Polymer 43 (2002) 7307–7313

polymer

www.elsevier.com/locate/polymer

Perfectly alternating ethylene–carbon monoxide copolymer crystallized epitaxially on alkali halides. 2. Energy calculation

Masahiro Fujita[a,1], Kyo Jin Ihn[b], Masaki Tsuji[a,*], Shinzo Kohjiya[a]

[a]Laboratory of Polymer Condensed States, Division of States and Structures III, Institute for Chemical Research, Kyoto University, Uji, Kyoto-fu 611-0011, Japan

[b]Department of Chemical Engineering, Kangwon National University, Chunchon 200-701, South Korea

Received 30 April 2002; received in revised form 11 September 2002; accepted 30 September 2002

Abstract

As reported in our previous papers, edge-on lamellar crystals of poly(3-oxotrimethylene) (POK) can be epitaxially grown from solution on alkali halides, and the crystal orientation depends on the type of substrate utilized: The chain-stems on the (001) surface of NaCl orient in the ⟨110⟩ directions of the substrate, while those on the (001) surface of KI orient in the ⟨100⟩ directions of the substrate. In order to explain this experimental evidence, calculations of the interaction energy between one rigid POK chain-segment of planar–zigzag conformation and a given substrate (the (001) surface of NaCl or KI) were carried out: in the calculations, the Lennard–Jones 6–12, the Coulomb and the induced-dipolar potentials were utilized. The calculation results were in good agreement with the experimental facts. Particularly in the calculation for POK on KI, the Coulomb energy was found to be the most important of the three to explain the the ⟨100⟩ orientation of the chain-segment. It was concluded that the interaction energy between the oxygen atoms in the POK chain-segment and the positive ions in a given alkali halide is an important factor to determine the orientation of the chain-stem in epitaxial crystallization of POK on these alkali halides. © 2002 Published by Elsevier Science Ltd.

Keywords: Polyolefine ketone; Poly(3-oxotrimethylene); Edge-on lamellar crystal

1. Introduction

It is well known that some linear polymers are epitaxially crystallized from solution on the (001) surface of an alkali halide [1,2]. In the epitaxial growth of polyethylene (PE), the resultant morphology appears as rodlike crystals oriented in the ⟨110⟩ directions of the substrate. Transmission electron microscopy (TEM) reveals that each of the rodlike crystals is a chain-folded edge-on lamella, i.e. the molecular chain-stems in the rodlike crystal are set perpendicular to the long axis of the rod and parallel with the substrate surface [3,4]. Such morphology has been reported on the epitaxy of other polymers on alkali halides [2]. For example, the epitaxy of polyoxymethylene (POM) on an alkali halide (NaCl or KCl) has also been extensively studied, and the rodlike crystals or the chain-stems are oriented in the ⟨110⟩ directions of the substrate [5,6], as in the case of PE.

Recently, we have found that poly(3-oxotrimethylene) (POK), which is a kind of polyolefin ketones (abbreviated as POK in this paper), can be grown as single crystals of α-form [7] from a dilute solution in nitrobenzene [8], and also that this POK can be epitaxially crystallized isothermally from such a solution on an alkali halide (NaCl or KI) [9–11]. TEM revealed that POK chain-stems in the resulting rodlike crystals which were grown on the substrate are parallel with the substrate surface and perpendicular to the long axis of the rod (actually, these rodlike crystals are edge-on folded-chain lamellae, as in the case of PE and POM), and that the crystal orientation depends on the kind of substrate [9–11]: the rodlike crystals on the (001) surface of NaCl orient in the ⟨110⟩ directions of the substrate, and ones on the (001) surface of KI orient in the ⟨100⟩ directions of the substrate. That is to say, the chain-stem axis (namely, the *c*-axis in the crystals of α-form [7] and β-form [12,13] which are both orthorhombic) on the (001) surface of NaCl

---

[*] Corresponding author. Tel.: +81-774-38-3061; fax: +81-774-38-3069. *E-mail address:* tsujimas@scl.kyoto-u.ac.jp (M. Tsuji).

[1] Research Fellow of the Japan Society for the Promotion of Science (1998–2001). Present address: Polymer Chemistry Laboratory, The Institute of Physical and Chemical Research (RIKEN Institute), Hirosawa, Wako, Saitama-ken 351-0198, Japan.

0032-3861/02/$ - see front matter © 2002 Published by Elsevier Science Ltd.
PII: S0032-3861(02)00701-2

orients in each of the ⟨110⟩ directions of the substrate (epitaxial mode: $\langle 001\rangle_{\mathrm{POK}}\|\langle 110\rangle_{\mathrm{NaCl}}$) [9,10], while the chain-stem axis on the (001) surface of KI orients in each of the ⟨100⟩ directions of the substrate ($\langle 001\rangle_{\mathrm{POK}}\|\langle 100\rangle_{\mathrm{KI}}$) [9–11].

In order to explain these TEM observations, energy calculation for polymer–substrate interaction is carried out using our homemade program in this work. The calculation method used here is basically similar to the one introduced by Mauritz et al. [14], which is a computer-simulation method to calculate static interaction energy between a single rigid chain-segment and an alkali halide. The calculation method was applied not only for the epitaxy of PE and POM on alkali halides [2,14,15] but also for vapor-deposited PE on cycloparaffin and normal-paraffin crystals [16] or for epitaxial synthesis of poly(*p*-xylylene) on alkali halides [17], and each of the calculation results was consistent with the corresponding experimental fact. It is noted, however, that in the calculations for PE and POM and so on, however, the dispersion-repulsive energy (for example, due to the Lennard–Jones 6–12 (LJ 6–12) potential; see Section 2.2) made a major contribution to the total interaction energy. [2,14,15]

In this paper, the computation of the interaction energy between one rigid POK chain-segment and a given substrate (the (001) surface of NaCl or KI) is carried out and demonstrates that the Coulomb potential makes the greatest contribution to the total energy in such a system, and the results are discussed in comparison with our experimental evidence [9–11] obtained by TEM observations.

2. Calculation method

2.1. Geometry

In a similar way as the work in the past [2,14–17], the static interaction between the rigid POK chain-segment of planar–zigzag conformation, consisting of six monomer units, and the (001) surface of NaCl or KI was considered in the present calculation. As illustrated in Fig. 1, six independent spatial parameters x, y, z, ϕ, μ, θ are required to define the position and orientation of the chain segment relative to the (001) surface of substrate [2,14,15]. The origin, O, is centered on a given positive ion of the substrate, and the X- and Y-axes correspond, respectively, to [100] and [010] directions of the substrate. Coordinates (x, y, z) denote an end of the chain axis: the parameter z represents the height from the surface, and the x and y are the fractional coordinates in the crystal lattice of the alkali halide. The parameter μ ($0° \leq \mu \leq 90°$) is the rotation angle of the chain-segment around its own axis, and this parameter is defined so that $\mu = 0°$ corresponds to the case where the plane of the zigzag chain-segment is perpendicular to the substrate surface. The θ is the azimuthal angle of the chain axis measured from the [001] direction of the substrate. The present calculation was carried out under the condition that the chain-segment was set parallel with the substrate surface, viz., $\theta = 90°$ so that the other five parameters were used as variables in the calculation. Accordingly here, the ϕ is the angle of this chain-segment measured from the [100] direction of the substrate. The increment of each of the five spatial variables used in this calculation is as follows: $\phi = \mu = 15°$, $z = 0.01$ nm, and $x = y = 1/16$.

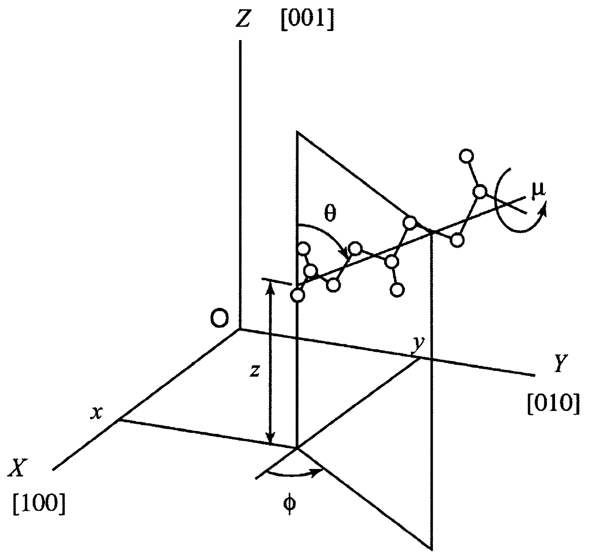

Fig. 1. Schematic representation of a POK chain-segment on the (001) surface of substrate. The origin O is centered on a positive ion; (x, y, z) is a coordinate of an end of the chain segment, where x and y correspond to fractional coordinates of substrate crystal and z is the height from the (001) surface of the substrate.

2.2. Molecular energetics

The coordinates of atoms and ions in the POK chain-segment and the substrate, respectively, were calculated by using the data shown in Fig. 2 for POK and for the lattice constant of substrate. The dispersion–repulsion force (LJ 6–12 potential; U_{LJ}) and electrostatic interactions (Coulomb

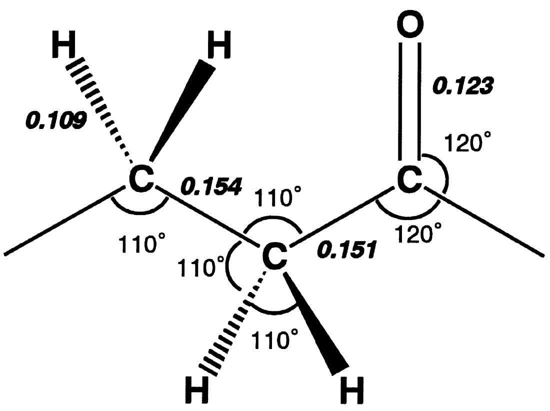

Fig. 2. Chemical structure of POK. Bond lengths (italic, in nanometers) and angles of POK used for the present calculation are indicated. These parameters were quoted from Ref. [7].

and induced-dipolar potentials are to contribute to interaction energy between the chain-segment and a substrate; U_C and U_{ID}) [2,14–17]. The energy was, therefore, estimated by summing up these three functions ($U_{LJ} + U_C + U_{ID}$).

As well known, the U_{LJ} is a function of the distance (r) between two points. The LJ 6–12 potential function between the ith atom in the chain-segment and the jth ion in the substrate can be expressed as

$$U(r_{ij}) = -\frac{A_{ij}}{r_{ij}^6} + \frac{B_{ij}}{r_{ij}^{12}} \tag{1}$$

where r_{ij} is the distance between the ith atom and the jth ion, and A_{ij} and B_{ij} are the attractive and repulsive energy constants, respectively (Table 1). These constants were calculated in the same way as those in Ref. [14]. The U_{LJ} for $r > 1$ nm makes a minor contribution to the interaction energy. In this study, hence, the ions located in a region of $r_{ij} > R$ ($= 1$ nm) apart from a given atom in the chain-segment were treated as being uniformly distributed in a continuum (see Refs. [2,14–17]), with the following expression

$$\begin{aligned} U(r_{ij} > R) &= U_{ik}(z_i) \\ &= -\frac{\pi}{2R^3}\left(\frac{R - z_i}{R} + \frac{1}{3}\right)n_k A_{ik} + \frac{\pi}{5R^9} \\ &\quad \times \left(\frac{R - z_i}{R} + \frac{1}{9}\right)n_k B_{ik} \end{aligned} \tag{2}$$

where the subscript k means the k-type ion in the substrate, and the parameters, n_k and z_i, are, respectively, the number density [2,14,15] of the k-type ions and the height of ith atom in the chain-segment from the substrate surface. Thus the total LJ 6–12 potential is described as

$$U_{LJ} = \sum_i \sum_j \{U(r_{ij} \le R) + U(r_{ij} > R)\} \tag{3}$$

According to Ref. [14], for alkali halide crystals, the electrostatic potential in kcal mol^{-1} at an external position (x_i,y_i,z_i) is given by

$$\begin{aligned} \phi(x_i, y_i, z_i) &\approx 3712\left(\frac{\nu e}{d}\right) \\ &\quad \times \exp(-2\pi z_i/d)\cos(2\pi x_i/d)\cos(2\pi y_i/d) \end{aligned} \tag{4}$$

where ν is the valence of the substrate ions, e is the electronic charge, viz., elementary electric charge (in this study, $\nu = 1$ and $e = 1$ (e.u.)), and d is the lattice constant (viz., unit cell dimension; $d_{NaCl} = 0.564$ nm, $d_{KI} = 0.7065$ nm [18]) of the substrate [14] (therefore, in Eq. (2), for example, the number density n_k for Na$^+$ in NaCl is calculated to be $4(d_{NaCl})^{-3}$). Thus, the total Coulomb potential (U_C) is calculated by multiplying the ϕ of ith atom by the partial charge (e_i; Table 2) residing on the atom, and summing over all atoms in the chain segment

$$U_C = \sum_i e_i \cdot \phi(x_i, y_i, z_i) \tag{5}$$

Table 1

Attractive and repulsion energy constants of atom–ion pairs in LJ 6–12 function

	$A_{ij} \times 10^4$ (kcal nm^6 mol^{-1})				$B_{ij} \times 10^8$ (kcal nm^{12} mol^{-1})			
Ion	C	C(O)[a]	O	H	C	C(O)[a]	O	H
Na	0.964	1.18	0.956	0.336	1.71	2.14	1.14	0.175
K	3.60	4.50	3.64	1.28	13.9	17.4	9.74	1.67
Cl	7.86	9.84	7.94	2.79	7.35	92.0	54.2	10.4
I	14.7	18.3	14.8	5.20	258	323	196	40.1

The values of A_{ij} were calculated from the following equation; $A_{ij} = (A_{ii}A_{jj})^{1/2}$. Here, A_{ii} and A_{jj} are attractive energy constants for identical atoms in polymers and for identical ions in substrate, respectively [14]. In this work, the A_{ii}'s were calculated according to Slater–Kirkwood [19], and the A_{jj}'s were quoted from Ref. [20]. The values of B_{ij} were calculated according to the Slater–Kirkwood [19].

[a] Carbon atom in the carbonyl group.

The induced-dipolar contribution is described as

$$U_{ID} = -\frac{1}{2}\sum_i \alpha_i E_i \tag{6}$$

where α_i is the atomic polarizability of the ith atom and E_i is the magnitude of electric field at the position of that atom: $\mathbf{E}(x_i,y_i,z_i) = -\nabla\phi(x_i,y_i,z_i)$ [14]. The parameters required for the potential functions are listed in Tables 1 and 2. The sum of the three potential energies, $U_{LJ} + U_C + U_{ID}$, was regarded as the total interaction energy between the single POK chain-segment and the substrate (NaCl or KI), as mentioned above.

3. Results

3.1. POK chain-segment on the (001) surface of NaCl

Potential contour maps showing the interaction energy of the POK chain-segment on the (001) surface of NaCl substrate are plotted in kcal mol^{-1} of 6 monomer units: The zones with negative energy values are darkened in Fig. 3(a)–(c) (and also in Fig. 4(a)–(c)), and the darker

Table 2

Partial charges (e_i) and polarizabilities (α_i) of atoms in POK

Atom	e_i,[a] (e.u.)	$\alpha_i \times 10^3$ (nm^3)[b]
C	−0.0794	0.93
C(O)	0.599	1.30
O	−0.531	0.84
H	0.0223	0.42

[a] These values are Mulliken charges calculated by MINDO/3 for the rigid POK chain-segment consisting of six monomer units. Calculation was performed on a personal computer using a commertial software package, 'CS Chem3D version 4.0' (CambridgeSoft Corp., USA) for Macintosh.

[b] Data from Refs. [19,21].

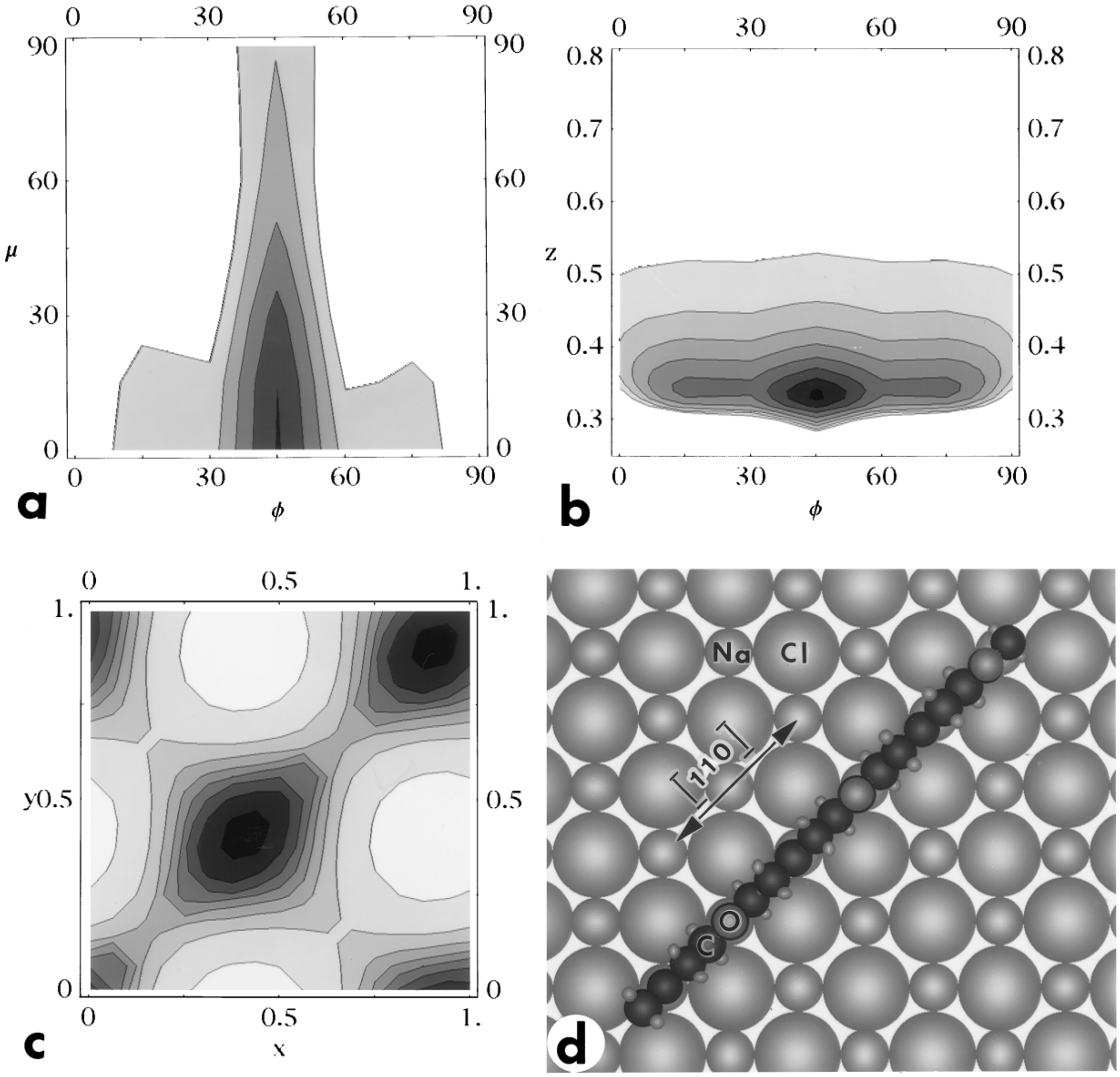

Fig. 3. Energy contour maps of POK chain-segment on the (001) surface of NaCl plotted as a function of (ϕ, μ) (a), of (ϕ, z) for $\mu = 0°$ (b) and of (x, y) for $\phi = 45°$ and $\mu = 0°$ (c), and the preferred position of the chain-segment relative to the substrate surface (d). In (a)–(c), contour lines were drawn only for the negative energy values at arbitrary intervals (the darker in gray level, the lower in energy), and ϕ, μ in degrees; z in nanometers; x, y: fractional coordinates.

in gray level, the lower in energy. Fig. 3(a) illustrates the potential contour map as a function of ϕ and μ. The value at each set of (ϕ, μ) in this figure is the lowest in the calculation results for all (x, y, z) with the given ϕ and μ. The deep potential valley appears at $\phi = 45°$ along μ and its minimum locates at $\phi = 45°$ and $\mu = 0°$. The potential energy is more affected by the variable ϕ than by μ. The valley at $\phi = 45°$ may cause the absorbed chain-segment to take its preferential orientation, that is, the POK chain-segment is expected to orient most stably in the [110] direction of NaCl. This result strongly supports the experimental fact in our previous work [9,10].

Fig. 3(b) shows the potential contour map as a function of ϕ and z for $\mu = 0°$, in order to see the influence of the variable z on the potential energy. In analogy with the results in Refs. [14–16] and [21], this figure well demonstrates that the potential energy is less affected by the substrate if the chain segment is far from the surface, while there appears a potential minimum according to the distance from the substrate surface when $z < 0.4$ nm.

Fig. 3(c) shows the potential map when the POK chain-segment moves on the xy-plane, namely the (001) plane, of NaCl. This figure is plotted for $\mu = 0°$, $\phi = 45°$, and z which yields the lowest potential value at a given set of (x, y). This figure indicates that the chain-segment on the array of the positive ions (Na^+) is far more stable than on that of the negative ions (Cl^-), as illustrated in Fig. 3(d). The interaction energy is, however, very sensitive to the translation of the chain segment even on that of the positive ions. This is due to the existence of the carbonyl groups in

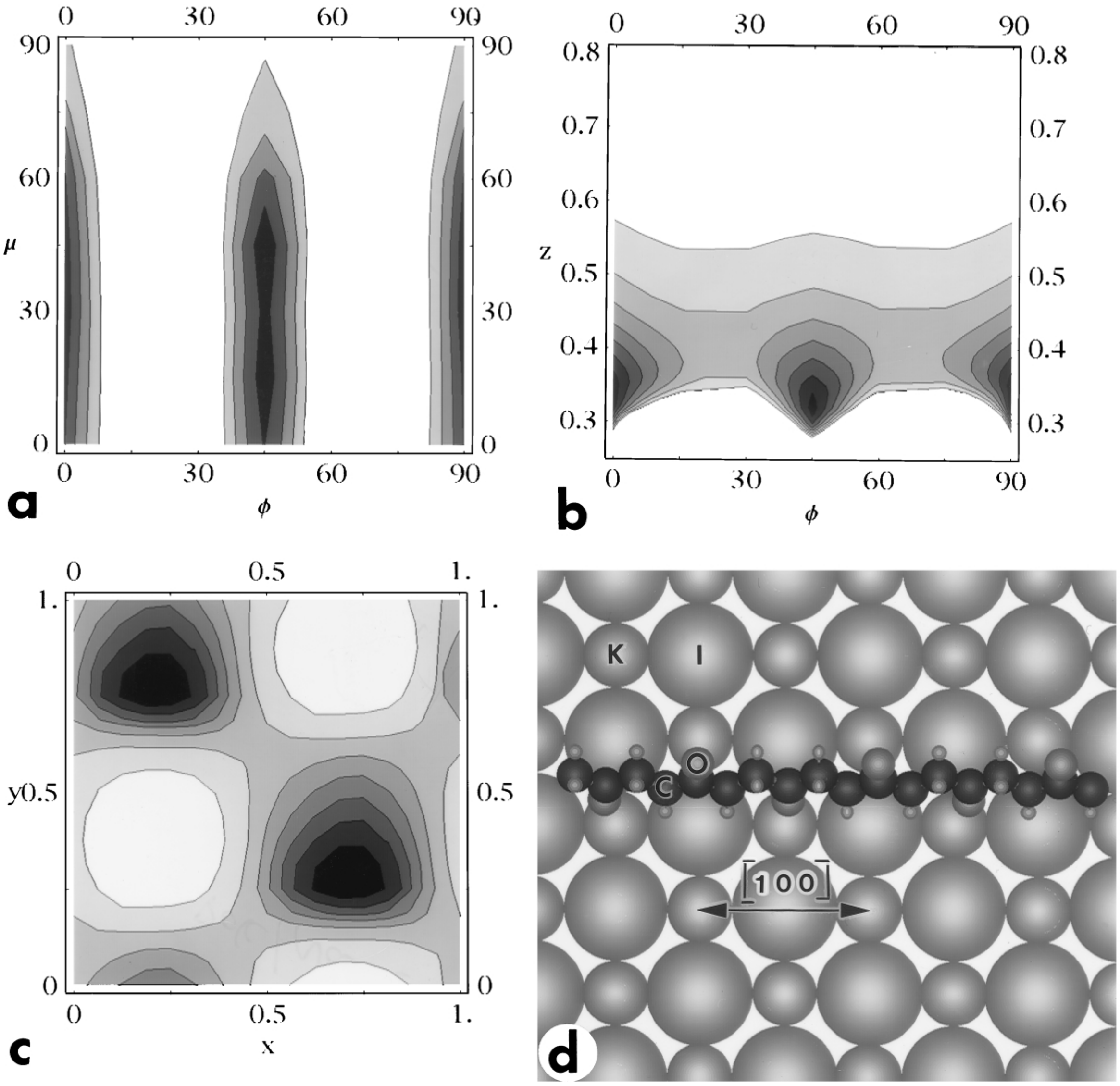

Fig. 4. Energy contour maps of POK chain-segment on the (001) surface of KI plotted as a function of (ϕ, μ) (a), of (ϕ, z) for $\mu = 30°$ (b) and of (x, y) for $\phi = 0°$ and $\mu = 30°$ (c), and the preferred position of the chain-segment relative to the substrate surface (d). In (a)–(c), contour lines were drawn only for the negative energy values at arbitrary intervals (the darker in gray level, the lower in energy), and ϕ, μ in degrees; z in nanometers; x, y: fractional coordinates.

the POK chain, i.e. the interaction between oxygen atoms and sodium ions is responsible for such sensitivity. This seems to mean that the lattice matching between the POK chain-segment and alkali halide crystals is important, not similarly to the cases of PE and POM on alkali halides [2,14,15].

3.2. POK chain-segment on the (001) surface of KI

Potential energy of a rigid POK chain-segment with 6 monomer units on the (001) surface of KI was calculated (in kcal mol^{-1} of 6 monomer units), as in the case of the NaCl substrate. Fig. 4(a)–(c) are potential contour maps for the

Table 3
Minimum interaction energy and relative energy contributions for each alkali halide

Preferred orientation	NaCl	KI	
	[110]	[100]	[110]
Minimum interaction energy (kcal mol^{-1} of 6 monomers)	−63.8	−58.3	−57.9
LJ 6–12[a] (%)	10.2	22.4	28.1
Coulomb energy[a] (%)	69.7	61.6	42.1
Induced-dipolar energy[a] (%)	20.1	16.0	29.8

[a] Each 'energy' itself is a negative value.

POK chain-segment on the (001) surface of KI, and are plotted as functions of (ϕ, μ), (ϕ, z) and (x, y), respectively. In contrast to the results calculated for the POK–NaCl system, two energy minima were obtained at $\phi = 0°$ (which is identical with 90°) and $\phi = 45°$, corresponding to the directions of [100] and [110] of KI, respectively (see Fig. 4(a) and (b)). As summarized in Table 3, the calculation in this work suggested that the POK chain-segment orients the most stably in the [100] direction of KI, and this result agrees well with our experimental evidence obtained in our previous work [9–11]. On the other hand, the present calculation also predicts the second stable orientation of the chain-segment in the [110] direction. In this case, the chain-segment should lie over the array of positive ions, although we have never observed the POK edge-on crystals oriented in the [110] direction of KI.

Fig. 4(c) shows the potential contour map as a function of x and y at $(\phi, \mu) = (0°, 30°)$. The POK chain-segment corresponding to the energy minimum in this figure is situated with each of the oxygen atoms on the substrate side being deposited on the positive ion, as shown in Fig. 4(d). The degree of mismatching between the distance of the oxygen atoms on the substrate side and that of potassium ions along the [100] direction is small and estimated at 7.6%. This value is within the accepted range of lattice mismatches for epitaxy ($<15\%$) [22]. Accordingly the POK chains oriented in this direction can be expected to crystallize epitaxially on KI. All the present calculation results, including the results for NaCl, have given the conclusion that the interaction between the oxygen atom in the polymer and the positive ion in an alkali halide is one of the important factors to determine the chain orientation on epitaxial crystallization. When the (001) surface of KI is utilized as a substrate, it is, thus, deduced that the interaction energy causes the POK chain to orient not in the [110] direction but in the [100] one of the substrate, although the energy difference between these two orientations was rather small in our present results of computation (see Table 3).

4. Discussion

In the present computation, the sum of LJ 6–12, Coulomb and induced-dipolar potential energies was regarded as an interaction energy between the single chain-segment and each of the substrates. The relative energy contributions at each minimum interaction energy are listed in Table 3 (it should be emphasized here that every contribution to the minimum interaction energy is negative). The main contribution to the interaction energy was presumed to be not due to the LJ 6–12 potential for each substrate. In contrast to the result for PE [14] in which the contribution due to the LJ 6–12 potential was more than 70% for NaF, NaCl and RbBr, and also to the result for POM [15] in which Coulomb energies were weak, our calculation has proved that other contributions exceeding the contribution due to the LJ 6–12 potential must be taken into account for POK. In the case of epitaxy of POK on alkali halides, the electrostatic interaction is orientation-sensitive, and dominates the preferred orientation and position of the POK chain-segment. The Coulomb potential, therefore, between the chain-segment and the substrates seems to be an important factor to determine the orientation and position of the chain-segment, because the induced-dipolar potential tends to lower the total energy under any condition [2,14,15]. It is, thus, more advantageous for the POK chain that its oxygen atoms lie on the positive ions of the substrates, if possible.

In the case of the (001) surface of NaCl as a substrate, the present calculation indicates that the POK chain-segment orients in the [110] direction of the substrate, which agrees well with the TEM evidence, and that the chain-segment orients along a row of positive ions. Even if only the LJ 6–12 potential energy was considered, the result also predicted that the chain-segment is to lie on the array of sodium ions. The [110] orientation of the POK is not hindered by the electrostatic interaction energy, especially by the Coulomb potential energy, because the oxygen atoms are apt to deposit themselves on the positive ions. The other orientations of the POK chain-segment except for the [110] orientation, however, are not possible, because they lead to the increase of electrical repulsion force. Thus, the present calculation supported that the chain-segment orients exclusively in the [110] direction of NaCl.

As for the (001) surface of KI, the situation is slightly different from the case of NaCl. The calculation result showed the most stable orientation where the chain-segment orients in the [100] direction, although the second stable potential minimum corresponding to the [110] direction was also obtained. The most stable orientation on KI is apparently due to a small misfit between the distance of the oxygen atoms of the chain and that of the potassium ions along the [100] direction of the substrate. As mentioned above, the misfit is 7.6% for the [100] orientation of POK chain-segment, so that even the [100] orientation leads to the gain by the Coulomb potential energy because the oxygen atoms are able to deposit themselves over the potassium ions, as shown in Fig. 4(d). Furthermore, the protruding oxygen atom from the chain axis fits in with the basin of substrate, which corresponds to the position of potassium ion. This condition also leads to the gain of the LJ 6–12 potential energy. In practice, an energy minimum was given at $\phi = 0°$ even when the interaction energy was estimated only with the LJ 6–12 potential function.

The energy calculation in this work was carried out under some restrictions and assumptions; for example, the rigid short chain-segment, the infinite and flat surface of substrate, and so on. It may be necessary to remove such restrictions and assumptions or to improve the calculation method, in order to simulate the real phenomenon (namely, the fact that only the [100] orientation of POK chain-stem axis was experimentally recognized). The present calculation results, however, have given the information that the interaction between the oxygen atom in the polymer and the positive ion in the substrates is an important factor to determine the chain orientation of the polymer on the substrate, and accordingly that this interaction causes preferred orientation dependent on the kind of substrate.

5. Concluding remarks

The energy analysis was performed to confirm and support the dependence of the orientation of POK chain-stems (or its rodlike crystals) on the kind of alkali halide by calculating the interaction energy between the single rigid chain-segment and each of the substrates, similarly to the work by Mauritz et al. [2,14,15]. The calculation results were well consistent with our previous experimental evidence [9–11], and it is concluded that the interaction energy between the oxygen atom in the polymer and the positive ion in each substrate is an important factor to determine the chain orientation.

For the epitaxy of POK on the (001) surface of NaCl, the calculation result supported that the preferred orientation of the POK chain is exclusively in the [110] direction. On the other hand, for the epitaxy on the (001) surface of KI, the POK chain orients stably in the [100] direction, although the calculation result also suggested the other possible orientation (the [110] direction) which has not been observed so far by TEM [9–11]. Although the three kinds of potentials (namely, the LJ 6–12, the Coulomb and the induced-dipolar potentials) were introduced in our computation for POK, the contribution of the Coulomb potential to the total interaction energy was much more greater than those of the other two, as shown in Table 3. In particular, the Coulomb energy was found to be the most important in order to explain the experimentally observed [100] orientation of the POK chain axis on KI. It is noted, however, that the preferential orientation of chain axis for PE [2,14], POM [15] and polythiomethylene [15] on alkali halides could be satisfactorily explained even with only the dispersion-repulsive force (the LJ 6–12 potential) because the contribution of the force to the total interaction energy is the largest of the three kinds of potentials.

Acknowledgements

M. Fujita expresses his thanks to the Research Fellowships of the Japan Society for the Promotion of Science (JSPS). This work was supported partly by a Grant-in-Aid for JSPS Fellows (No. 9796; to M. Fujita), and also partly by a Grant-in-Aid for Scientific Research on Priority Areas (B), ‘Mechanism of Polymer Crystallization’ (No. 12127207; to M. Tsuji), both of which were from the Ministry of Education, Culture, Sports, Science and Technology, Japan.

References

[1] Wunderlich W. Macromolecular physics. Crystal structure, morphology, defects, 1. New York: Academic Press; 1973. p. 275–80.
[2] Mauritz KA, Baer E, Hopfinger AJ. J Polym Sci Macromol Rev 1978; 13:1–61.
[3] Ihn KJ, Tsuji M, Kawaguchi A, Kayayama K. Bull Inst Chem Res, Kyoto Univ 1990;68:30–40.
[4] Tsuji M, Ihn KJ. Bull Inst Chem Res, Kyoto Univ 1995;72:429–42.
[5] Fujita M, Hamada N, Tosaka M, Tsuji M, Kohjiya S. J Macromol Sci Phys 1997;B36:681–7.
[6] Fujita M, Tsuji M, Kohjiya S. Polymer 1999;40:2829–36.
[7] Lommerts BJ, Klop EA, Aerts J. J Polym Sci: Part B: Polym Phys 1993;31:1319–30.
[8] Fujita M, Tsuji M, Murakami S, Kohjiya S, Wittmann JC. Macromolecules 2001;34:6147–51.
[9] Fujita M, Tsuji M, Kohjiya S, Wittmann JC. Macromolecules 1999; 32:4383–9.
[10] Fujita M, Ihn KJ, Tsuji M, Kohjiya S. Kobunshi Ronbunshu 1999;56: 786–96.
[11] Fujita M, Tsuji M, Kohjiya S. Macromolecules 2001;34:7724–9.
[12] Chatani Y, Takizawa T, Murahashi S, Sakata Y, Nishimura Y. J Polym Sci 1961;55:811–9.
[13] Chatani Y, Takizawa T, Murahashi S. J Polym Sci 1962;62:S27–S30.
[14] Mauritz KA, Baer E, Hopfinger AJ. J Polym Sci, Polym Phys Ed 1973; 11:2185–97.
[15] Mauritz KA, Hopfinger AJ. J Polym Sci, Polym Phys Ed 1975;13: 787–98.
[16] Ihn KJ, Tsuji M, Isoda S, Kawaguchi A, Katayama K. Macromolecules 1990;23:1788–93.
[17] Isoda S. Polymer 1984;25:615–24.
[18] ‘Kagakubinran Kisohen II’, 4th ed. Maruzen, Tokyo: The Chemical Society of Japan; 1993. Chapter 15.
[19] for example, Tadokoro H. Structure of crystalline polymers. New York: Wiley Interscience; 1979. chapter 6.
[20] Balik CM, Tripathy SK, Hopfinger AJ. J Polym Sci, Polym Phys Ed 1982;20:2017–26.
[21] Scott RA, Sheraga HA. J Chem Phys 1966;45:2091–101.
[22] Swei GS, Lando JB, Ricket SE, Mauritz KA, in ‘Encyclopedia of polymer science and engineering’, 6. New York: Wiley; 1986. p. 209–24.