

## **Yushu Matsushita**

Professor Emeritus, Nagoya University

### **Education:**

Mar. 1977 Bachelor of Engineering, Nagoya University

Mar. 1979 Master of Engineering, Nagoya University

Feb. 1984 Dr. of Engineering, Nagoya University

### **Professional Experience:**

1982-1987 Assistant Professor, Department of Synthetic Chemistry, Nagoya University

1987-1993 Lecturer, Department of Synthetic Chemistry, Nagoya University

1994-1999 Associate Professor, The Institute for Solid State Physics, The University of Tokyo

1999-2020 Professor, Department of Applied Chemistry, Nagoya University

2004- 2007 Presidential Advisor, Nagoya University

2007- 2013 Vice-President, Nagoya University

2012- 2013 Dean, Graduate School of Pharmaceutical Sciences, Nagoya University

2013- 2015 Dean, Graduate School of Engineering, Nagoya University

2015- 2019 Trustee and Vice-President, Nagoya University

2020- Research Fellow, Toyota Physical and Chemical Research Institute

### **Awards and Honors:**

1999 Wiley Polymer Science Award, 1999 Physics

2007 Journal of Polymer Science, 2007 Polymer Physics Award

2007 The Award of the Society of Polymer Science, Japan

### **Research Interest**

Morphology Control of Complex Polymer Systems

- Crystalline Symmetry and Quasi-crystalline Symmetry

New Self-assembly Manners of Complex Polymers with Supramolecular Interactions

## Research Achievements

As described in the research proposal section, block polymers with incompatible components naturally form periodic patterns in condensed state. These multiphase structures vary their morphologies depending on molecular parameters such as block chain connectivity and the degree of molecular asymmetry. This section introduces and describes the principal achievements brought by Matsushita's research group. The common feature of each section from the viewpoint of molecules is "frustration included in the system" introduced variously by complex molecular design. The main text is written along with slides attached.

### 1. The superlattice structures created by symmetric ABC linear terpolymers (slide #1)

The centered B block chains in ABC linear block terpolymers have to adopt bridge conformation in bulk and the B chains are forced to be suffered from weak stress, and hence they naturally give featured structures. A variety of compositional change is possible, but here we focus on "symmetric" molecules whose volume fractions of A and C chains are the same each other. Polymer species adopted are **I(polyisoprene), S(polystyrene) and P(poly(2-vinylpyridine))**, and many ISP terpolymers were prepared based on the anionic polymerizations under vacuum. The morphologies observed for symmetric terpolymers, whose volume fractions of I and P block are almost the same, are shown in the slide #1. Four structures, i.e., three-phase four-layered lamellar structure, **tricontinuous gyroid structure**, a cylindrical one with tetragonal symmetry and spherical structure of the CsCl type, can be seen from left to right with increasing the fraction of the center S block. It should be noting the fact that all these have the feature of **the superlattice structures**, in which I and P domains can be switched mutually.

### 2. Tricontinuous double diamond structure from binary blends of linear ABC triblock terpolymers (slide #2)

As displayed in the slide #2, two asymmetric ISP terpolymers were prepared and the structures of their binary blends were observed, where the overall compositions were designed to keep symmetry. **Four-branched double diamond structure** was stably observed by realizing weak localization of two molecules on the phase-separated surface, in contrast to the three-branched double gyroid structure for monodisperse symmetric molecules as introduced in the previous section. This structure is conceived to be formed to earn the conformational entropy for the end block chains by sacrificing both the surface energy and junction-placement entropy. This symmetric but unusual morphology was created under weak stress due to the present molecular design.

### 3. Periodic and aperiodic tiling patterns from ABC star molecules (slides #3 and #4)

If each end of three different polymer chains are connected at one junction point, we can get **ABC star molecules**, whose junction points have to be aligned on lines not on surfaces, and therefore block chains are restricted to stay within the very frustrated spaces. And they naturally form rod-based morphology. If we explore to check the order the cross-sectional view, we naturally meet the two-dimensional space-sharing patterns, i.e., tilings. The slide #3 shows schematic expression of the self-assembly manner of this type of molecules, and several examples of the observed patterns are assembled on the right, which includes many **periodic Archimedean tiling patterns** and also an aperiodic pattern at the center. This characteristic one in a pentagon has the feature of **aperiodicity with dodecagonal symmetry** as shown in the slide #4 in detail. This non-classic ordered pattern was first discovered in metal alloy in 1982 by Prof. D. Shechtman, who was honored as a single Nobel Prize laureate in chemistry section in 2011, whereas our discovery of quasi-crystal with mesoscopic tile size in 2007 was in the memorial year of silver jubilee of Shechtman's first discovery in 1982.

#### **4. Non-classical structures in tetrablock terpolymers of the $A_1BA_2C$ type (slide #5)**

Tetrablock terpolymers of the  $A_1BA_2C$  type naturally hold frustration in molecules themselves since  $A_1$  and  $A_2$  have to play different roles in self-assembly, and so forth they represent featured morphologies. The slide #5 displays the double hexagonal and tetragonal structures from  $S_1IS_2P$  terpolymers at the left-hand side, while **the 3.3.4.3.4 AT structure** and another **quasicrystalline tiling with dodecagonal symmetry** from well-annealed binary blends of terpolymers are compared at the right-hand side of the slide. These non-classical structures have been found to appear covering wide composition region, indicating that these are evidently thermodynamically stable structures. These results are owing to the frustrated molecular systems adopted in the present study.

## Yushu Matsushita, List of publications

### 1) Reviewed articles

1. "Preparation and Morphological Properties of a Triblock Copolymer of the ABC Type"

Y. Matsushita, H. Choshi, T. Fujimoto, M. Nagasawa

*Macromolecules* **13**, 1053-1058, (1980).

2. "Preparation and Characterization of Block Copolymers of Ordinary and Deuterated Styrenes"

Y. Matsushita, H. Furuhashi, H. Choshi, I. Noda, M. Nagasawa, T. Fujimoto, C.C. Han  
*Polym. J.* **14**, 489-493(1982).

3. "Morphologies of ABC-Type Triblock Copolymers with Different Compositions"

Y. Matsushita, K. Yamada, T. Hattori, T. Fujimoto, Y. Sawada, M. Nagasawa, C. Matsui

*Macromolecules* **16**, 10-13(1983).

4. "Expansion Factor of a Part of a Polymer Chain in a Good Solvent Measured by Small-Angle Neutron Scattering"

Y. Matsushita, I. Noda, M. Nagasawa, T.P. Lodge, E.J. Amis, C.C. Han

*Macromolecules* **17**, 1785-1789(1984).

5. "Preparation and Characterization of Poly(2-vinylpyridine)s with Narrow Molecular Weight Distributions"

Y. Matsushita, K. Shimizu, Y. Nakao, H. Choshi, I. Noda, M. Nagasawa

*Polym. J.* **18**, 361-366(1986).

6. "Studies of Styrene and 2-Vinylpyridine Block Copolymers; Preparation and Characterization"

Y. Matsushita, Y. Nakao, R. Saguchi, H. Choshi, M. Nagasawa

*Polym. J.* **18**, 493-499(1986).

7. "Conformations of Diblock Copolymers in Dilute Solutions"

Y. Matsushita, Y. Nakao, K. Shimizu, I. Noda, M. Nagasawa

*Macromolecules* **21**, 2790-2793(1988).

8. "Phase Contrast Matching in Lamellar Structures Composed of Mixtures of Labeled and Unlabeled Block Copolymers for Small-Angle Neutron Scattering"

Y. Matsushita, Y. Nakao, R. Saguchi, K. Mori, H. Choshi, Y. Muroga, I. Noda, M. Nagasawa, T. Chang, C.J. Glinka, C.C. Han

*Macromolecules* **21**, 1802-1806(1988).

9. "Molecular Weight Dependence of Lamellar Domain Spacing of Diblock

Copolymers in Bulk"

Y. Matsushita, K. Mori, R. Saguchi, Y. Nakao, I. Noda, M. Nagasawa

*Macromolecules* **23**, 4313-4316(1990).

10. "Chain Conformation of a Block Polymer in a Microphase-Separated Structure"

Y. Matsushita, K. Mori, Y. Mogi, R. Saguchi, I. Noda, M. Nagasawa, T. Chang, C.J. Glinka, C.C. Han

*Macromolecules* **23**, 4317-4321(1990).

11. "Chain Conformations and Locations of Parts of a Block Polymer in a Lamellar Structure"

Y. Matsushita, K. Mori, R. Saguchi, I. Noda, M. Nagasawa, T. Chang, C.J. Glinka, C.C. Han

*Macromolecules* **23**, 4387-4391(1990).

12. "Chain Conformation of Block Copolymers in Dilute Solutions Measured by Small-Angle Neutron Scattering"

Y. Matsushita, K. Shimizu, I. Noda, T. Chang, C.C. Han

*Polymer*, **23**, 2412-2415(1992).

13. "Preparation and Morphology of Triblock Copolymers of the ABC Type"

Y. Mogi, H. Kotsuji, Y. Kaneko, K. Mori, Y. Matsushita, I. Noda

*Macromolecules*, **25**, 5408-5411(1992).

14. "Tricontinuous Morphology of Triblock Copolymers of the ABC Type"

Y. Mogi, K. Mori, Y. Matsushita, I. Noda

*Macromolecules*, **25**, 5412-5415(1992).

15. "Molecular Weight Dependence of Lamellar Domain Spacing and Chain Conformation of ABC Triblock Copolymers in Microphase-separated Structure."

Y. Mogi, K. Mori, H. Kotsuji, Y. Matsushita, I. Noda, C.C. Han

*Macromolecules* **26**, 5169-5173(1993).

16. "Localization of a Homopolymer Dissolved in a Lamellar Structure of a Block Copolymer Studied by Small-Angle Neutron Scattering"

Y. Matsushita, Y. Mogi, N. Torikai, I. Noda, C.C. Han

*Macromolecules* **26**, 6346-6349(1993).

17. "Preparation and Morphology of Multiblock Copolymers of the (AB)<sub>n</sub> Type"

Y. Matsushita, J. Watanabe, Y. Mogi, H. Mukai, I. Noda

*Polymer* **35**, 246-249(1994).

18. "Tricontinuous Double-Diamond Structure Formed by a Styrene-Isoprene-2-Vinylpyridine Triblock Copolymer"

Y. Matsushita, M. Tamura, I. Noda

- Macromolecules* **27**, 3680-3682(1994).
19. "Preparation and Morphologies of 4-Arm and 12-Arm Styrene-Isoprene Star-Shaped Block Copolymers"  
Y. Matsushita, T. Takasu, K. Yagi, K. Tomioka, I. Noda  
*Polymer* **35**, 2862-2866(1994).
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Y. Matsushita, Y. Mogi, K. Mori, I. Noda, C.C. Han  
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21. "Super Lattice Structure in Morphology of the ABC Triblock Copolymers"  
Y. Mogi, H. Kotsuji, M. Nomura, K. Ohnishi, Y. Matsushita, I. Noda  
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22. "Alternating Lamellar Structure of Triblock Copolymers of the ABA Type"  
Y. Matsushita, M. Nomura, J. Watanabe, Y. Mogi, I. Noda, M. Imai  
*Macromolecules* **28**, 6007-6013(1995).
23. "Preparation and Characterization of ABB Graft Copolymers"  
Y. Matsushita, J. Watanabe, F. Katano, Y. Yoshida, I. Noda  
*Polymer* **37**, 321-325(1996).
24. "Morphologies and Domain Sizes of Microphase-Separated Structures of Block and Graft Copolymers of Different Types"  
Y. Matsushita, I. Noda, N. Torikai  
*Macromol. Symp.* **106**, 121-133(1997).
25. "Lamellar Domain Spacing of the ABB Graft Copolymers"  
Y. Matsushita, H. Momose, Y. Yoshida, I. Noda  
*Polymer* **38**, 149-153(1997).
26. "Neutron Reflection Studies on Segment Distribution of Block Chains in Lamellar Microphase-separated Structures"  
N. Torikai, I. Noda, A. Karim, S.K. Satija, C.C. Han, Y. Matsushita, T. Kawakatsu  
*Macromolecules* **30**, 2907-2914(1997).
27. "Lamellar Domain Spacings of Diblock Copolymer/Homopolymer Blends and Conformations of Block Chains in Their Microdomains"  
N. Torikai, N. Takabayashi, I. Noda, S. Koizumi, Y. Morii, Y. Matsushita  
*Macromolecules* **30**, 5698-5703(1997).
28. "Order-Disorder Transition of Symmetric Poly(styrene-*b*-2-vinylpyridine) in Bulk and Solution"  
Y. Takahashi, S. Kitade, M. Noda, N. Ochiai, I. Noda, M. Imai, Y. Matsushita

- Polym. J.* **30**, 388-393(1998).
29. "Chain Dimensions of Mid-blocks of ABA Triblock Copolymers with Lamellar Structures in Bulk"  
Y. Matsushita, J. Suzuki, N. Takabayashi, N. Torikai, M. Nomura, I. Noda  
*Macromolecules* **30**, 2378-2380(1998).
30. "Surfaces of Tricontinuous Structures formed by ABC Triblock Copolymers in Bulk"  
Y. Matsushita, J. Suzuki, M. Seki  
*Physica B*, 248, 238-242(1998).
31. "Concentration Dependence of Radius of Gyration of Sodium Poly(styrene sulfonate) over a Wide Range of Concentration Studied by Small-Angle Neutron Scattering"  
Y. Takahashi, N. Matsumoto, S. Iio, H. Kondo, I. Noda, M. Imai, Y. Matsushita  
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32. "Ring Structure of Cyclic Poly(2-vinylpyridine) Proved by Pyrolysis-GC/MS"  
H. Ohtani, H. Kotsuji, H. Momose, Y. Matsushita, I. Noda, S. Tsuge  
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33. "Miscibility of Isotactic Polypropylene/Ethylene-Propylene Random Copolymer Binary Blends"  
M. Seki, H. Nakano, S. Yamauchi, J. Suzuki, Y. Matsushita  
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34. "Small-angle X-ray Scattering Analysis of the Periodic Tricontinuous Network Structure of Symmetric ABC Triblock Copolymers"  
M. Seki, J. Suzuki, Y. Matsushita  
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35. "The Tricontinuous Double-Gyroid Structure from a Three-component Polymer System"  
J. Suzuki, M. Seki, Y. Matsushita  
*J. Chem. Phys.* **112**, 4862-4868(2000).
36. "Studies on Equilibrium Structures of Complex Polymers in Condensed Systems"  
Y. Matsushita  
*J. Polym. Sci., Part B, Polym. Phys. Ed.* **38**, 1645-1655(2000).
37. "Study on the Thermodynamic Interactions between Isotactic Polypropylene and Ethylene-hexene Random Copolymers by SANS"  
M. Seki, H. Uchida, Y. Maeda, S. Yamauchi, K. Takagi, Y. Ukai, Y. Matsushita  
*Macromolecules* **33**, 9712-9719(2000)

38. “Mesoscopic patterns of block and graft copolymers in condensed systems” Y. Matsushita  
*Macromol. Symp.* **160**, 151-158 (2000).
39. “Preparation and Morphology of Model Graft Copolymers of the A<sub>3</sub> B<sub>2</sub> Type with Different Graft Junction Points”  
 A. Takano, K. Kondo, M. Ueno, K. Ito, S. Kawahara, Y. Isono, J. Suzuki, Y. Matsushita  
*Polym. J.* **33**, 732-740(2001)
40. Morphology of ABC Triblock Copolymer/Homopolymer Blend System  
 Jiro Suzuki, Motofumi Furuya, Minobu Iinuma, Atsushi Takano, Yushu Matsushita  
*J.Polym.Sci. Polym. Phys.Ed.* **40**, 1135-1141(2002)
41. Preparation and Characterization of Cyclic Polystyrene with Short Poly (2-*tert*-butyl-butadiene) sequence  
 A. Takano, A. Nonaka, O. Kadoi, K. Hirahara, S. Kawahara, Y. Isono, N. Torikai, Y. Matsushita  
*J.Polym.Sci., Polym. Phys.Ed.* **40**, 1582-1589(2002)
42. Preparation and Morphology of Ring-shaped Polystyrene-*block*-polyisoprenes  
 D. Takano, O. Kadoi, K. Hirahara, S. Kawahara, Y. Isono, J. Suzuki, Y. Matsushita  
*Macromolecules* **36**, 3045-3050(2003)
43. Effect of Composition Distribution on Microphase-separated Structure from Diblock Copolymers  
 Y. Matsushita, M. Iinuma, A. Noro, J. Suzuki, H. Ohtani, A. Takano  
*Macromolecules* **36**, 8074-8077(2003)
44. Observation of Four Phase Lamellar Structure from a Tetrablock Copolymer of the ABCD Type  
 A. Takano, K. Soga, T. Asari, J. Suzuki, S. Arai, H. Saka, Y. Matsushita  
*Macromolecules* **36**, 8216-8218 (2003).
45. Non-centrosymmetric Structure with Nanoscopic Periodicity from a Tetrablock Copolymer of the ABCA Type  
 A. Takano, K. Soga, J. Suzuki, Y. Matsushita,  
*Macromolecules* **36**, 9288-9291 (2003).
46. Effect of Composition Distribution on Microphase-Separated Structure from BAB Triblock Copolymers  
 A. Noro, M. Iinuma, J. Suzuki, A. Takano, Y. Matsushita  
*Macromolecules* **37**, 3804-3808(2004).
47. Chain Elongation Suppression of Cyclic Block Copolymers in Lamellar Microphase-Separated Bulk  
 Y. Matsushita, H. Iwata, T. Asari, T.Uchida, G. ten Brinke, A. Takano



- J. Chem. Phys.* **121**, 1129-1132(2004)
48. Preparation of Partially Deuterium-labelled Poly(4-trimethylsilylstyrene)s and Their Unperturbed Dimensions in Bulk  
M.Harada, T. Suzuki, M.Ohya, A. Takano, Y. Matsushita  
*Polymer J.* **38**, 538-541(2004)
49. Observation of Cylinder-based Microphase-separated Structures from ABC Star-shaped Terpolymers Investigated by Electron Computerized Tomography  
A. Takano, S. Wada, S. Sato, T.Araki, T. Kazama, S. Kawahara, Y. Isono, A. Ohno, N.Tanaka, Y. Matsushita  
*Macromolecules* **37**, 9941-9946(2004)
50. Self Assembly Template During Morphological Transition of a Linear ABC Triblock Copolymer from Lamellar to Gyroid Structure  
J.Suzuki, K. Nakane, A. Takano, Y. Matsushita  
*Polymer* **45**, 8989-8997(2004).
51. Nobel Miscible Polymer Blend of Poly(4-trimethylsilylstyrene) and Polyisoprene  
M. Harada, T. Suzuki, M.Ohya, A.Takano, Y. Matsushita  
*Macromolecules*, **38**, 1868-1873(2005).
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M. Harada, M. Ohya, T. Suzuki, D. Kawaguchi, A. Takano, Y.Matsushita  
*J. Polym. Sci. Polym. Phys.* **43**,1214-1219(2005).
- 53.TGIC Separation of PS-b-P2VP Diblock & P2VP-b-PS-b-P2VP Triblock Copolymers According to Chemical Composition  
D. Cho, A. Noro, A. Takano, Y. Matsushita  
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M. Harada, T. Suzuki, M. Ohya, D.Kawaguchi, A. Takano, Y. Matsushita, N. Torikai  
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55. Effect of Molecular Weight Distribution on Microphase-separated Structures from Block Copolymers  
A. Noro, D. Cho, A. Takano, Y. Matsushita  
*Macromolecules* **38**, 4371 – 4376(2005).
56. Preparation and Characterization of Cyclic Polystyrenes  
D.Cho, K.Masuoka, K. Kohguchi, T. Asari, A. Takano, Y. Matsushita  
*Polymer J.* **37**, 506 – 511(2005)
57. A Mesoscopic Archimedean Tiling Having a New Complexity in an ABC Star Polymer

- B. Takano, W. Kawashima, A. Noro, Y. Isono, N. Tanaka, T. Dotera, Y. Matsushita  
*J. Polym. Sci. Polym. Phys. Ed.* **43**, 2427-2432(2005).
58. Novel Synthesis and Characterization of Bioconjugate Block Copolymers Having Oligonucleotides  
A. Noro, Y. Nagata, M. Tsukamoto, Y. Hayakawa, A. Takano, Y. Matsushita  
*Biomacromolecules* **6**, 2328-2333(2005)
59. Three Phase Hierarchical Structures from AB/CD Diblock Copolymer Blends with Complemental Hydrogen Bonding Interaction  
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A. Takano, I. Kamaya, Y. Takahashi, Y. Matsushita  
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61. Effect of Loop/Bridge Conformation Ratio on Elastic Properties of the Sphere-Forming A-B-A Triblock Copolymers under Uniaxial Elongation  
Y. Takahashi, Y. Song, N. Nemoto, A. Takano, Y. Akazawa, and Y. Matsushita,  
*Macromolecules* **38**, 9724-9729(2005).
62. Preparation and Characterization of a Styrene-Isoprene Undecablock Copolymer and Its Hierarchical Microdomain Structure in Bulk  
Y. Nagata, J. Masuda, A. Noro, D. Cho, A. Takano, Y. Matsushita  
*Macromolecules* **38**, 10220-10225 (2005).
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T. Asari, S. Matsuo, A. Takano, Y. Matsushita  
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T. Asari, S. Arai, A. Takano, Y. Matsushita  
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65. A Diblock-Type Supramacromolecule via Biocomplementary Hydrogen Bonding  
A. Noro, Y. Nagata, A. Tanaka, Y. Matsushita  
*Biomacromolecules* **7**, 1696-1699(2006).
66. Archimedean Tiling Patterns of ABC Star-shaped Terpolymers Studied by Micro-beam Small Angle X-ray Scattering  
K. Hayashida, W. Kawashima, A. Takano, Y. Shinohara, Y. Amemiya<sup>2</sup> Y. Nozue,

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D.Kawaguchi, K. Masuoka, A. Takano, K. Tanaka, T. Nagamura, N. Torikai, R. M. Dalgliesh, S. Langridge, Y. Matsushita

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J. Masuda, Y.Nagata, A. Noro, A. Takano, Y. Matsushita

*Phys. Rev. Lett.* **97**, 098301(2006).

69. Chain Localization and Interfacial Thickness in Microphase-Separated Structures of Block Copolymers with Variable Composition Distributions

A. Noro, M. Okuda, F. Odamaki, D. Kawaguchi, N. Torikai, A. Takano, Y. Matsushita

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K. Hayashida, A. Takano, S. Arai, Y. Shinohara, Y. Amemiya, Y. Matsushita

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Y. Nozue, T. Sakurai, H. Hozumi, T. Kasahara, N. Yamaguchi, M. Shibayama, Y. Matsushita

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73. The Creation of Hierarchically-Ordered Nanophase Structures In Block Polymers Having Various Competing Interactions

Y. Matsushita

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74. Polymeric Quasicrystal: Mesoscopic Quasicrystalline Tiling in ABC Star Polymers

K. Hayashida, A. Takano, T. Dotera, Y. Matsushita

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K. Hayashida, A. Takano, N. Tanaka, Y. Matsushita  
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75. Composition-Dependent Morphological Transition of Hierarchically-Ordered Structures formed by Multiblock Terpolymers  
J. Masuda, A. Takano, J. Suzuki, Y. Nagata, A. Noro, K. Hayashida, Y. Matsushita  
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A. Takano, W. Kawashima, S. Wada, K. Hayashida, S. Sato, K. Hirahara, S. Kawahara, Y. Isono, N. Tanaka, D. Kawaguchi and Y. Matsushita  
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N. Torikai, N. Yamada, A. Noro, M. Harada, D. Kawaguchi, A. Takano, Y. Matsushita  
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78. Temperature Dependence of Surface Segregation in Miscible Polymer Blend of Poly(4-trimethylsilylstyrene)/Polyisoprene  
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- 84 Nanophase-Separated Structures of AB block Copolymer/C Homopolymer Blends with Complimentary Hydrogen Bonding Interaction  
 K. Dobrosielska, S. Wakao, A. Takano, Y. Matsushita  
*Macromolecules* **41**, 7695-7698(2008)
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