determined calibration curve, which can be obtained by drawing a sample stepwise and measuring the actual draw ratio with ink marks. To be sure that neck formation starts in the symmetry centre of the oven, a V-shaped mark is cut in the sample (the origin of the neck). Test runs with ink-marked samples were satisfactory.

It should be noted here that the device presented can easily be accommodated in a Fourier transform infrared spectrometer or in a small-angle light scattering (SALS) set-up for real-time drawing studies.



(*a*)

(*b*)

Fig. 2. Series of isointensity contour plots of real-time WAXS (*a*) and SAXS (*b*) patterns, obtained by drawing melt-crystallized UHMW-PE. The contour lines were drawn at equal intensity intervals. The meridian and the drawing direction are denoted by a dashed line. × denotes the centre of the primary beam. The corresponding draw ratios are indicated in the lower right corner of the patterns.

3. Application to melt-crystallized UHMW-PE

The performance of a real-time X-ray study necessitates a drastic reduction of the X-ray exposure time, which can be accomplished by using a synchrotron source in combination with a two-dimensional position-sensitive X-ray detector.

As an example a small series of real-time WAXS and SAXS patterns are presented in Fig. 2, recorded *during* a drawing experiment of melt-crystallized UHMW-PE tapes (Hostalen-GUR 412, $M_r \simeq 1700$ kg mol^{-1}). The samples were drawn at 373 K with a drawing velocity of 0·004 s^{-1}. The X-ray patterns were obtained using the synchrotron facilities at DESY, Hamburg. The X-rays were generated at 5·260 GeV and 25–30 mA and monochromatized to a wavelength of 1·61 Å. Combination of the use of the intense synchrotron X-ray source and a Westinghouse Vidicon detector made it possible to record a WAXS or SAXS pattern every 10 s. The distance between the sample and the detector was chosen to be about 9 and 205 cm for the WAXS and SAXS experiments respectively. The resulting contour plots were obtained straightforwardly without any background correction.

The observed WAXS and SAXS patterns can be understood in terms of the deformation model, as proposed by Peterlin (1965, 1971). In a forthcoming paper results obtained by conventional and real-time X-ray drawing studies of melt-crystallized PE will be discussed and compared in detail (van Aerle & Braam, 1988).

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