# Experimental Investigation of the Morphology Development and Mechanical Properties of Waste Ethylene Propylene Diene Monomer/Polypropylene Blend in Modular Intermeshing Corotating Twin-Screw Extruder

JIN KUK KIM, SUNG HYO LEE, SUNG HYUK HWANG

Department of Polymer Science and Engineering, Research Institute of Industrial Technology, Gyeongsang National University, Chinju, Gyeongnam 660-701, South Korea

*Received 27 August 2001; accepted 11 December 2001*

**ABSTRACT:** Nowadays, with the increase in the number of automobiles, waste EPDM (ethylene propylene diene monomer) is causing a significant environmental problem. From environmental and economical perspectives, recycling is one of the popular methods to solve environmental problems. This study, which involved waste EPDM/PP (polypropylene) blends with the ratio range of 70/30 and 75/25, set out to ascertain the relevance of the mass percentage of the dispersed phase, the influence of the screw geometry, the screw rpm, and the melting temperature of PP materials on the morphology and mechanical properties of the waste rubber blend. The purpose of this study is to develop a high-value thermoplastic elastomer from waste EPDM. This investigation concentrated on determining the optimum conditions for producing a blend by extrusion, relative to screw geometry, screw rational speed, and operating temperature.
 J Appl Polym Sci 85: 2276–2282, 2002

**Key words:** screw configuration; waste EPDM/PP blend; high-value thermoplastic elastomers, recycling

## INTRODUCTION

Commercial polyblends consist of at least two polymeric components. They allow the combination of favorable properties of known polymers as well as the development of new quality characteristics.[1] In general, the development of a new polyblend is far more cost-effective than that of a new polymer.[2]

During the processing of polyblends and depending on material combination, the mostly single-phase starting materials form different microstructures inside the solid.

Blends of polypropylene (PP) with ethylene propylene diene monomer (EPDM) have been widely used as engineering polymeric materials. Blends with a high content of PP have been used as a high-impact PP, whereas blends with a high content of EPDM have been used as thermoplastic elastomers. Many studies[3–6] reported that the dynamic vulcanization of EPDM during melt mixing with PP can improve some properties of high-impact PP.

The barrel and screw configurations as well as the processing conditions such as screw speeds and temperature strongly affect the phase morphology and mechanical properties of the blends. These dependencies were experimentally ascertained relative to various screw configurations of the extruder in this work. A substantial reduction

*Correspondence to:* J. Kim.

Journal of Applied Polymer Science, Vol. 85, 2276–2282 (2002)

**Table I Characteristics of the Polymers Used in This Work**

| Material | Properties | Source |
|---|---|---|
| Random co-PP | MFI = 4.5 g/10 min | Grade R930Y (SK Chemical Co., Korea) |
| | $M_w$ = 200,000 | |
| | Density = 0.9 g/cm$^3$ | |
| | Melting point = 152°C | |
| Waste EPDM powder | Size = 5–20 $\mu$m | Cooling crush method (Dong-a Hwasung Co., Korea) |
| | Density = 1.057 g/cm$^3$ | |
| | Component | |
| | C = 90.09% | |
| | H = 8.42% | |
| | N = 0.122% | |
| | S = 0.066% | |

of the waste EPDM particle size in PP matrix can be observed in the melting zone of the extruder. The final morphology and mechanical properties of the blend are determined by the design of the geometry loads to the flow and mixing conditions as well as the screw speeds.

## EXPERIMENTAL

### Materials

The characteristics of the polymers used in this study are summarized in Table I. The PP was the commercial extrusion grade of SK Chemicals (R930Y) and Korea Petrochemical (1088). The waste EPDM powder was obtained by the crush method from the vulcanized EPDM compound.

### Blending

The waste EPDM/PP blends were prepared as 25/75 and 30/70 ratios, as shown in Table II. We used a corotating intermeshing twin-screw extruder ($D$ = 19 mm, $L/D$ = 40) and five different screw configurations, which are shown in Figure 1. Such configurations involve different combinations of right-handed and left-handed screws and neutral kneading disc elements. The geometrical parameters and dimension data of the kneading block of the used extruder are given in Table III. The radial clearance was the distance between the cylinder inner diameter and the tip of the kneading block (0.6 mm). The screw speeds were varied at 50, 70, 100, and 150 rpm in a corotating twin-screw extruder. The cylinder temperature was maintained at 200, 220, 230, and 235°C from the hopper to the die.

### Testing

#### *Morphology*

The morphology was investigated by using a scanning electron microscope (SEM); (Philips XL 30S; Holland) after sputtering the samples with gold (fine coat sputter, JFC-1100E; JEOL, Peabody, MA). The surface analysis was studied by using cryogenically fractured etched samples in $p$-xylene.

**Table II Formulation and Blend Conditions of Waste EPDM/PP Blend**

| Waste EPDM/PP Blend Ratio | | | |
|---|---|---|---|
| Waste EPDM | PP | Barrel Temperature | Screw rpm |
| 75 | 25 | 200–235°C | 150 |
| 70 | 30 | | 100 |
| | | | 70 |
| | | | 50 |

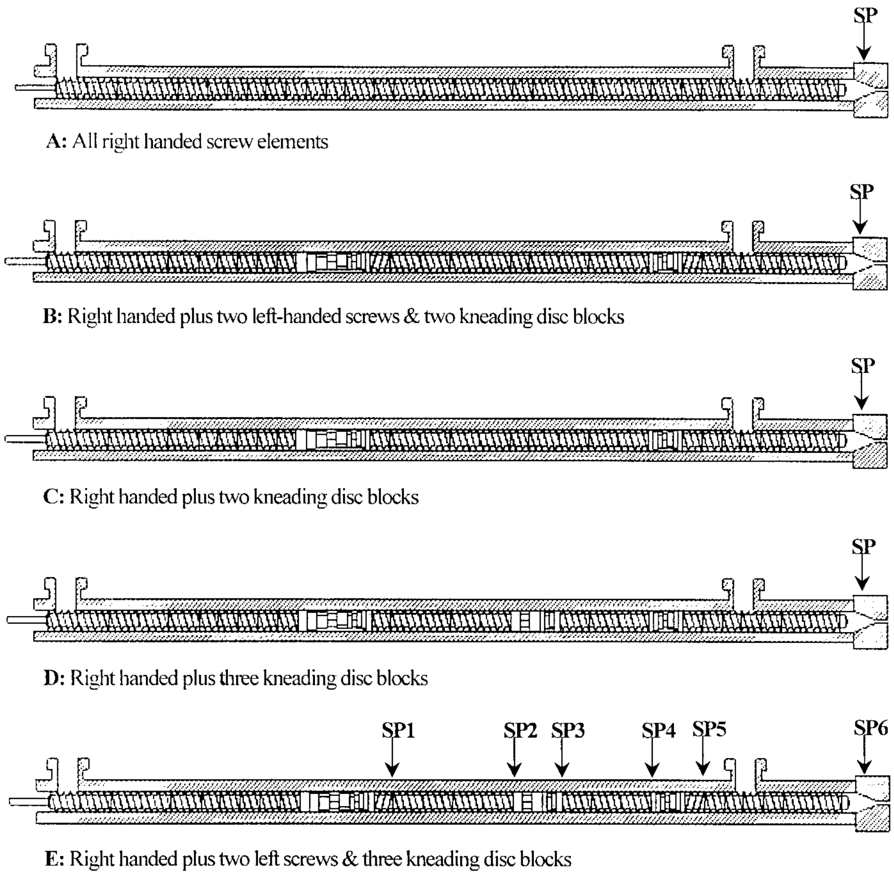


**Figure 1** Screw configurations with sampling points (SPs). (A) All right-handed screw elements. (B) Right-handed plus two left-handed screws and two kneading disc blocks. (C) Right-handed screw plus two kneading disc blocks. (D) Right-handed screw plus three kneading disc blocks. (E) Right-handed plus two left-handed screws and three kneading disc blocks.

### *Mechanical and Rheological Properties*

The extrudates were pealletized and then injection molded. The test specimens were prepared according to ASTM standards [dumbbell-shape samples for tensile test (ASTM D412) and elongation test (ASTM D412)]. The temperature of the injection-molding cylinder was kept at 235°C, the mold temperature was 35–40°C, and the injection pressure was maintained at 2000–2400 psi. The mechanical properties (tensile strength and elongation at break) were determined by using a Lloyd LR10K tensile testing machine (RHC, Inc., Ontario, Canada). The

**Table III Geometry and Dimension Data of the Kneading Block**

| | First Kneading Block Set | | Second Kneading Block Set | | Third Kneading Block Set | |
|---|---|---|---|---|---|---|
| Item | L[a] | S[b] | L[a] | S[b] | L[a] | S[b] |
| Number of kneading disc | 5 | 5 | 3 | 4 | — | 7 |
| Paddle orientation | 30° | 30° | 90° | 30° | — | 30° |

[a] L; 9.55-mm kneading disc.
[b] S; 4.75-mm kneading disc.

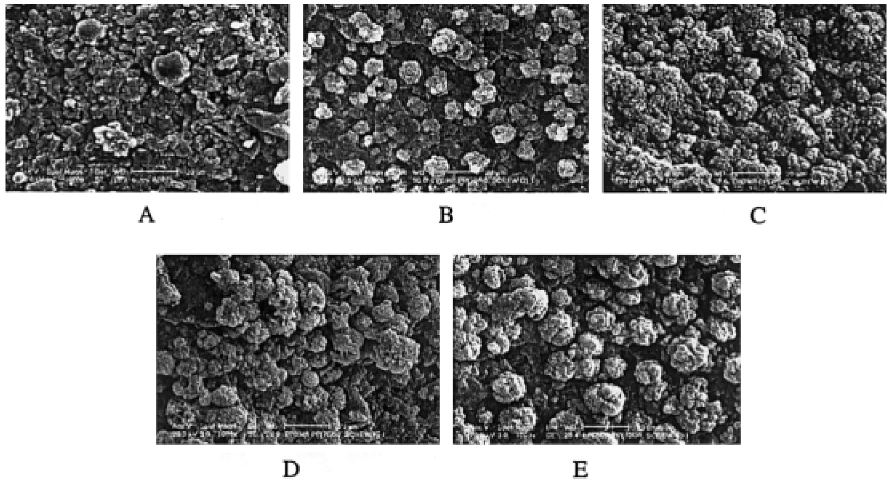

**Figure 2** SEM micrographs of composite formed by dynamic vulcanization in extruder: (A) screw configuration A; (B) screw configuration B; (C) screw configuration C; (D) screw configuration D; (E) screw configuration E.

crosshead speed was 500 mm/min for tensile measurements and a load cell of 10 kN capacity was used.

The rheological properties of the samples were studied using a capillary rheometer Model Galaxy V8052 (Kayeness, Inc., Pennsylvania) with the length and diameter of 0.591 and 0.0276 in., respectively. The *L/D* of the capillary was 20 and measurements were carried out at 250°C.

### Swelling Experiment

Samples of 0.1 to 0.3 g were placed in *n*-heptane for 48 h to swell. The swollen piece was quickly removed from the *n*-heptane and excess *n*-heptane was blotted off, after which the sample was weighed in a stoppered preweighed vial, deswollen in a 60°C vacuum oven for 3 h, and weighed again.

We calculated the number of crosslinking chains per unit volume to determine the crosslinking density by eq. (1):

$$\upsilon_e = -[\ln(1 - V_R) + V_R + \chi_1 V_R^2] \div [V_1 (V_R^{1/3} - V_R/2)] \quad (1)$$

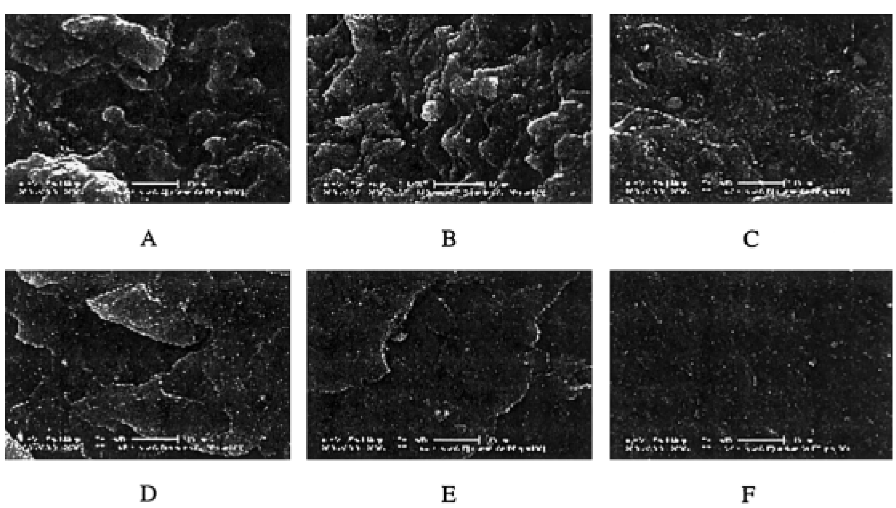

**Figure 3** Morphology development for sampling points (SPs) of screw configuration E: (A) SP1; (B) SP2; (C) SP3; (D) SP4; (E) SP5; (F) SP6.

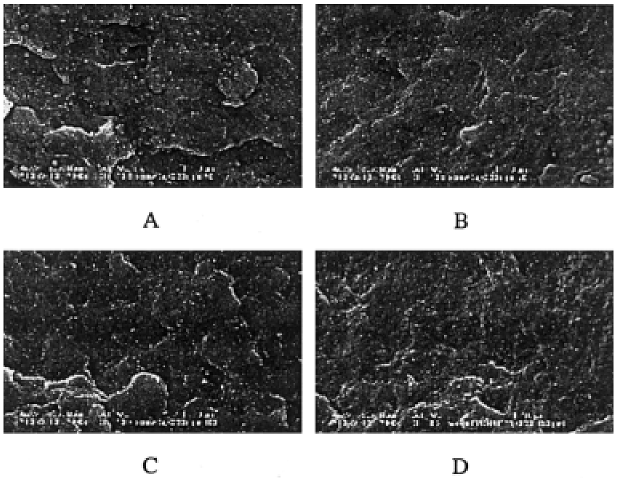


**Figure 4** SEM micrographs of samples prepared at different screw speeds in screw configuration E: (A) 50 rpm; (B) 70 rpm; (C) 100 rpm; (D) 150 rpm.

where $\upsilon_e$ is the effective number of chains in a real network per unit volume (mol/cm$^3$), $V_1$ is the molecular volume of solvent (cm$^3$/mol), $V_R$ is the volume fraction of rubber in the swollen state, and $\chi_1$[7] is the parameter expressing the first-neighbor interaction free energy.

## RESULTS AND DISCUSSION

### Morphology

Different screw configurations result in different blend morphologies.

Figure 2 shows the SEM micrographs with different screw configurations of the waste EPDM/PP (70/30) blend. The results indicate that the better morphology of blends is shown in screw configurations B and E because of the kneading disc block and left-handed screw element in their screw configurations. The kneading disc or left-handed screw induced a complicated flow, which leads to good dispersion.

Figure 3 shows morphology changes at different sampling points of screw configuration E, which is shown in Figure 1(E). We found that the interface between two materials disappeared from the SP4 position. The results indicated that the screw configurations of well-dispersed samples at long residence times show good results.

The screw speed is one of the important variables relative to blend properties. Figure 4 shows the morphology changes of screw configuration E with blend EPDM/PP (70/30) and different reactions. Figure 4 tells us that different rates of dynamic vulcanization occurred with the different screw speeds. The best morphology at screw configuration E for the same blend [EPDM/PP (70/30)] shows the screw rpm of 100 rather than at 50, 70, or 150 rpm. The reason is that a screw rpm of 100 induces a higher shear rate than at the 50 and 70 rpm screw speeds and higher residence time at the 150 rpm screw speed.

### Mechanical Properties

The tensile strength elongation at break and the 50% modulus with different screw configurations are shown in Figure 5. The better mechanical properties are also shown in configurations B and

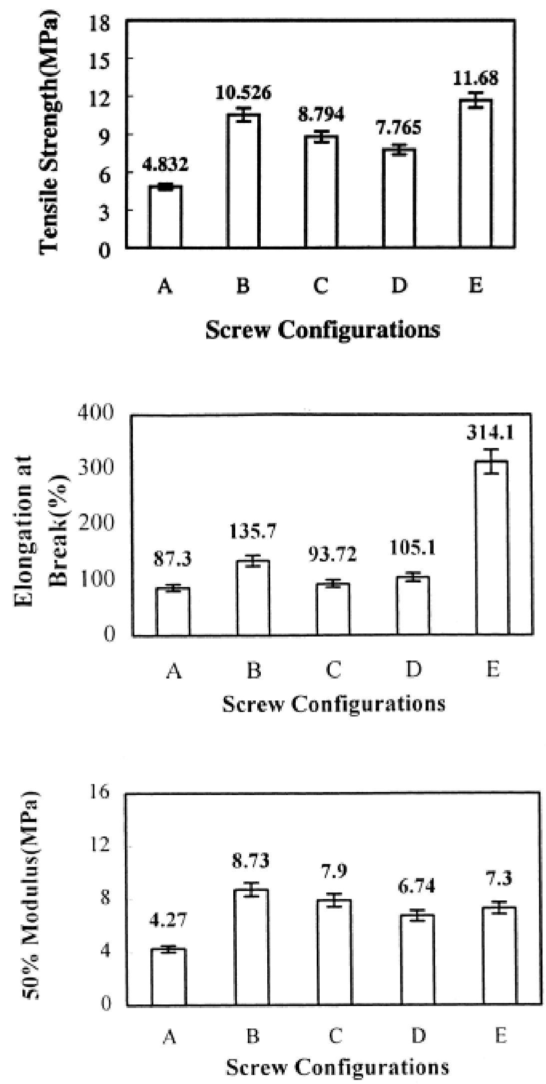


**Figure 5** Mechanical properties of the composite as a function of screw configuration.

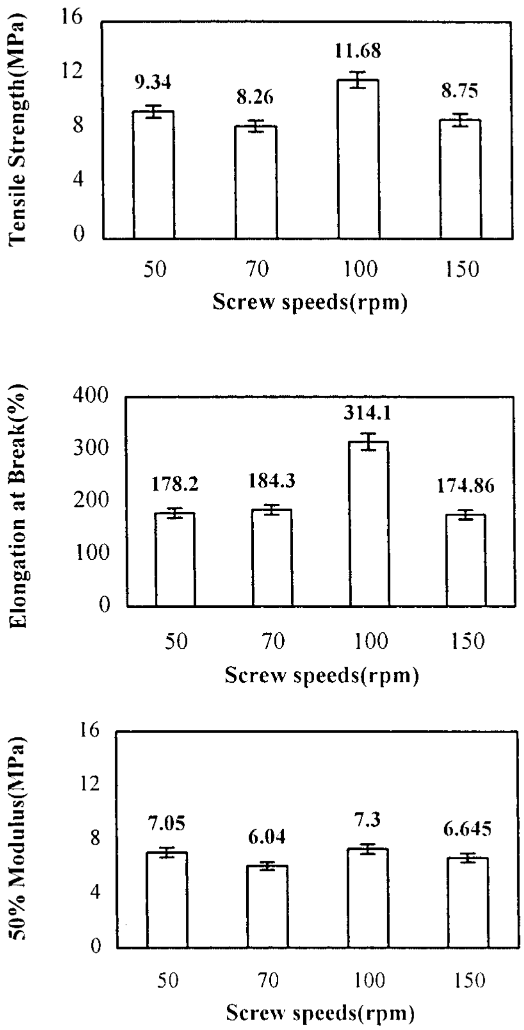


**Figure 6** Mechanical properties of samples prepared at different screw speeds in screw configuration E.

E than in other screw configurations. These results indicated that the mechanical properties were affected by shear stress and residence time during the process.

Figure 6 shows the mechanical properties with different screw speeds in the case of screw configuration E. The blends containing 30 wt % PP at 100 rpm showed better tensile strength, elongation at break, and 50% modulus compared to those of other samples that were compounded at 50, 70, and 150 rpm (see Table IV). The reason is that not only is there a higher shear rate at 100 rpm than the shear rate at either 50 or 70 rpm but there is also a longer residence time compared to that at a screw speed of 150 rpm.

**Table IV Mechanical Properties of Waste EPDM/PP Blends of Different Blend Ratios**

| Blend Ratio | Tensile Strength (MPa) | Elongation at Break (%) | 50% Modulus (MPa) |
|---|---|---|---|
| 75/25 | 11.11 | 321 | 6.0 |
| 70/30 | 11.68 | 314 | 7.3 |

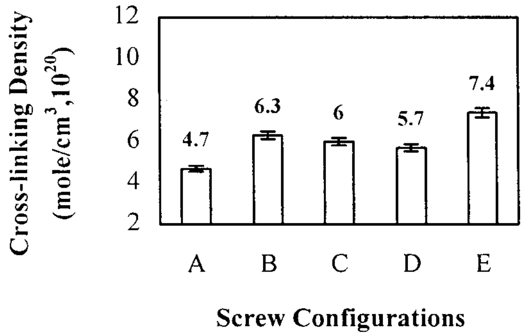


**Figure 7** Crosslinking density of the composite as a function of screw configuration.

## Crosslinking Density

Figures 7 and 8 show the crosslinking density of blends at various screw configurations and at various screw speeds in screw configuration E, respectively.

The crosslinking density decreases with increase of shear stress and decrease of residence time during the process because of mechanochemically produced radicals under the high-intensity blending condition.

## Rheology

The effect of the screw configurations and blend ratio on the rheological behavior of the waste

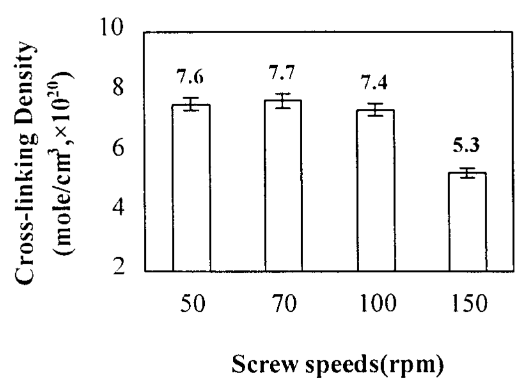


**Figure 8** Crosslinking density of samples prepared at different screw speeds in screw configuration E.

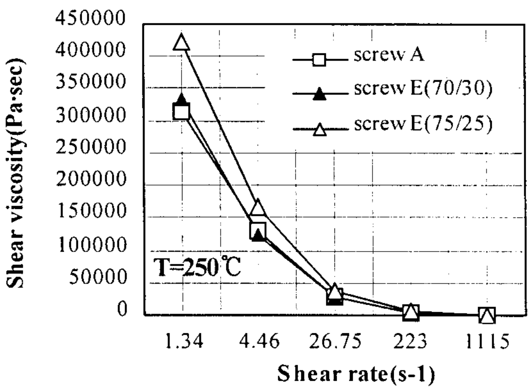


**Figure 9** Shear viscosity with shear rate of waste EPDM/PP blends for different screw configurations and blend ratios at 100 rpm.

EPDM/PP blends is shown in Figure 9. The figure shows that the shear viscosity strongly depends on the waste EPDM/PP blend ratio rather than on screw configuration. It is obvious that the thermoplastic elastomer vulcanizate shows shear thinning behavior, which follows the power law model over the entire range of the studied shear rates. These are attributed to the formation of crosslinks between the rubber chains within the rubber particles, which increases their stability toward shear breakdown during mixing and, therefore, less reduction in the shear viscosity of the blends.

## CONCLUSIONS

The purpose of this study is to recycle the waste EPDM powder by blending with PP. Therefore, we set out to determine the optimum conditions of extrusion relative to screw configurations and screw speed. We investigated the morphology and mechanical properties (tensile strength, elongation at break, and 50% modulus) of the waste EPDM/PP blends to ascertain the optimum conditions. From experimental results, the best conditions are shown in screw configuration E, which contains two left-handed screws and three kneading disc blocks for the best conditions for dynamic vulcanization. In screw configuration E, the best screw speed is at 100 rpm. These results indicate that the properties strongly depend on the dynamic vulcanization during blending. This study shows a possibility for producing TPR (thermoplastic rubber) from waste rubber.

## REFERENCES

1. N/N, Aufbereiten von Polymerblends; VDI: Düsseldorf, 1982.
2. Utracki, L. A. Polymer Alloys and Blends: Thermodynamics and Rheology; Hanser Verlag: Munich, 1989.
3. Dao, K. C. Polymer 1984, 25, 1527.
4. Kresge, E. N.; Lohse, D. J.; Datta, S. Makromol Chem Macromol Symp 1992, 53, 173.
5. Krulis, Z.; Fortenlny, I.; Kovar, J. Collect Czech Chem Commun 1993, 58, 2642.
6. Inouse, T. J Appl Polym Sci 1994, 54, 723.
7. Bandrup, J.; Immergut, E. H. Polymer Handbook, 3rd ed.; Wiley: New York, 1989; p 180.