About the Lorentz Correction Used in Interpretation of Small-Angle X-ray Scattering Data of Semicrystalline Polymers

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ABSTRACT: Lorentz correction is used to correct the intensities of X-ray scattering of single-crystal diffractometry in order to recalculate intensities to obtain structure factors. This correction reduces the intensities to zero at zero diffraction angle. Small-angle scattering is used to study the dimensions of heterogeneities in polymeric materials. The scattering intensities at a near to zero scattering angle originate partly from periodic systems (reciprocal lattice) and partly from dispersed particle systems. Periodic systems should result in individual Gaussian or Lorentzian peaks with the position of a peak maximum depending on the length of the periodicity. Particle scattering results in a Gaussian peak centered at zero scattering angle. The effect of the Lorentz correction on the interpretation of small-angle X-ray scattering data is shown for some semicrystalline polyethylenes (high-density, linear low-density, and low-molecular-weight waxy polyethylenes). The data are compared to those for amorphous block copolymers (styrene–butadiene), in which there is a periodic system with homogeneous lamellar thickness. Lorentz correction destroys the characteristics of the particle scattering and can be applied only for periodic systems. It should not be used to produce a peak on scattering data, which do not show periodicity (peaks) without correction.

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INTRODUCTION

The structures of semicrystalline polymers, including polyethylenes of different types, are supposed to consist of periodically arranged crystalline lamellae, which are separated by amorphous phases. The layer thicknesses of the crystalline and amorphous layers are fairly homogeneous.[1–2] The length of the periodicity (d) is calculated from the position of the peak maximum in the X-ray intensities using the Bragg equation

$$d = \frac{\lambda}{2\,\sin(\Theta)} \qquad (1)$$

where Θ is half the diffraction angle of the peak maximum and λ is the wavelength of the X-ray.

This formula is applied to calculate the length of periodic systems, that is, systems that can be described by a reciprocal lattice. The lamellar thickness of the crystals is generally calculated from the Bragg periodicity and modified by the percent crystallinity of the system, determined

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from either wide-angle X-ray diffraction (WAXS), DSC heat-flow data, or density measurements.[3]

Small-angle X-ray scattering (SAXS) is also used to determine particle size and its distribution in systems where particles (or a second phase in a broader sense) are dispersed in a continuous phase called the matrix.[4–5] This is called particle scattering.

For these types of systems, the following conditions necessary to use SAXS:

a. the electron densities of the particle and the matrix must be different;
b. the electron density of the matrix must be constant at larger distances from the particles;
c. the structure must be statistically isotropic; and
d. there should be no long-range order, so no correlation will exist between two points separated widely enough.

These conditions are generally fulfilled in a diluted solution of macromolecules but generally not in semicrystalline polymeric bulk. There is a 10–15% difference in the electron densities of the amorphous and the crystalline phase, so condition a is generally fulfilled. As the crystallinity varies from 10% to 60% in most systems used for this study, conditions b and d are not fulfilled: the dimension and the volume of the amorphous phase are commensurable to those of the crystalline one. If the crystals form folded-chain lamellae with a fairly homogeneous thickness,[1] condition c is also not fulfilled, as lamellae with extended lateral dimensions with respect to their thickness cannot be packed isotropically. There must be a local order of the lamellar particles as well as of the amorphous environment around the particles.

The scattering intensities of a particle system are the Fourier transform of the electron densities that reduce for the conditions shown above to the form described by eq. (2):

$$I(s) = \int 4\pi r^2 \, dr \bar{\rho}^2(r) \, \frac{\sin(sr)}{sr} \tag{2}$$

where r represents the position vector in the real space and s represents that of the reciprocal space. This latter term is expressed by eq. (3):

$$s = \frac{4\pi}{\lambda} \sin(\Theta) \tag{3}$$

The solution of eq. (2) in the case of spheres with radius r is given by eq. (4)[6]:

$$I(s) = V * (\rho_p - \rho_m)^2 * 3 * \left(\frac{\sin(sr) - sr * \cos(sr)}{(sr)^3} \right)^2 \tag{4}$$

where ρ_p and ρ_m are the electron densities of the particle and of the matrix, respectively; and V is the volume of the particles. This function approximately forms a Gaussian peak centered at $s = 0$.

Guinier[6] presented an approximation to describe the scattering intensity near to the zero-scattering angle using the radius of gyration of the particle system shown by eq. (5):

$$I = I_0 \exp(-KR^2\Theta^2) \tag{5}$$

where I_0 represents the scattering intensity at zero angle, R is the radius of gyration of the particle, Θ is half of the scattering angle, and K is a universal constant ($16\pi^2/3\lambda^2$).

It can be seen in eqs. (2), (4), and (5) that X-ray scattering of particles has a maximum at zero angle, with an approximately Gaussian shape of the scattering toward greater angles. This means X-ray scattering for particles has nonzero values at the zero diffraction angle. The log I versus square of the scattering vector s (the Guinier plot) results in a straight line with a slope corresponding to the Gaussian scattering curve. This slope can be used to determine the dimension (radius of gyration) of the scattering particle.

There is another relationship used in SAXS data processing, the so-called invariant (Q),[5] which is expressed by eq. (6):

$$Q = \int_{r=0}^{\infty} I(s) s^2 \, ds \tag{6}$$

The invariant is expressed as an integral of the intensities multiplied by the square of the scattering vector. The importance of the invariant is its expression of factor by which the structural data can be reduced to an absolute scale without having to measure the absolute intensities. The invariant represents the scattering power of the particles with respect to the matrix and their concentration as it is expressed by eq. (7):

$$Q \approx (\rho_p - \rho_m)^2 * \Phi_p * (1 - \Phi_p) \tag{7}$$

where ρ_p and ρ_m are the electron densities for the particles and for the matrix, respectively, and Φ_p is the volume fraction of the particles. It is stated in eq. (7) that the integral of the corrected intensities is proportional to the square of the difference in the electron densities of both the matrix and the particles as well as to their volume. In the case of semicrystalline polymers, this function has a maximum at 50% of crystallinity. In a number of investigations,[7–9] the invariant was calculated and used for representing the actual crystallinity of the polymer as a function of the temperature.

Another approach to a real polymeric structure is a fractal type.[10] In this case the intensity function is reciprocally proportional to the tth power of the scattering vector, s, as it is expressed by eq. (8).

$$I(s) = I(0) * s^{-t} \tag{8}$$

where t is the fractal coefficient. In this case the intensity at zero angle also has a finite value.

Lorentz Correction

The Lorentz correction means a multiplication of the scattering intensities by a factor proportional to the sine of the diffraction angle. This correction is generally used in single-crystal X-ray crystallography to correct the X-ray intensities obtained from crystalline materials in order to calculate correct structure factor values independent of the geometry of the data collection (see the second equation provided by Kavesh and Schultz[11]). It corrects the differences in the time that the individual reciprocal lattice point, with different indices and consequently at different distance from the origin ([000]), spends on the surface of the Ewald sphere, that is, it is in a diffracting position. It is applied only to scattering from the reciprocal lattices where there is a periodicity with a great number of consecutive cells in the crystalline systems. This correction eliminates the intensity at zero angle, as [000] is always in a diffracting position. The factor contains a sin (2Θ) element and other elements that are dependent on the geometry of the data collection, that is, the intensity measurement.

Recently much of the work dealing with the SAXS of semicrystalline materials has used the so-called Lorentz correction[3,7–9,12–16] in trying to determine the periodicity of the structure. Those who use this correction generally use the squared form of sin 2Θ, instead of a multiplication factor of it. This who use this correction use it in most cases, even with no peak on the uncorrected SAXS intensity curve. Thus, there is no indication that a reciprocal lattice would be present in the system. Some other authors do not use this correction.[10,17–18]

As can be seen above, with particle scattering the maximum of scattering intensities is at zero scattering angles. If the Lorentz correction is applied, the nearly Gaussian shape of the scattering intensities is destroyed, reducing the intensities to zero at zero diffraction angle, and an artificial maximum might be produced on the curve. The position of this maximum depends on the dimension and the volume of the phases as well as on the difference in the electron densities of the particles and the matrix.

The scattering curve of a real system is always the sum of the scattering intensities of the different phases and the interactions between the phases. In a periodic lamellar system a reciprocal axis is present and therefore produces a peak (or series of peaks) in the scattering curves. Their exact position might be determined from the Lorentz-corrected intensities. If there are also particles or fractals in the system, their characteristic scattering will be superimposed on the scattering caused by the reciprocal lattice, and a Lorentz correction will destroy the shape of the curve. In this case it cannot be used for structural calculations. The sum of the intensities originating from the particle scattering might shift the position of the peaks derived from the reciprocal lattice, and their position will also be incorrect. Therefore, the two scattering types must be separated from each other and handled individually, as Dlugosz et al. suggested[12] when they introduced the Lorentz correction and used it for obtaining the exact peak position of a Bragg peak near to the zero-diffraction angle.

Polyethylene (PE) has always been a subject of SAXS investigations. PE has a broad maximum in SAXS intensities.[1,8,16,19,20] It has been claimed that this maximum represents the repeating period of the lamellar structure of the crystallites.[1] In some previous and only partially published[21] works on the SAXS of semicrystalline polymers, the SAXS curves of many polyethylenes (PE)—high-density polyethylene (HDPE), linear low-density polyethylene (LLDPE), and low-density polyethylene (LDPE), as well as polypropylene (PP) and its blends—did not show definite maxima to indicate the dominance, or even the presence, of a reciprocal lattice type of structure. Therefore, the Lorentz correction was abandoned, and the system was handled as particles dispersed in a matrix or as a fractal. In this article the SAXS intensities of some polyethylenes are presented as examples to show the effect of the Lorentz correction on the structural data derived from the original experiments. The same data are shown in two interpretations—as a reciprocal lattice and as dispersed particles—to enable a discussion of the effect of the Lorentz correction on possible interpretations of the data. Hypothetical models of polymeric crystals were disregarded in the interest of this study. WAXS data are also shown in order to properly characterize the semicrystalline polymeric systems. And a comparison also presented are SAXS data on block copolymers with polystyrene and polybutadiene blocks of a narrow molecular-weight distribution.

EXPERIMENTAL

Materials

The following materials were used:

- HDPE,extrusion-grade polymer—MFI: 1 g/min.
- C6–LLDPE,gas-phase-polymerized LLDPE with hexene comonomer—$\rho = 922$ kg/m^3, MFI: 0.78 g/10 min.
- PE–wax, oligoethylene extracted from LLDPE—$M_w \approx 1000$ g/mol, $\rho = 980$ kg/m^3.
- SEBS [poly(styrene-*block*-ethylene-*co*-butadiene)]—$M_w = 43{,}000$, styrene content: 25%.
- SBS [poly(styrene-*block*-butadiene)]—$M_w = 39{,}000$, styrene content: 25%.

Sample Preparation

Compression-molded sheets of HDPE, SBS, SEBS, and PE—wax with thicknesses of 2–3 mm were used for this study. From C6–LLDPE a stack of blown films, each 0.025 mm thick for an overall thickness of 2 mm, was studied. SAXS and WAXS intensities were recorded in the machine direction (MD) and the transverse direction (TD) using the axes of the blown cylinder as the MD.

X-ray Diffractometry

A Rigaku Geigerflex generator was used with a wide-angle and a Kratky-type small-angle goniometer. A 30-kV accelerating voltage and a 30-mA current were applied using Ni-filtered Cu Kα radiation.

WAXS intensities were collected from $2\Theta = 3$–$50°$ with steps of 0.05° using transmission techniques on the compression-molded or multiple-layered film samples. Data were collected and processed using separate graphic software.

SAXS intensities were collected from $2\Theta = -1$–$1°$ in steps of 0.002° using the same samples as for WAXS. Background intensities were collected for each type of sample (thickness, absorption) from the same sample positioned just in front of the counter. Data were collected and processed using a computer program for removing the background and calculating the average of the data from the two sides of the primary beam. Slit correction (desmearing) was applied. For this purpose the derivative of the intensity data was obtained by a smooth derivation, using two neighboring data points at each side. A numerical integration was carried out on smoothed derivatives using limits of the data collection (s_o, s_e) for the integration according to eq. (9).

$$I(s) = -\frac{1}{\pi}\int_{s_0}^{s_e} \frac{I(\sqrt{s^2+t^2})}{\sqrt{s^2+t^2}}\,dt \tag{9}$$

The maximum resolution of the SAXS camera was 120 nm, as the intensities could generally be collected up to $2\Theta = 0.06°$. SAXS intensities from PE–wax were collected from 0.06° to 5° in 2Θ. These data were not corrected for the background.

Melting enthalpies were determined by temperature-modulated DSC (TMDSC), integrating the total heat flow from 0°C to the end of the melting peaks. A heating rate of 2 K/min was used with 40 sec as the modulation period and 0.6 K as the modulation amplitude in a TA Instrument TMDSC equipped with an intercooler device. Helium flushing gas with a flow rate of 25 mL/min was used.

RESULTS AND DISCUSSION

Figures 1–3 show the WAXS intensities of the PE sample materials. HDPE and PE–wax show normal orthorhombic crystalline modification of PE.[17–18]

The amorphous scattering intensities have been removed from the scattering intensities and

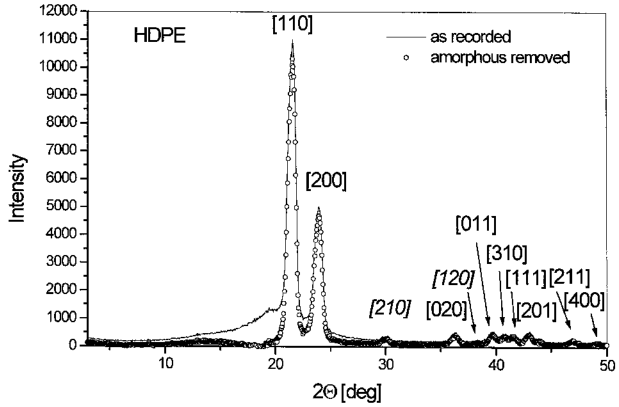

Figure 1 WAXS intensities of HDPE: total and crystalline diffraction.

the intensities of the pure crystalline phases are also shown in Figures 1 and 2. There is a broad maximum, with a peak position at 2Θ = 13°. This maximum is always present, and its intensity is proportional to the amount of the crystalline phase, that is, it cannot be included in the scattering intensities of the amorphous phase. Liquid polyisobutylene has a broad amorphous diffraction peak at this scattering angle. The crystallinity of the HDPE is 75%, and the crystallinity for PE–wax and C6–LLDPE is 67% and 53%, respectively (TMDSC result).

There are two peaks in the WAXS intensities of PE–wax corresponding to a lamellar period, as shown in Figure 2. The maxima of the peaks are at 3.3° and 5.1°, respectively. The *l* indices of the lamellar phase are underlined, those without underlining indicate the *l* indices of the PE subcell (*c* = 2.53 Å).

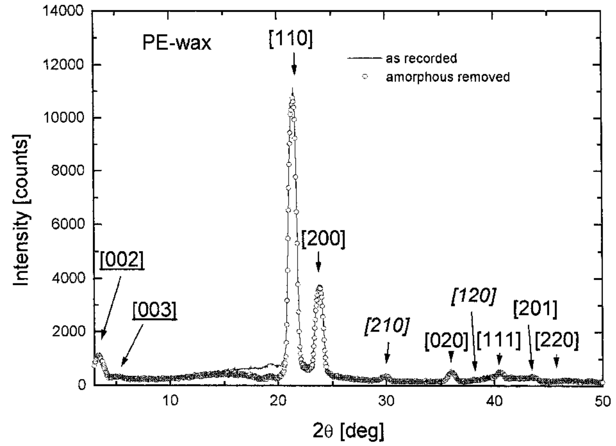

Figure 2 WAXS intensities of a high-molecular-weight wax: total and crystalline diffraction.

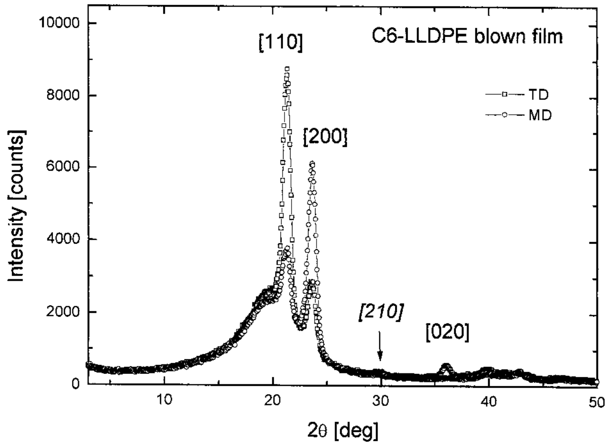

Figure 3 WAXS intensities of C6–LLDPE blown films recorded in two orthogonal directions.

The film blown from C6–LLDPE shows a strong orientation effect. There is an *a*–*b* orientation in the plane of the film. The diffraction peak with the index of [200] is high in the MD and low in the TD. The [020] peak is absent in the MD and present in the TD. The [110] peak is low in the MD as well.

The SAXS intensities of C6–LLDPE systems are shown in Figure 4. The intensities recorded in the TD and in the MD are different. There is no indication of a peak in the TD, but a very broad peak can be seen in the MD. The corresponding Bragg periodicity is 23 nm. The crystallinity of C6–LLDPE is calculated as 44.8% from the melting enthalpy (130 J/g) and as 55% on the basis of

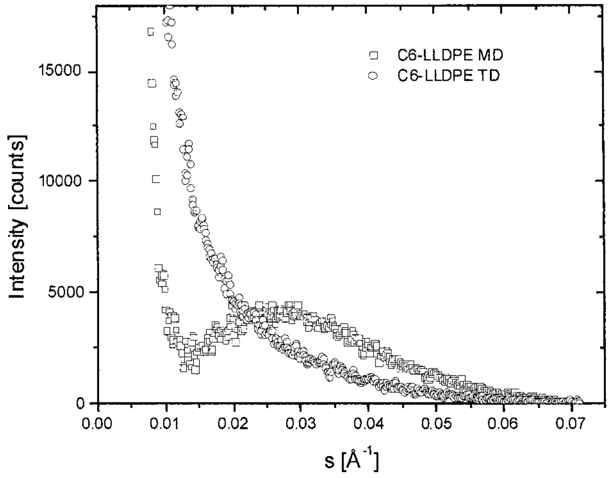

Figure 4 Desmeared SAXS intensities of C6–LLDPE film recorded in two orthogonal (machine and transverse) directions.

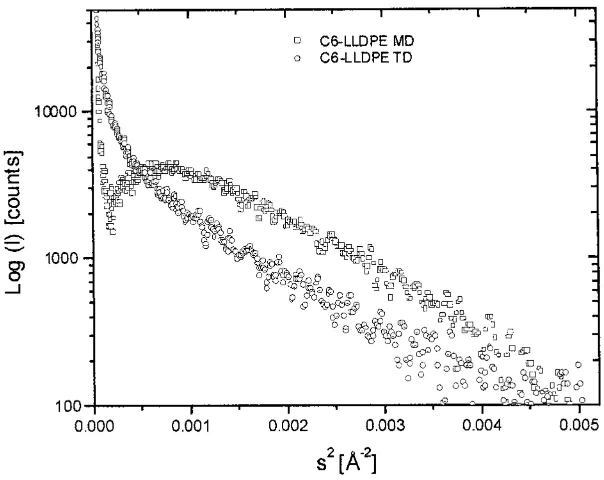

Figure 5 Desmeared SAXS intensities of C6–LLDPE. Guinier representation of the data is shown in Figure 4.

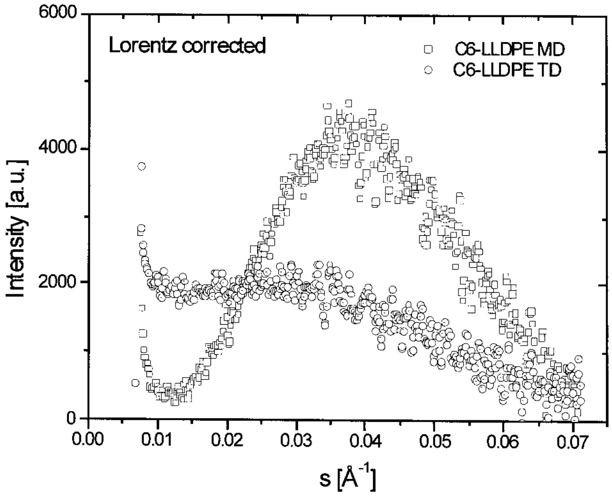

Figure 6 Lorentz-corrected SAXS intensities of C6–LLDPE derived from the data shown in Figure 4.

the density using 850 kg m^{-3} for the amorphous and 1005 kg m^{-3} for the crystalline PE, respectively.[10] The lamellar thickness of the crystallites is estimated to be ≈10 nm.

Figure 5 shows the Guinier representation of the same intensities shown in Figure 4. The curve recorded in the TD can be represented by the combination of two straight lines, which relate to particle scattering in the TD. The value for the radii of gyration determined from the initial slope in the TD is 17 nm and that for the second part is 5.2 nm. The corresponding lamellar thicknesses are 59 nm and 21 nm. These values show a much greater lamellar thickness than those estimated from the Bragg period. There is a broad peak superimposed on the straight lines, representing a combined particle scattering in the MD.

Figure 6 shows the Lorentz-corrected intensities of the same system. Both curves show a maximum. The maxima correspond to 25 nm in the TD and 16 nm in the MD, leading to a crystallite lamellar thickness of 7–11 nm.

The measured peak melting temperature of C6–LLDPE is 125.3°C, which corresponds to a lamellar thickness of 16 nm using eq. 10, the Thomson–Gibbs formula.[12]

$$T_m = T_m^0 * \left(1 - \frac{2\sigma_c}{\Delta H_u * L_c}\right) \tag{10}$$

where T_m is the melting temperature of a crystal with a thickness of L_c (cm), T_m^o is the extrapolated melting temperature of the crystals with infinite thickness (414 K), σ_c is the surface free energy (8.7 μJ cm^{-2}), and ΔH_u is the molar heat of fusion (2.9 × 10^2 J^{-1} cm^{-3}). The reference data in brackets is for poly(methylene).[12]

The Lorentz-corrected scattering curves resulted in much smaller lamellar thicknesses for the crystallites than was estimated from the melting peak temperatures. The particle size obtained from the Guinier approximation is much closer to the value expected from the melting peak temperature.

Figure 7 shows the SAXS intensities of HDPE, both as recorded and desmeared. There is a peak at s = 0.026 (Å^{-1}) corresponding to a lattice pa-

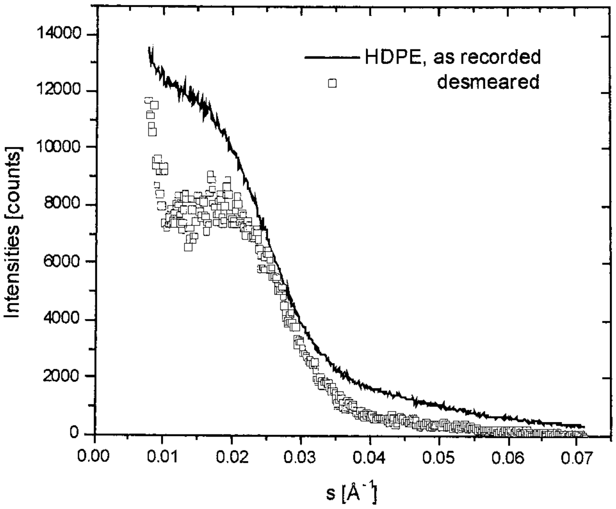

Figure 7 As-recorded and desmeared SAXS intensities of HDPE.

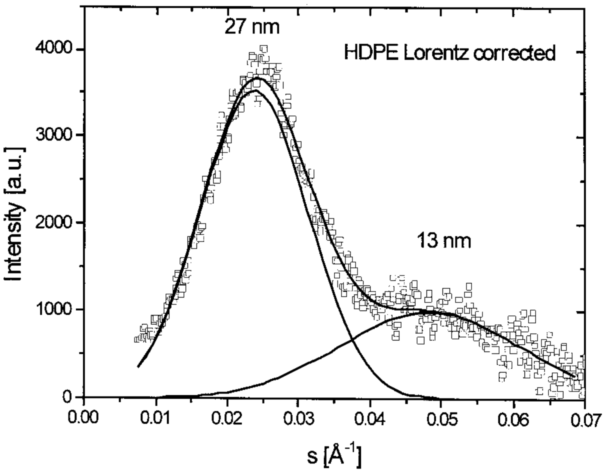

Figure 8 Lorentz-corrected SAXS intensities of HDPE: evolution of two Bragg peaks.

rameter of 25 nm. The melting enthalpy of HDPE is 205 J/g, corresponding to 70.6% crystallinity. The lamellar thickness of the crystallites is 17 nm. Figure 8 shows the Lorentz-corrected intensities of HDPE. There are two peaks in the curve, and they correspond to 26- and 13-nm Bragg periods. The melting temperature of this HDPE is 131°C, as observed from the TMDSC endotherm. Using the actual peak melting temperature and eq. (10), the calculated lamellar thickness is 36 nm. The Bragg periods read from the peak positions are much smaller than the value expected on the basis of the melting peak temperature.

The two peaks have half-line widths of 0.0021 rad and 0.0036 rad. This corresponds to $N_c = 2.9$ cells and $N_c = 3.5$ cells within a crystalline "particle," according to the Scherer formula .

$$B = \frac{K * \lambda}{N_c * d * \cos(\Theta_0)} \tag{11}$$

where N_c is the number of crystalline units with a periodical length of d, B is the half width of the diffraction peak expressed in radians, and K is a constant with a value of 1.07. The N_c numbers are very small. This means the peaks cannot be diffraction peaks as a result of a true reciprocal lattice. These numbers reflect a liquid structure rather than a crystalline one. The same conclusion can be drawn for C6–LLDPE, where the line widths—0.0051 rad and 0.0063 rad—yielded, respectively, 2.4 and 1 for the number of unit cells of the particle in the two directions.

The desmeared data were now handled as a superposition of the particle and crystalline scattering, as suggested by Dlugosz et al.[12] A least-square fit was calculated using two kinds of particles and a reciprocal lattice, according to eq. (12).

$$I = I_1^0 * \exp(-B_1 * s^2) + I_2^0 * \exp(-B_2 * s^2) + I_3^0 * \exp(-B_3 * (s_0^3 - s)^2) \tag{12}$$

Figure 9 shows the fit, which is excellent, and the refined parameters. If the areas under the individual peaks are considered, the reciprocal lattice-type diffraction has less than 10% of the weight of the total scattering intensities. The line width of the Gauss peak for a reciprocal lattice scattering was 0.0016 rad, another unrealistic value. The maximum corresponds to a periodicity of 57 nm, resulting in 1.8 unit cells based on the Scherer formula. With a peak in the scattering intensities, the Lorentz correction produces a smaller effect on the calculated periodicities.

SAXS studies on PE–wax served to prove that the equipment suitable for producing proper SAXS data as well as to show the upper limit of line broadening because of the equipment (instrumental line broadening). Figure 10 shows the SAXS intensities of the PE–wax as recorded together with the desmeared value and the Lorentz-corrected values of the latter one. As expected, [001] is present together with [002], which is indicated in Figure 2 (at 2Θ =3.3°), and the peaks

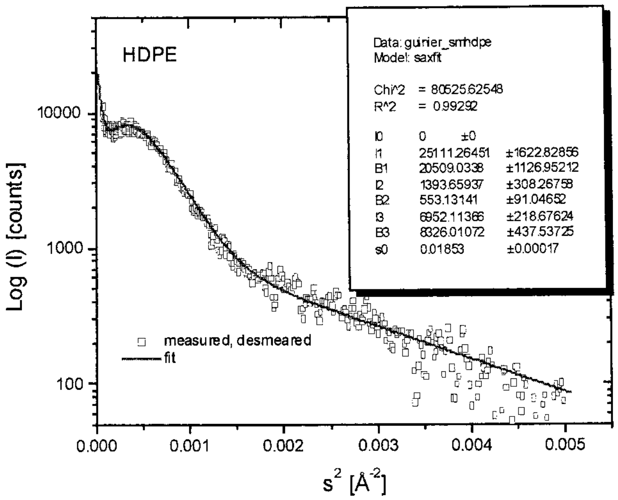

Figure 9 Desmeared SAXS intensities of HDPE. The fit of the two-phase model (reciprocal lattice and particle scattering).

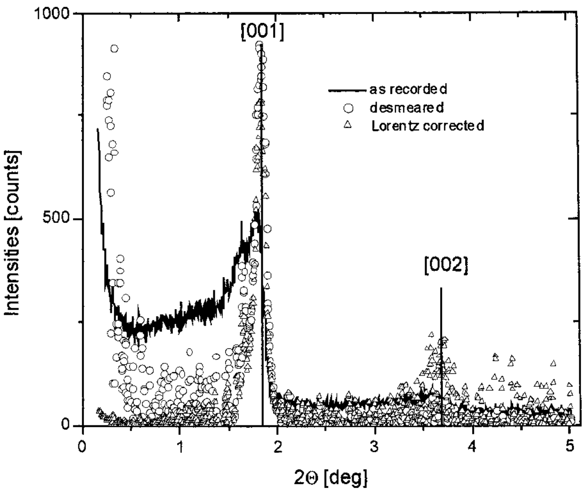

Figure 10 Comparison of as-recorded, desmeared, and Lorentz-corrected SAXS intensities of high-molecular-weight wax.

are asymmetric in the as recorded data because of the smearing of the line slit. The Lorentz correction brought [002] to an intensity commensurable with [001]; nevertheless the position of the maximum did not change. The maxima correspond to a 4.85-nm periodicity. The particle size of the crystals along the *c*-axis is 72 or 47 nm, estimated from the line broadening without the correction of the instrumental broadening (0.0023 rad and 0.0030 rad). This means there are at least 15–19 consecutive unit cells in this direction, which is in the range of relatively small crystals. Using proper instrumental line broadening, this number is higher. There is negligible scattering intensity caused by particle scattering in spite of the presence of a measurable amorphous phase. The melting enthalpy of the PE–wax is 194 J/g, which corresponds to 67% crystallinity. The WAXS curves suggest a much greater value (95%). The melting temperature of the PE–wax is 60°C, corresponding to a lamellar thickness of 3.1 nm. Using the measure of crystal particle size determined without the Lorentz correction (i.e., 45 nm) and the TMDSCX crystallinity, the crystalline lamellar thickness is calculated as 3.3 nm. This fits quite well with the value estimated by the Thomson–Gibbs formula.

As a comparison again, Figure 11 shows the SAXS intensities of two block copolymers of styrene and butadiene in a Guinier representation. One of the copolymers has been partially hydrogenated. There are peaks on the scattering intensity curves corresponding to 27–29 nm periodicity. When the desmeared intensities are compared to those of the Lorentz-corrected ones (Fig. 12), only a small shift in the peak position can be detected in the direction of the smaller periodicities (26 nm). The expected change in the peak position by a Lorentz correction is greater when the peak is closer to the zero scattering angle. This is the case for SBS and the wax; however, even in this case the shift is small (less than 10%). The TEM pattern of the blends containing the block copolymer with unsaturation reveals a 25–30 nm periodic arrangement of alternating styrene and butadiene blocks.[21]

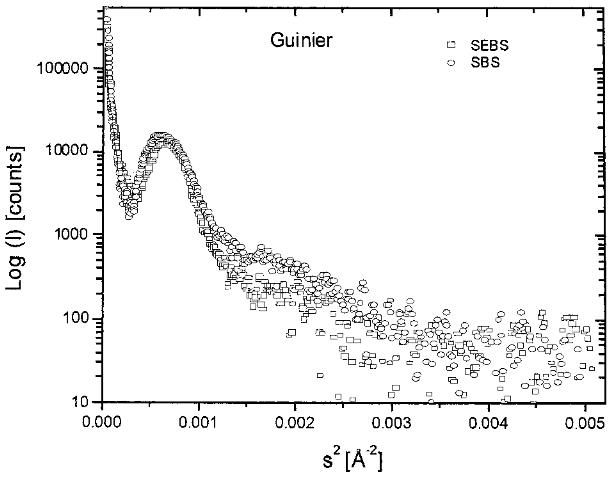

Figure 11 Desmeared SAXS intensities of styrene–butadiene block copolymers in Guinier representation.

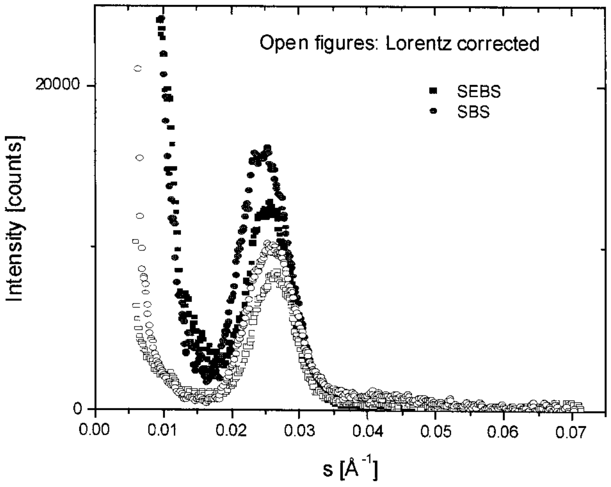

Figure 12 Desmeared (filled figures) and Lorentz-corrected (open figures) SAXS intensities of styrene–butadiene block copolymers.

CONCLUSIONS

The experiments carried out on different semicrystalline polymers and amorphous block copolymers with narrow molecular-weight distributions showed that peaks in the SAXS intensities are present when the system contains a periodic structural arrangement. In this case Lorentz correction might result in diffraction peaks corresponding to lesser Bragg periods near to zero-diffraction angles.

Many of the semicrystalline polymers such as PE did not produce Bragg diffraction peaks at SAXS angles, possibly because of structural peculiarities. The source of the scattering is either particle type or fractal, and there is no Bragg periodicity in the system. Lorentz correction produces artificial Bragg periodicity in these systems. The parameters obtained from the so-called diffraction peaks as a result of the applied Lorentz correction are misleading. The Lorentz correction, particularly its squared form, should not be used to interpretat SAXS intensities when there is no true indication of the presence of reciprocal lattice scattering. Naturally, the invariant should be calculated in the usual way.

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