

# Melting Aspects of Filled Compounds in A Modular Co-rotating Twin Screw Extruder

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Received 4 January 2005; accepted 28 March 2006

DOI 10.1002/app.24578

Published online in Wiley InterScience (www.interscience.wiley.com).

**ABSTRACT:** In the polymer industry, precompounded materials are widely used. Carefully designed melting experiments were carried out to investigate melting mechanisms in master batched polymer compounds, using an intermeshing co-rotating twin screw extruder. Calcium carbonate or aluminum powder was master batched with linear low-density polyethylene (LLDPE). The calcium carbonates, which were used in the compounds, have differences in weight fraction and particle size. The compounds containing

filler have higher thermal conductivities and viscosities than neat polyethylene. We observed melting initiation and propagation mechanisms of LLDPE compounds by removal and characterization of polymer compound carcasses in the melting region. © 2006 Wiley Periodicals, Inc. *J Appl Polym Sci* 102: 1990–2012, 2006

**Key words:** twin screw extruder; melting mechanism; compound; filler

## INTRODUCTION

In 1959, Maddock<sup>1</sup> made the first careful melting experiments in single screw extruders in detail. In 1966, Tadmor<sup>2</sup> modeled melting based on Maddock's melting mechanism. There were many models of melting in single screw extruders in the literature from that year to the 2000s.

Melting studies<sup>3–11</sup> in twin screw extruders started from the 1990s. Todd<sup>3</sup> observed melting on the kneading disc block elements in 1992. Potente<sup>4</sup> suggested the first theoretical model of melting based on the theory of single screw extruders in 1993. Bawiskar and White<sup>7,8</sup> observed melting initiation from the barrel surface and melting growth from the barrel surface. These melting experiments had been done using barrel temperatures over the crystalline melting temperature or critical melt flow temperature at the barrel surface. Gogos et al.<sup>9,10</sup> have proposed that it is plastic deformation energy that enables pellets to melt in co-rotating twin screw extruders.

We have done systematic experiments by varying the barrel temperature, screw speed, and feed rate<sup>11</sup>. We related melting trends to barrel temperatures and screw speeds. The operating conditions of their melting experiments cover a range from below the crystalline melting temperature to above the crystalline melting temperature. We observed barrel melting at temperatures above the crystalline melting point and at

lower temperatures, pellet deformation melting, as well as, screw melting initiation that occurred at low temperatures and screw speeds. Diagrams of melting mechanism transition regimes were drawn. We also described mechanisms of melting propagation.

All studies of melting in twin screw extruders have involved neat thermoplastics. Here, we present the first study of the melting of thermoplastics containing fillers.

## EXPERIMENTAL

### Materials

Melting experiments were carried out for four compounds of LLDPE. These were prepared by mixing LLDPE with aluminum flakes or calcium carbonate, using a co-rotating twin screw extruder.

First, we premixed LLDPE with 10 vol % of a filler, using the JSW co-rotating twin screw extruder. We made three different LLDPE compounds for aluminum flakes, 0.07- $\mu\text{m}$  size calcium carbonate, and 3.0- $\mu\text{m}$  size calcium carbonate. This is equivalent to 25 wt % of aluminum or calcium carbonate. One additional compound we studied contained 20 vol % of 0.07- $\mu\text{m}$  calcium carbonate. This is equivalent to 43 wt % of calcium carbonate. We cooled the filaments emerging from the extruder by a water bath and cut them into pellets, using a pelletizer. The pellets had dimensions of 3.0 mm height and 2.6 mm diameter.

The specifications and thermal properties of the materials used are shown in Tables I and II.

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**TABLE I**  
Thermal Properties of LLDPE and Fillers

	Density (g/cm <sup>3</sup> )	Specific heat capacity (J/(g K))	Thermal conductivity (W/(m K))
Aluminum	2.7	0.921	225.94
CaCO <sub>3</sub>	2.7	0.837	4.393
LLDPE	0.9	2.092	0.331

### Thermal properties of materials

To obtain the thermal properties of the fillers aluminum and calcium carbonate, we used the data from Ref. 12 as a reference. Key data have been summarized in Table I.

The order of magnitude difference in conductivity between aluminum and calcium carbonate is two as shown in Table II. Even though the order of thermal heat conductivity of fillers is quite different, the difference of the order of thermal heat conductivity of these compounds is small as shown in Table II, where we have calculated the thermal conductivities by Hamilton and Crosser's equation<sup>13</sup> for the thermal conductivity of heterogeneous, two-component systems.

$$k = k_1 \left[ \frac{k_2 + (n-1)k_1 - (n-1)v_2(k_1 - k_2)}{k_2 + (n-1)k_1 + v_2(k_1 - k_2)} \right] \quad (1)$$

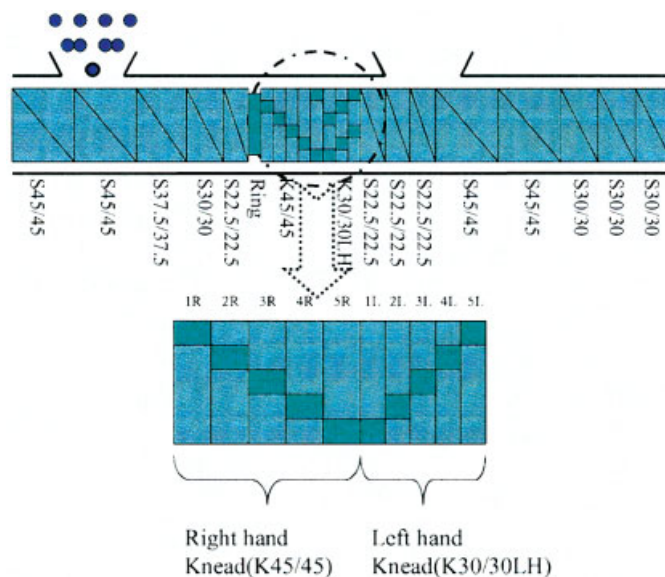
where  $k_1$  is the thermal conductivity of the matrix LLDPE,  $k_2$  is that of fillers,  $v_2$  is the volume fraction of filler, and  $n$  is the shape factor.

We calculated the other properties for compounds as shown in Table II, using the following equations:

$$c_p = w_1c_1 + w_2c_2 \quad (2)$$

$$1/\rho = w_1/\rho_1 + w_2/\rho_2 \quad (3)$$

$$\alpha = \frac{k}{\rho c_p} \quad (4)$$



**Figure 1** Screw configuration and the numbered disc of kneading blocks. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

where  $w_i$  is the weight fraction of an ingredient,  $c_i$  and  $c_p$  are the specific heat capacity, and  $\rho_i$  and  $\rho$  are the density,  $\alpha$  is the thermal diffusivity, and  $k$  is the thermal conductivity. The density of compounds is increased by larger amounts of filler, but the specific heat is reduced by the increased amounts of filler.

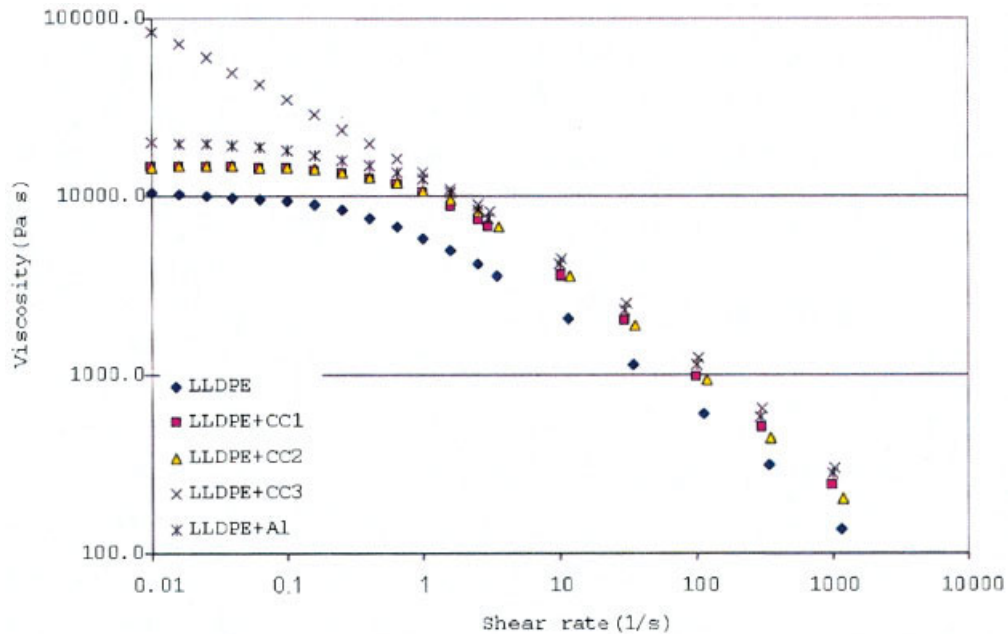
### Shear viscosity of compounds

The shear viscosity was determined from using capillary and cone-plate data at 140, 160, and 180°C as shown in Figures 1–3. The shear viscosity of the compound containing 43 wt % of calcium carbonate is the highest. The shear viscosity of the neat LLDPE is the lowest. The shear viscosity of the other two compounds containing 20 wt % of calcium carbonate is intermediate. For these two compounds, the shear viscosity of the small size of the calcium carbonate compound is somewhat lower than that of the large

**TABLE II**  
Thermal Properties of LLDPE and its Compounds

	Density, $\rho$ (g/cm <sup>3</sup> ) at 20°C	Specific heat capacity, $C_p$ (J/(g K))	Thermal conductivity, $k$ (W/(m K))	Thermal diffusivity, $\alpha$ (m <sup>2</sup> /s)
LLDPE	0.9	2.092	0.331	0.176E-6
LLDPE + A1	1.08	1.799	0.549	0.283E-6
LLDPE + CC1 or CC2	1.08	1.788	0.417	0.216E-6
LLDPE + CC3	1.26	1.554	0.520	0.266E-6

A1, 10 vol % of aluminum flakes; CC1, 10 vol % of calcium carbonate (ViCALity Heavy PCC, 3  $\mu$ m); CC2, 10 vol % of calcium carbonate (Multiflex, 0.07  $\mu$ m); CC3, 20 vol % of calcium carbonate (Multiflex, 0.07  $\mu$ m).



**Figure 2** Shear viscosity–shear rate relationships for neat LLDPE and its compounds used in this study at 140 °C. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

size of the calcium carbonate compound. The effect on shear viscosity by the smaller particle is larger.

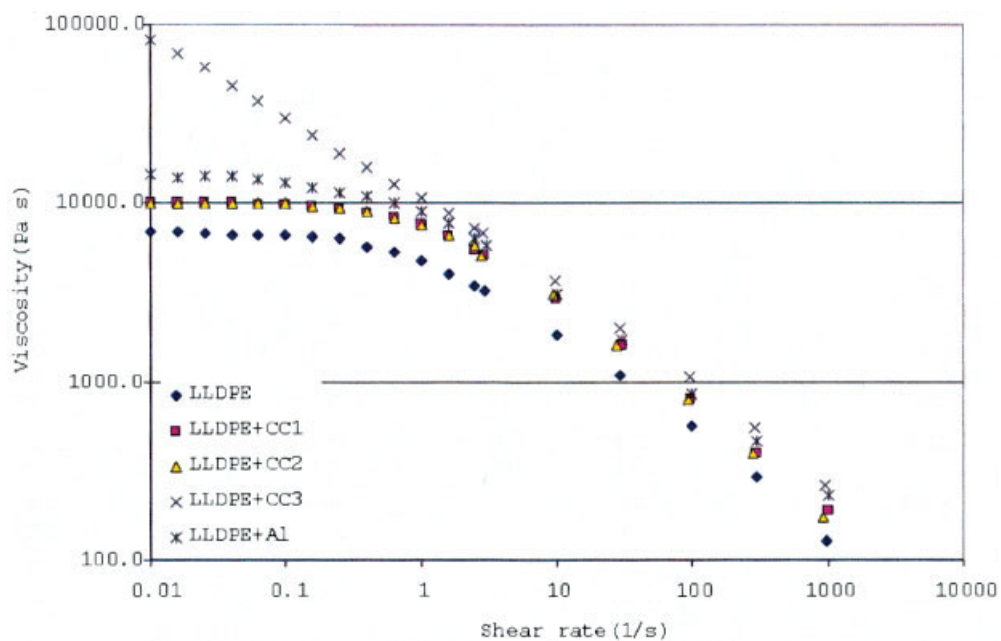
used for investigation of melting mechanisms is shown in Figure 4.

### Apparatus

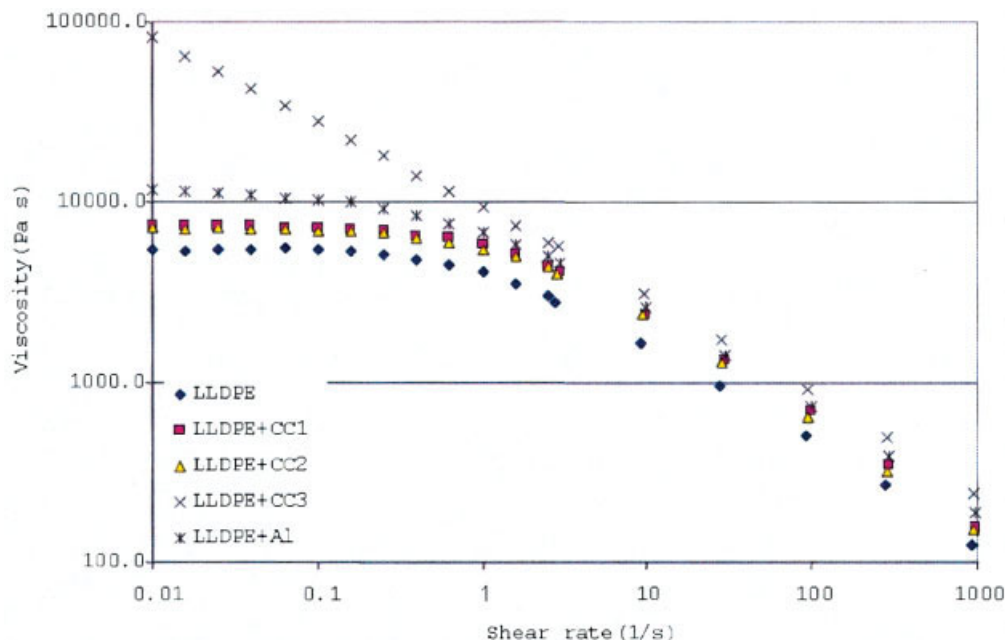
A co-rotating twin screw extruder (Japan Steel Works Labo Tex 30 mm) was used.<sup>11</sup> The screw configuration

### Procedure

In the melting experiments, the solid pellets were fed to the extruder at room temperature. The same melt-



**Figure 3** Shear viscosity–shear rate relationships for neat LLDPE and its compounds used in this study at 160 °C. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]



**Figure 4** Shear viscosity–shear rate relationships for neat LLDPE and its compounds used in this study at 180 °C. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

ing mechanisms with the melting mechanisms of neat polyolefins were observed as a function of barrel temperature and screw speed (Table IV). Generally barrel temperatures of 130, 110, and 70°C were used. The melting temperature of the LDPE compounds is 122°C. In melting experiments, the extruder was shut off and cooled down by cooling water, after the operating status of the extruder reached to a steady state. Carcasses were sectioned in the melting region and removed from the twin screw extruder, and digital photographs of carcasses were taken to characterize melting mechanisms. Before the carcass was removed from the twin screw extruder, the melting length was measured to study the melting effects by different types, amounts, and sizes of filler particles.

## RESULTS

Melting of polymer compounds containing aluminum flakes or calcium carbonate is initiated earlier than for

neat LLDPE under the conditions of our study. These polymer compounds were melted fully in the right-handed kneading disc block in front of the left-handed kneading disc block under most experimental conditions.

Neat LLDPE's digital photographs are shown in Figure 5.

### Aluminum flake compound

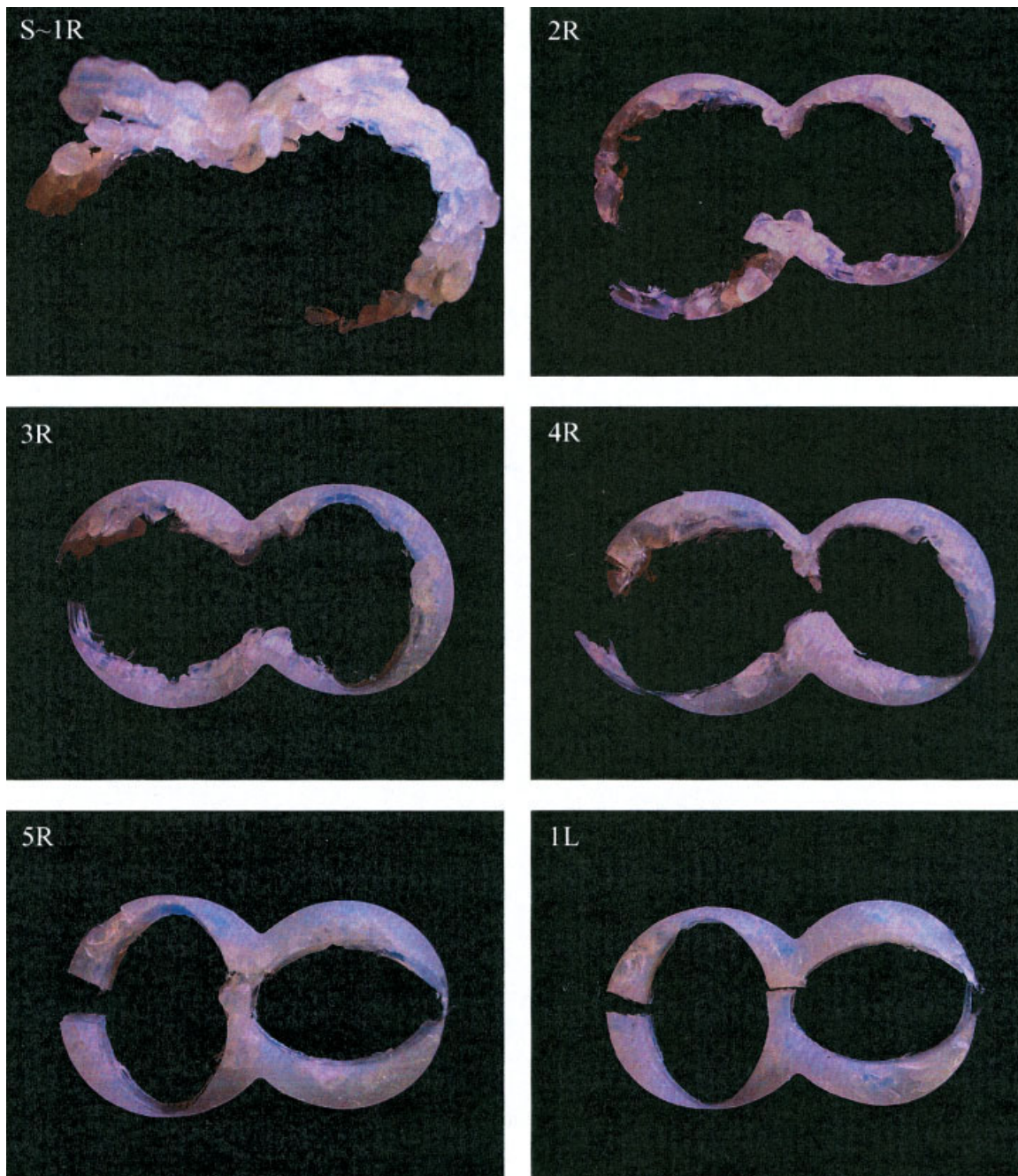
Here, we describe our melting experiments on LLDPE compounds containing 10 vol % aluminum flakes. For the barrel temperature 70°C, we did three experiments at three screw speeds: 50, 150, and 250 rpm. At 50 rpm, we observed screw melting initiation and propagation. The pellets observed on the barrel side were deformed throughout the screw elements. A melt layer was seen on the screw side, which became thicker in succeeding carcasses. At the screw speed 150 rpm (Fig. 6), tightly packed pellets are seen in the beginning of the carcass on the modular screw. The left side digital photograph at 3R shows less deformed

**TABLE III**  
Parameters of the Linear Relations Between Yield Stress and Inverse Kelvin Temperature for LLDPE, PP, PS, LLDPE + A1, LLDPE + CC1, LLDPE + CC2, and LLDPE + CC3

	Slope (Pa/K)	Intercept (Pa)
LLDPE	1.248E10	-2.992E07
PP	1.565E10	-3.772E07
PS	2.575E10	-6.259E07
LLDPE + A1	5.965E09	-1.425E07
LLDPE + CC1	5.894E09	-1.372E07
LLDPE + CC2	5.978E09	-1.398E07
LLDPE + CC3	7.825E09	-1.905E07

**TABLE IV**  
Melting Experimental Conditions

Compound	Feed rate (kg/h)	Barrel	
		temperature (°C)	Screw speed (rpm)
LLDPE + A1	6.0	70, 110, 130	50, 150, 250
LLDPE + CC1	6.0	70, 110, 130	50, 150, 250
LLDPE + CC2	6.0	70, 110, 130	50, 150, 250
LLDPE + CC3	6.0	70, 110, 130	50, 150, 250

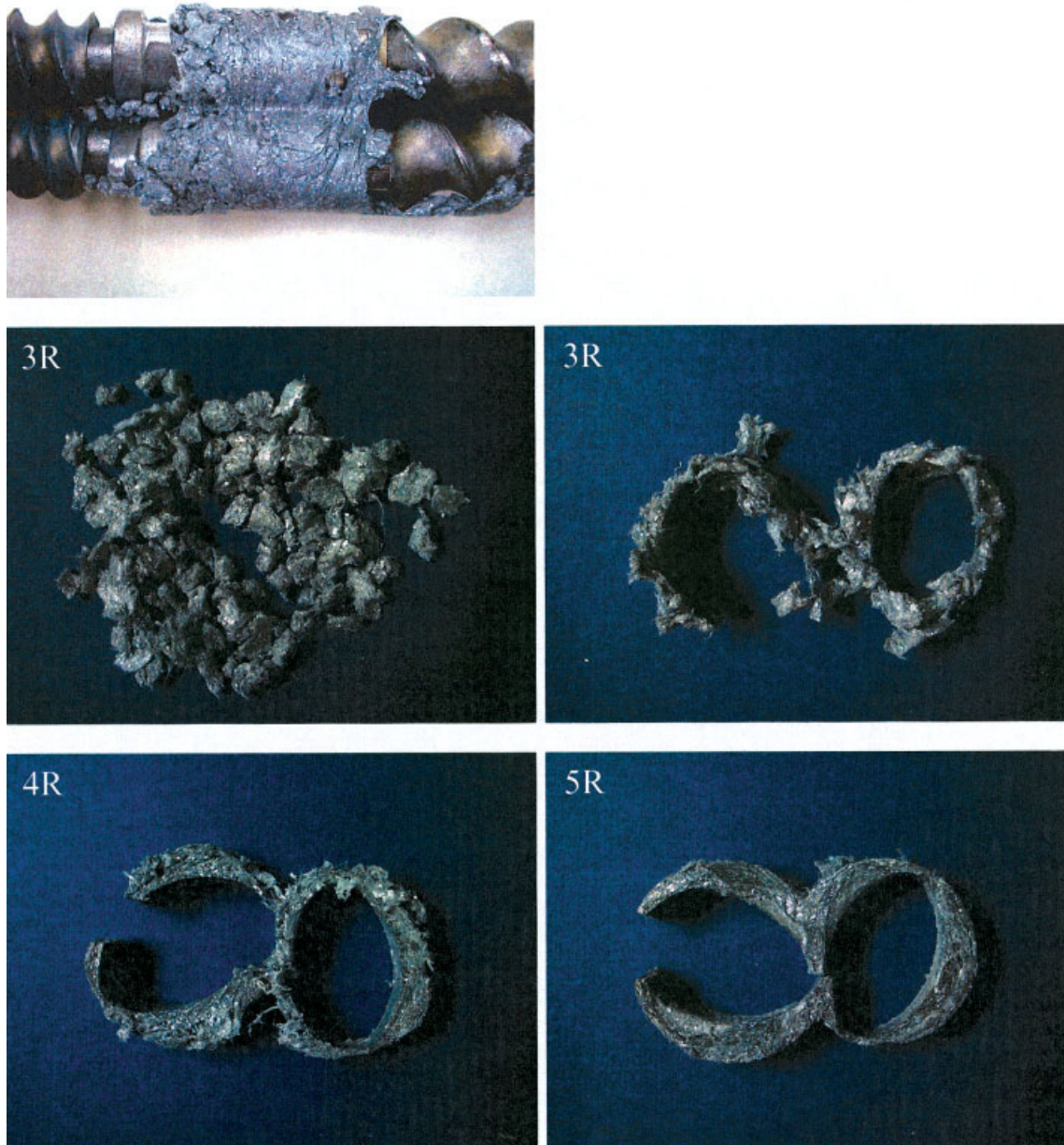


**Figure 5** Sectioned carcasses of LLDPE at feed rate 7.2 kg/h, barrel temperature 130°C, screw speed 50 rpm. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

pellets than those at 50 rpm, and the right side digital photograph at 3R shows clustered deformed pellets toward the screw surface. The digital photograph at 4R shows a growing melt layer from the screw surface. These indicate a melting initiation and propagation from the screw surface. At the screw speed 250 rpm, we obtained the same results for the melting initiation as at the lower screw speeds. At this screw speed, pellets melted very fast and deformed very little.

At the barrel temperature 110°C, we also did experiments at screw speeds 50, 150, and 250 rpm. At this

barrel temperature, all of the experiments for the aluminum compound suggest internal bulk melting initiation and propagation. At the screw speed 50 rpm, pellets did not exhibit as much deformation as experiments at 70°C and at 50 rpm. Melting was initiated between pellets, and the bed with deformed pellets and melt was observed. In Figure 7, at 150 rpm, we also observed internal melting initiation that was in compacted and clustered pellets with melt together at 3R. At 4R, there is more melt around the screw surface than between pellets. In Figure 8, we show digital



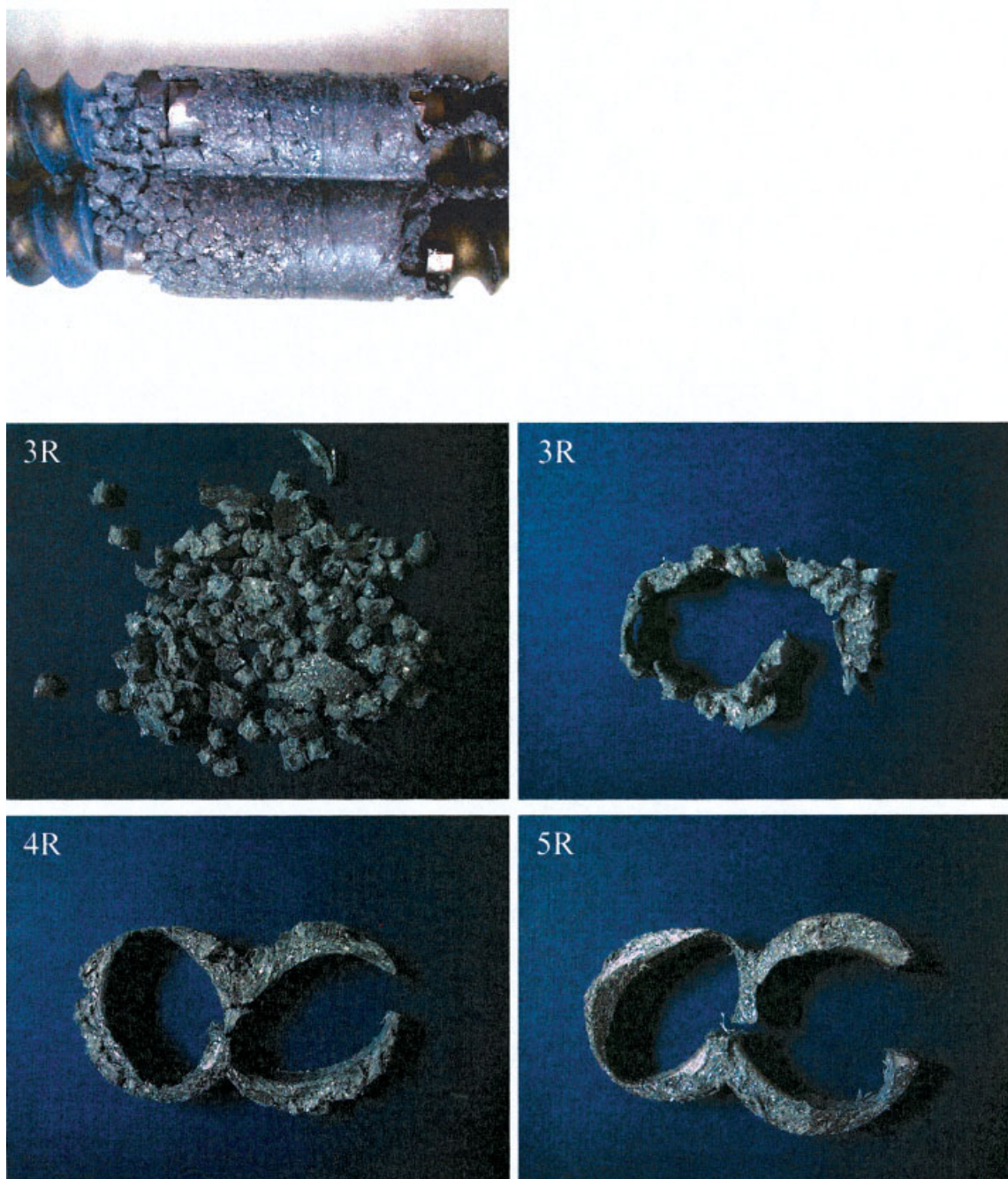
**Figure 6** The sectioned carcasses of master batched compound with 10 vol % of aluminum flakes at feed rate 6.0 kg/h, barrel temperature 70°C, screw speed 150 rpm. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

photographs at 250 rpm. The digital photograph on the modular screw shows pellets enter the kneading disc block zone in their original shape and then these begin to be compacted. Then, melt appears between pellets at 4R. This melting occurred rapidly.

For the barrel temperature 130°C, we observed barrel melting initiation and two different melting propagation mechanisms, i.e., propagation from the barrel side and bed instability. As described for neat LLDPE earlier and in our earlier article,<sup>11</sup> at 50 rpm, the digital photograph of the carcass on the modular screw shows melt and the digital photograph at the location

of 1R shows aluminum compound pellets fused near the barrel surface (Fig. 9). The aluminum compound pellets gradually disappeared from the barrel surface in the succeeding sectioned carcasses. This means that melting is initiated and propagated from the barrel. At 150 rpm, the melt was initiated by barrel melting and propagated by the same mechanism. At 250 rpm, pellets seem to be melted by the barrel melting and the bed instability mechanism. There is a very short melting length, when compared with the observations at 50 and 150 rpm.

We plot the melting initiation and propagation mechanisms of experimental results of aluminum



**Figure 7** The sectioned carcasses of master batched compound with 10 vol % of aluminum flakes at feed rate 6.0 kg/h, barrel temperature 110°C, screw speed 150 rpm. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

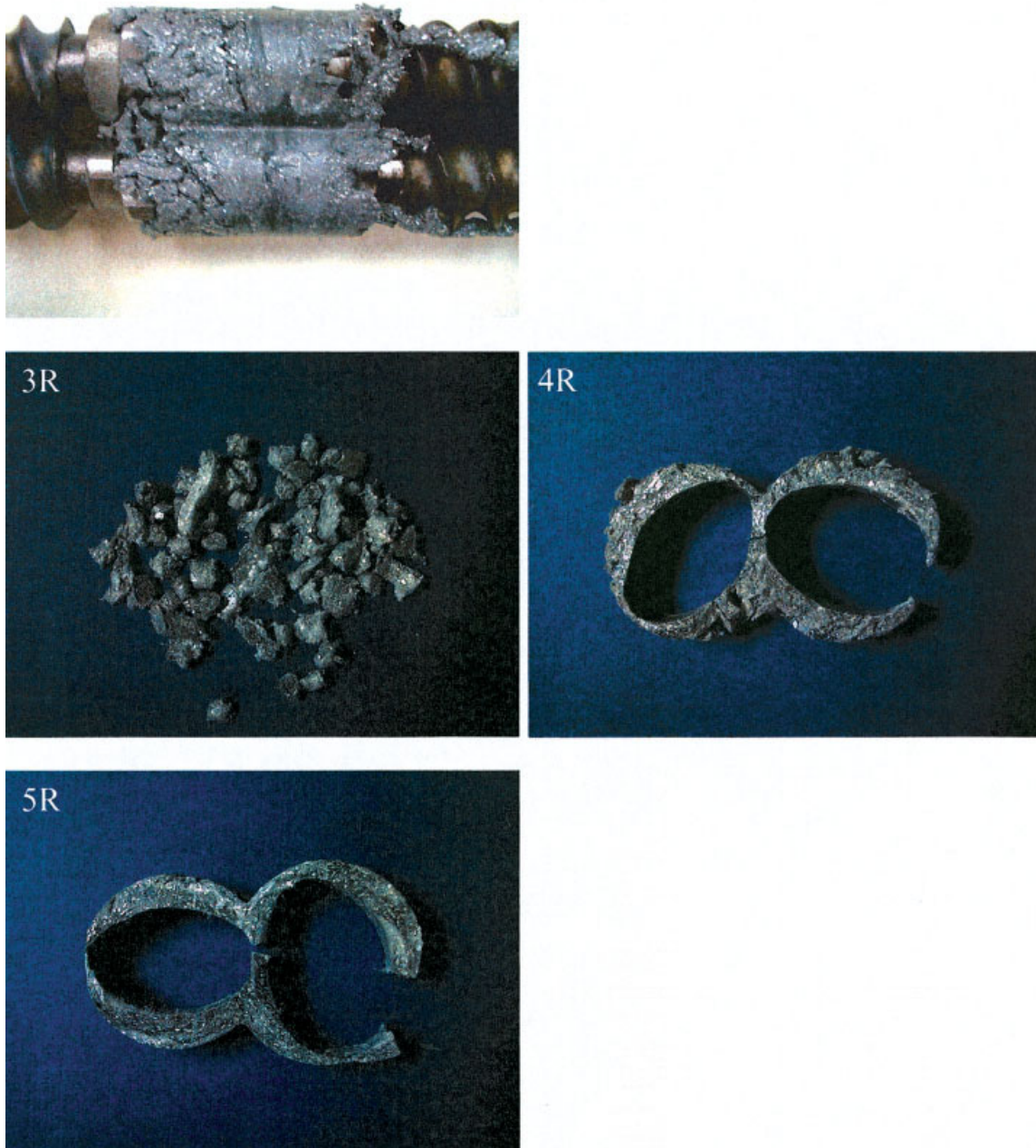
compound in Figure 10. This shows three melting initiation and four melting propagation mechanisms according to screw speeds and barrel temperatures.

#### Larger size particle calcium carbonate compound

Figures 11–13 show sectioned carcasses indicating typical melting initiation and propagation mecha-

nisms of the LLDPE compound with 10 vol % of calcium carbonate (ViCALity Heavy, 3  $\mu\text{m}$ ).

For the barrel temperature 70°C, we experimented at three screw speeds: 50, 150, and 250 rpm. At 50 rpm, melting was initiated from the screw surface and propagated in the same manner. In contrast to the experiments on aluminum compound at 70°C and 50 rpm, the calcium carbonate compound melted com-



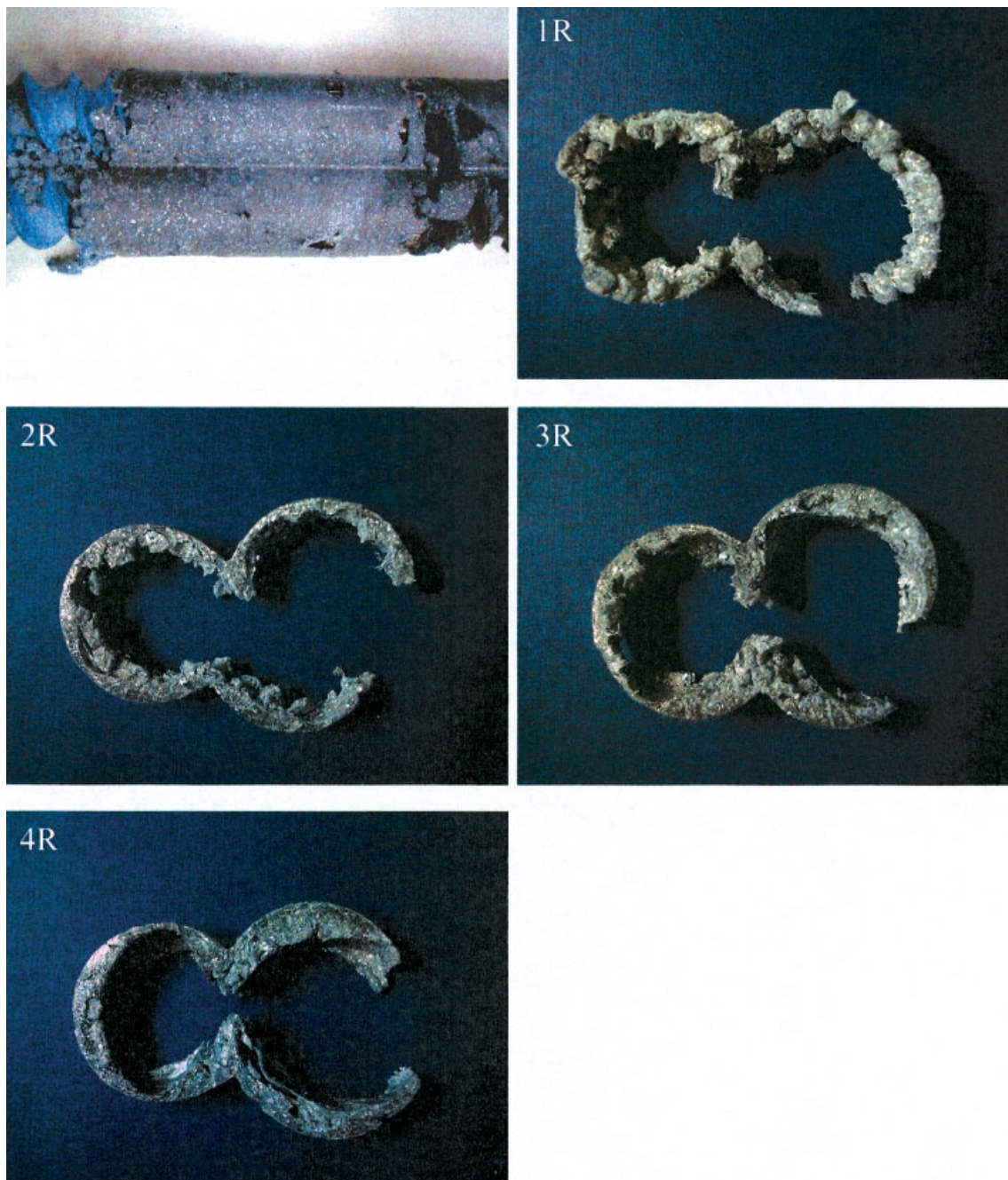
**Figure 8** The sectioned carcasses of master batched compound with 10 vol % of aluminum flakes at feed rate 6.0 kg/h, barrel temperature 110°C, screw speed 250 rpm. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

pletely in front of the left-handed kneading disc block element. In Figure 11, deformed pellets are shown in the digital photograph of the carcass on the modular screw at 150 rpm. Deformed pellets are found at the locations of 2R, 3R, and 4R. In these locations, we observed deformed and clustered pellets around the screw surface at 3R and a growing melt layer at 3R and 4R. These resulted in melting initiation from the screw surface and gradual melt layer growth from the screw surface. At the screw speed 250 rpm, we also observed the same melting initiation and propagation mechanism.

Figure 12, where the barrel temperature ( $T_b$ ) is 110 °C and the screw speed ( $N$ ) is 50 rpm, the digital photograph at the location of 2R shows us clustered melt and deformed pellets in the bed. Successive digital photographs indicate that melting is developing in the solid bed and melting being propagated in a bed mixed with pellets and melt. At screw speeds 150 and 250 rpm, we also observed the same bulk melting initiation and propagation mechanism.

In Figure 13, which represents behavior at  $T_b = 130^\circ\text{C}$  and  $N = 50$  rpm, melt is seen in the carcass on the modular screw. The melt layer grows grad-





**Figure 9** The sectioned carcasses of master batched compound with 10 vol % of aluminum flakes at feed rate 6.0 kg/h, barrel temperature 130°C, screw speed 50 rpm. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

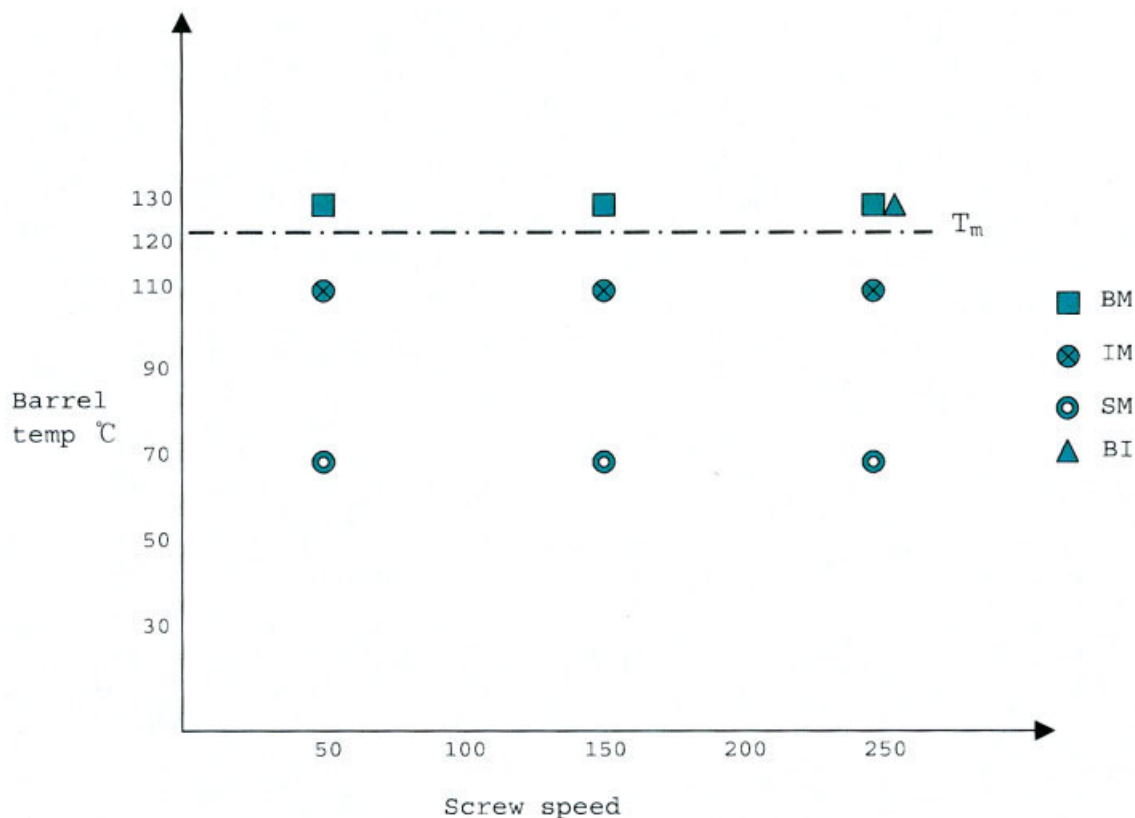
ually from the barrel in the successively sectioned carcasses. From our observation, the carcasses' melting was initiated and propagated from the barrel. At 150 and 250 rpm, melting was initiated by the same mechanism, but propagated by the bed instability process.

Figure 14 shows three melting initiation mechanisms and four melting propagation mechanisms for the LLDPE compound with 10 vol % of calcium carbonate (ViCALity Heavy, 3  $\mu\text{m}$ ).

#### **Smaller particle size and lower calcium carbonate concentration compound**

Figures 15–17 show typical melting initiation and propagation mechanisms of the LLDPE compound with 10 vol % of calcium carbonate (Multiflex, 0.07  $\mu\text{m}$ ).

For the barrel temperature 70°C, we observed the same melting initiation and propagation mechanisms at 50, 150, and 250 rpm as in the previous compounds



**Figure 10** Melting behavior at various screw speeds and barrel temperatures at 6.0 kg/h for aluminum flake compound. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

at the same experimental conditions. From Figure 11, at 150 rpm, we see the melting initiation and propagation mechanism in detail. Deformed pellets are observed in the digital photograph of the carcass on the modular screw. In the digital photographs, 2R and 3R, deformed pellets and clustered and deformed pellets toward the screw surface are observed, and a growing melt layer from the screw surface is observed in the digital photograph of 4R. Here, melt is initiated by the screw melting initiation mechanism and is propagated by the screw melting propagation mechanism. At the other screw speeds, 50 and 250 rpm, screw melting initiation and the screw melting propagation were also observed.

For the barrel temperature 110°C, we did experiments at three screw speeds. Here, we observed the same melting initiation and propagation mechanism through the screw speeds studied. At 250 rpm, we observed internal bulk melting initiation (IM) at 3R and fusing pellets in the bed at 4R (Fig. 16).

For the barrel temperature 130 °C, three melting experiments were carried out at 50, 150, and 250 rpm. At 50 rpm, the melting was initiated at the barrel, and melting proceeded slowly away from the barrel side toward the screw side in the succeeding carcasses. In Figure 17, at 150 rpm, melt is seen in

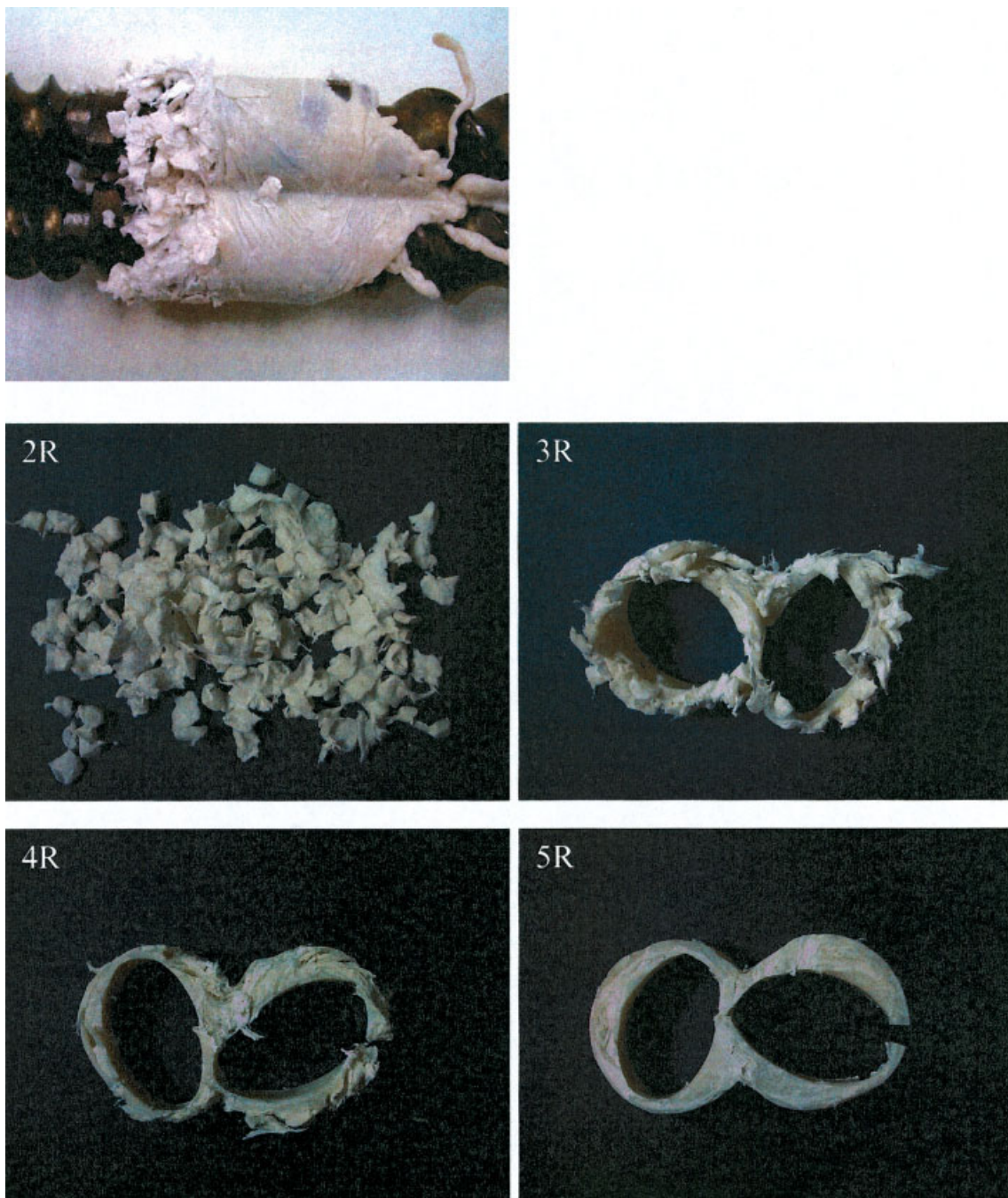
the carcass on the modular screw. Then pellets begin to be surrounded by melt at 3R and subsequently become completely surrounded by melt at 4R. In this case, melting was initiated by the barrel melting (BM) mechanism and propagated by the bed instability (BI) mechanism. At 250 rpm, the experiment results in the same mechanism as seen in the experiments at 150 rpm.

Figure 18 shows the melting initiation and propagation mechanisms of the calcium carbonate compound at three different barrel temperatures and screw speeds at a throughput of 6.0 kg/h. This figure shows three melting initiation mechanisms and four melting propagation mechanisms, which were observed in the experiments of neat polymers.

#### Smaller particle size and higher calcium carbonate concentration compound

Figures 19–21 present carcass cross sections showing the melting initiation and propagation mechanisms of the LLDPE compound with 20 vol % of calcium carbonate (Multiflex, 0.07  $\mu\text{m}$ ).

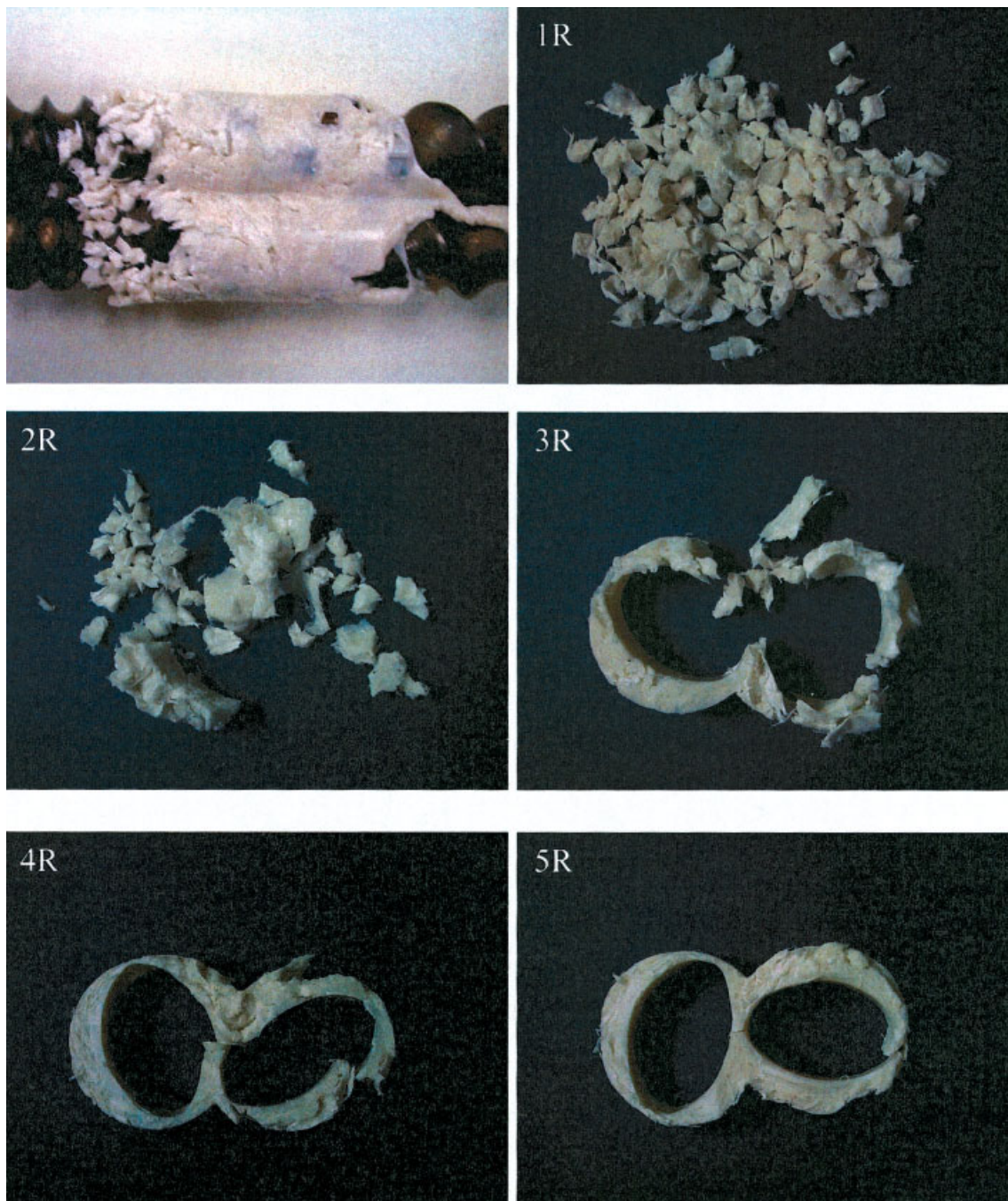
For the barrel temperature 70°C, we observed melting mechanisms for all the screw speeds stud-



**Figure 11** Sectioned carcasses of master batched compound with 10 vol % of calcium carbonate (ViCALity Heavy) at feed rate 6.0 kg/h, barrel temperature 70°C, screw speed 150 rpm. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

ied here. In Figure 19, when the screw speed is 50 rpm, deformed pellets are observed in 2R. These deformed pellets are clustered on the screw surface in 3R. A melt layer at the screw surface is observed first in 3R. The melt layer grew further from the screw surface as shown in 4R. For the experiments at 150 and 250 rpm screw speeds, melting was also initiated at the screw and propagated from the screw surface.

In Figure 20, where  $T_b = 110^\circ\text{C}$  and  $N = 50$  rpm before melting occurred, pellets were deformed as shown at 1R and 2R and compacted as shown in the digital photograph of the modular screw. The melting was initiated by the internal bulk melting initiation mechanism (IM) as shown in 3R, and melting was propagated further by the same mechanism as shown in 4R. When we did further experiments at 150 and 250 rpm, the internal bulk melting initiation



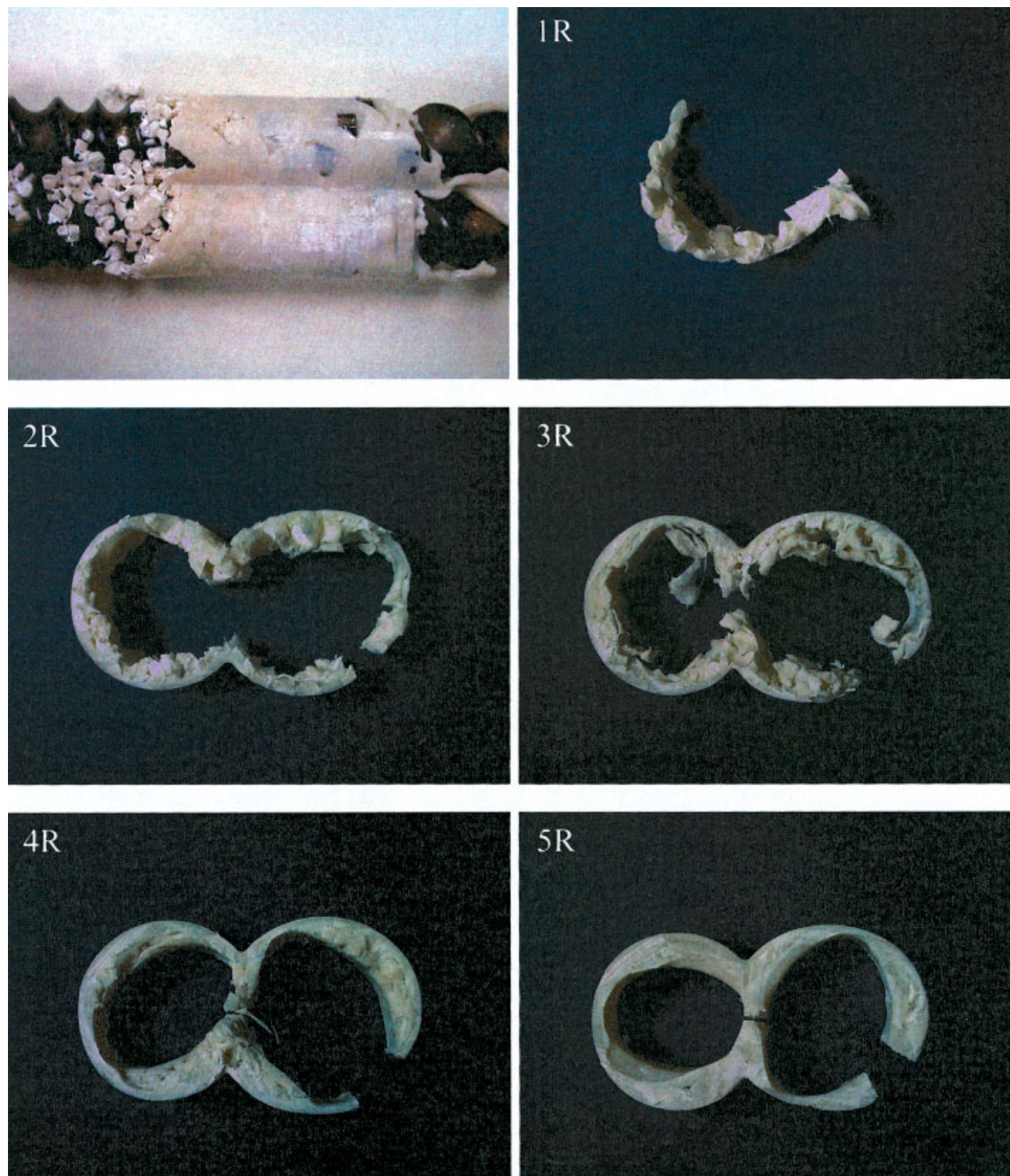
**Figure 12** Sectioned carcasses of master batched compound with 10 vol % of calcium carbonate (ViCALity Heavy) at feed rate 6.0 kg/h, barrel temperature 110°C, screw speed 50 rpm. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

and internal bulk melting propagation were also observed.

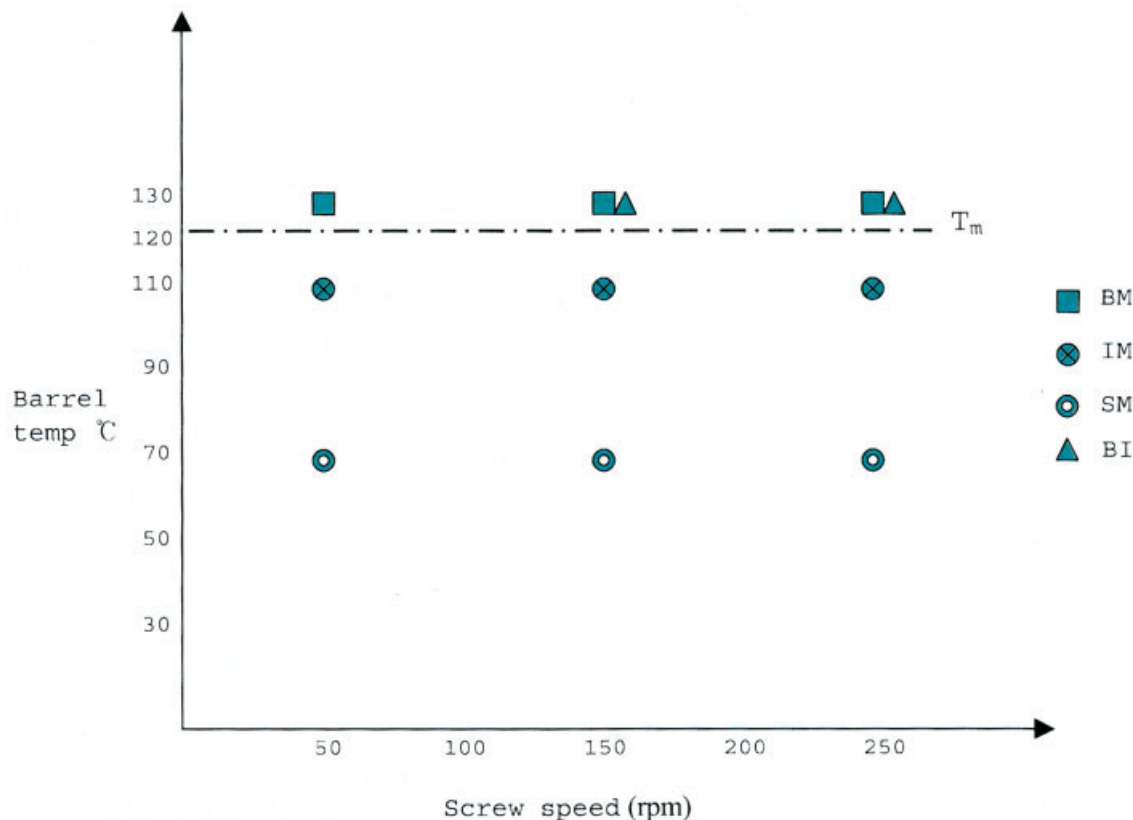
In the experiments at  $T_b = 130^\circ\text{C}$  and  $N = 50$  rpm, melting was initiated at the barrel and propagated from the barrel side in successive carcasses. In Figure 21, where  $T_b = 130^\circ\text{C}$ ,  $N = 150$  rpm, and  $G = 6$  kg/h, melting was initiated from the barrel surface (BM) as shown in the digital photograph of the carcass on the modular screw and in the digital

photograph at 2R. In successive carcasses, melt was propagated by the bed instability (BI) as shown in the digital photographs of 3R and 4R. The experiment, at  $T_b = 130^\circ\text{C}$  and  $N = 250$  rpm, also has a barrel melting initiation mechanism and a bed instability propagation mechanism.

Figure 22 summarizes melting initiation and propagation mechanisms at three screw speeds and three barrel temperatures and at 6.0 kg/h.



**Figure 13** Sectioned carcasses of master batched compound with 10 vol % of calcium carbonate (ViCALity Heavy) at feed rate 6.0 kg/h, barrel temperature 130°C, screw speed 50 rpm. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]



**Figure 14** Melting behavior at various screw speeds and barrel temperatures at 6.0 kg/h for LLDPE compound with 10 vol % of calcium carbonate (ViCALity Heavy, 3  $\mu\text{m}$ ). [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

## DISCUSSION

### General observations

We generally observe that at high temperatures a homogenous melting or molten common layer forms at the barrel. When the temperature is low and screw speed is low, a melt layer is observed near the screw surface. These must be results of the operating machine and not associated with its cooling.

### Effects of filler presence

There are three important factors that determine the influence of fillers. These are filler type, filler amount, and particle size. These influence the melting initiation and propagation in a modular co-rotating twin screw extruder.

To understand the effects of filler presence, filler type filler amount, and filler size, we first looked at the position where melting initiation and completion occur. Figures 23–25 show melting length variations between melting initiation and melting completion on the screw elements.

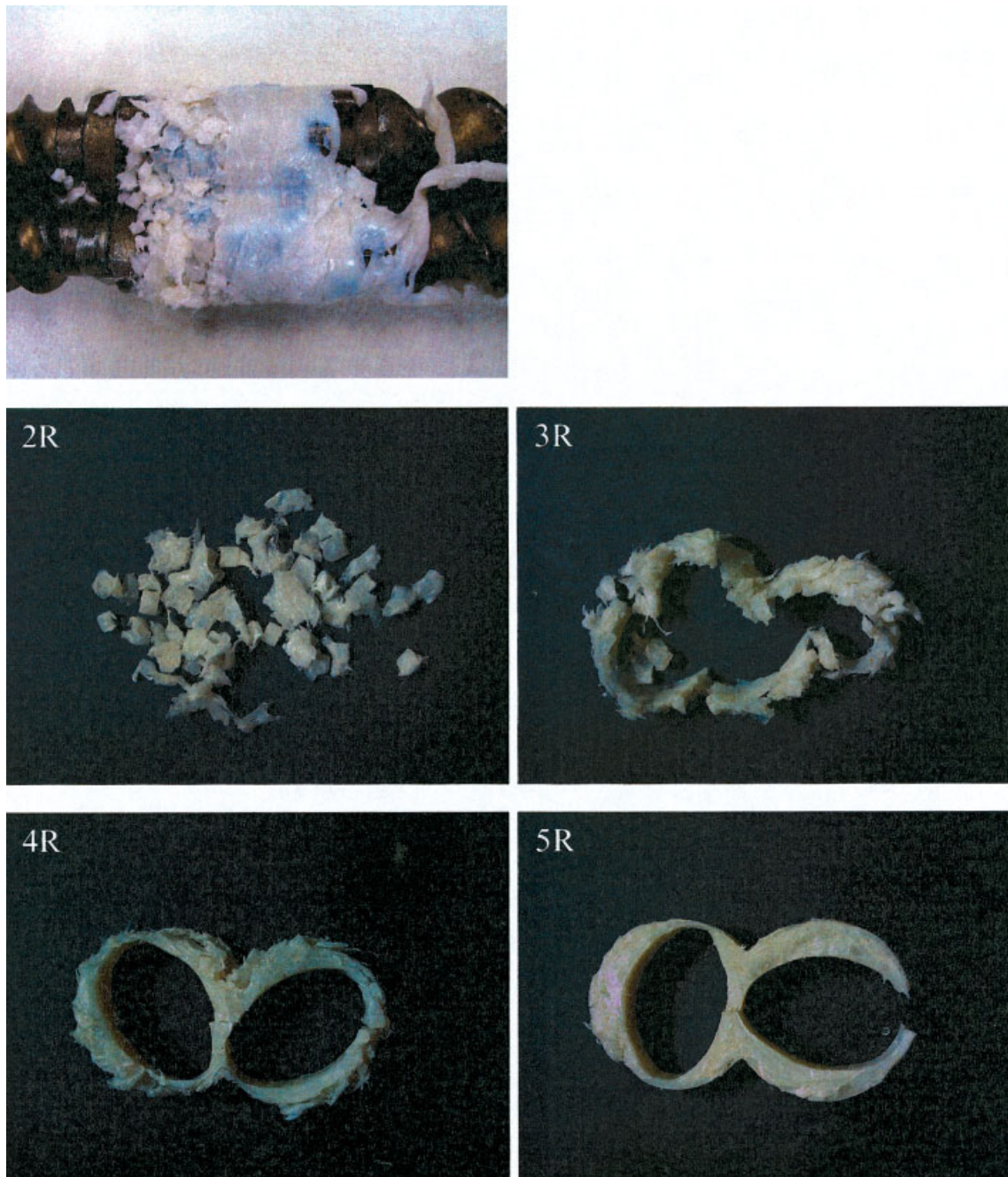
The LLDPE compounds started to melt closer to the hopper than the neat LLDPE in almost all experiments (see Figs. 23–25). Generally the melting rate increases

with increasing filler loading. Further, the rate increases with increasing screw speed. Melting of the LLDPE compounds occurs earlier along the screw than the neat LLDPE as shown in Figures 23–25.

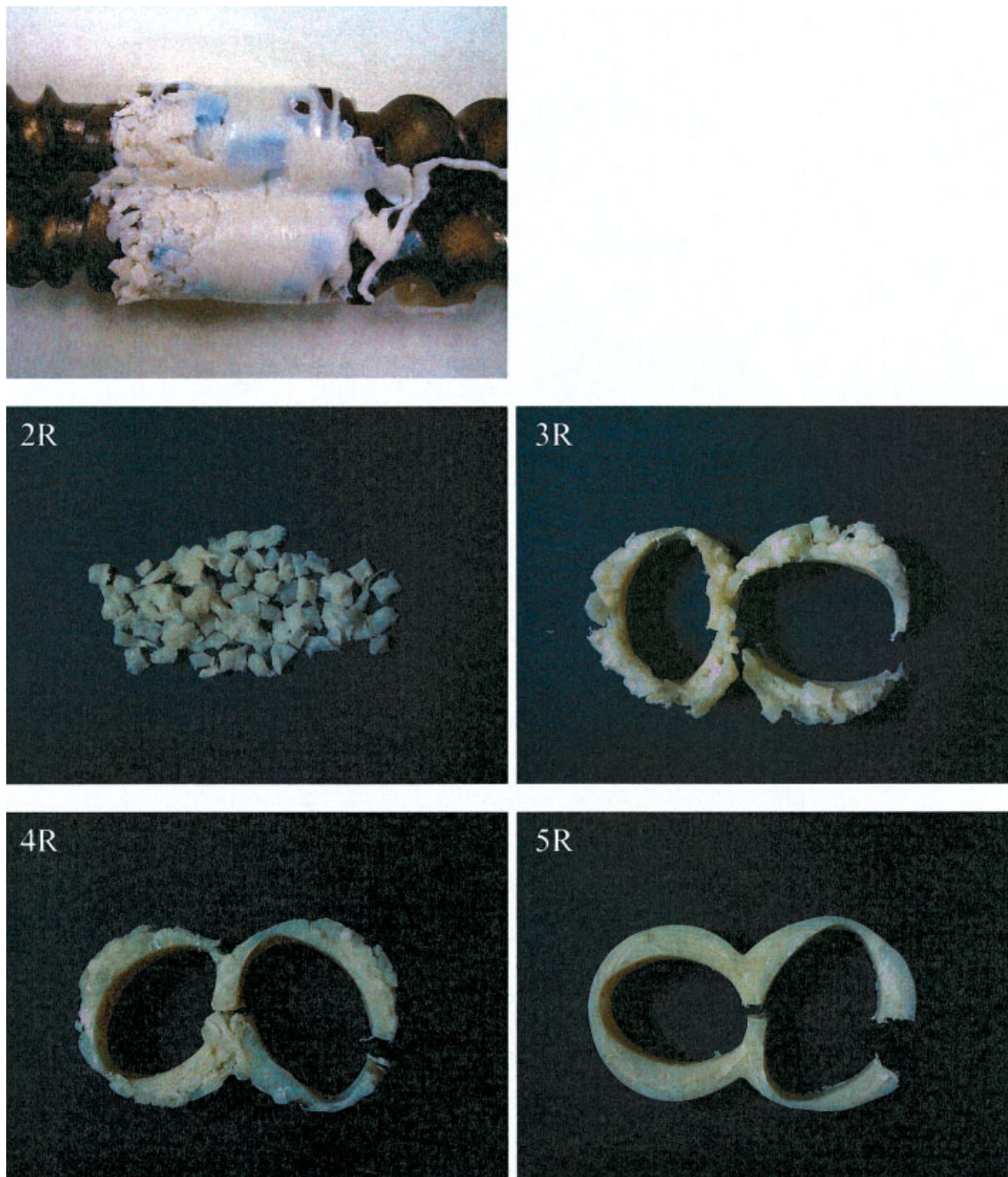
### Effects of filler type

Two compounds were considered to compare the effects of filler type on melting initiation and propagation of the compounds. One is a compound of LLDPE with aluminum flakes, and the other is a compound of LLDPE with the large size of calcium carbonate.

At the barrel temperatures of 70 and 110°C, the compound filled with calcium carbonate began to melt and melted completely at an earlier position than that of the compound filled with aluminum flakes. When the barrel temperature is lower than the crystalline melting temperature of the compound, the compound of aluminum flakes may lose more heat energy through the barrel than that of the other compound because of its higher thermal diffusivity, which would slow melting initiation of the aluminum flake compound. The conductivity and the thermal diffusivity of the aluminum flake compound are 0.549 W/(m k) and 0.283  $\text{m}^2/\text{s}$  and for the calcium carbonate com-

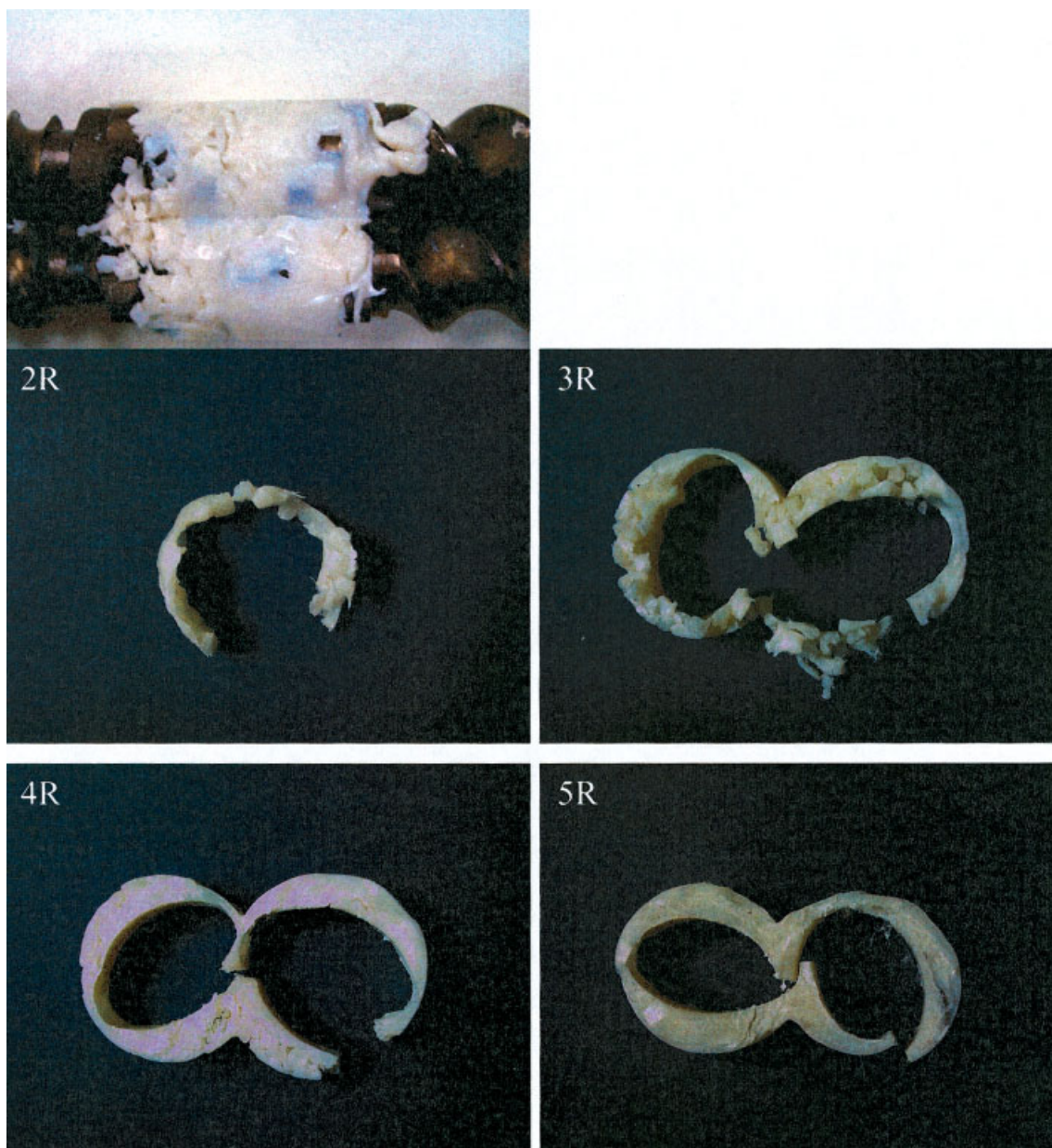


**Figure 15** Sectioned carcasses of master batched compound with 10 vol % of calcium carbonate (Multiflex) at feed rate 6.0 kg/h, barrel temperature 70°C, screw speed 150 rpm. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

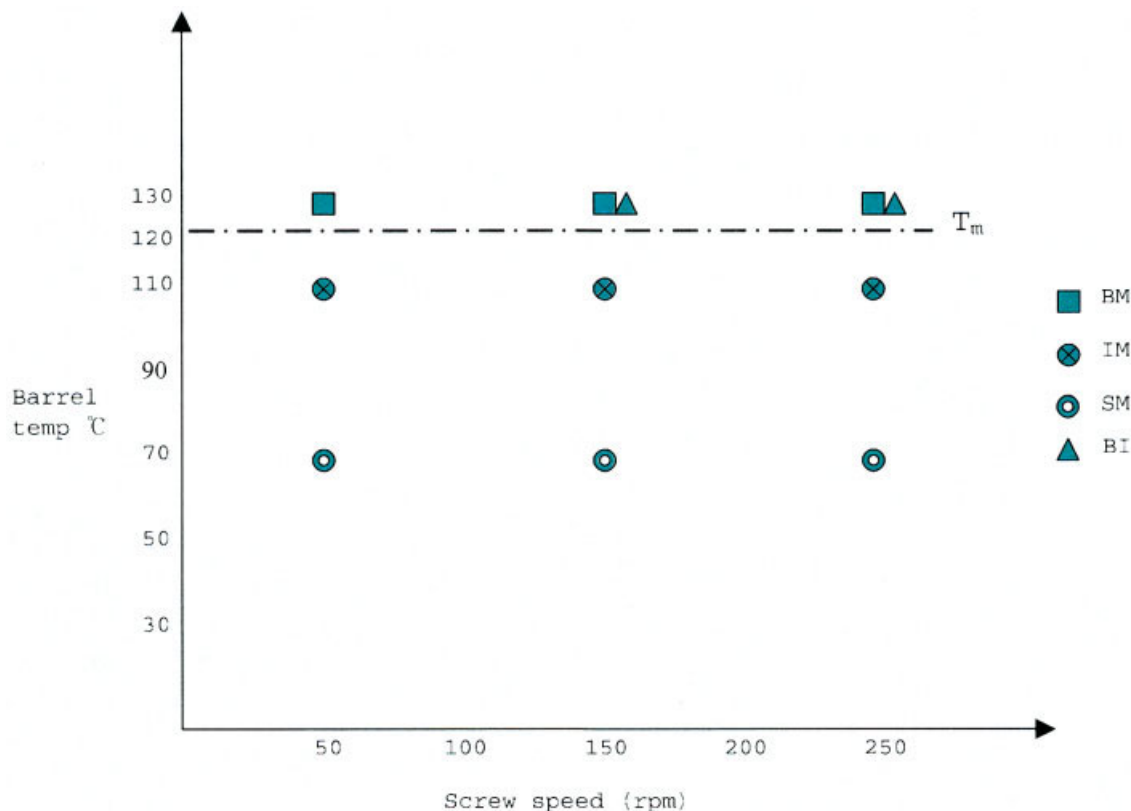


**Figure 16** Sectioned carcasses of master batched compound with 10 vol % of calcium carbonate (Multiflex) at feed rate 6.0 kg/h, barrel temperature 110°C, screw speed 250 rpm. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]





**Figure 17** Sectioned carcasses of master batched compound with 10 vol % of calcium carbonate (Multiflex) at feed rate 6.0 kg/h, barrel temperature 130°C, screw speed 150 rpm. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]



**Figure 18** Melting behavior at various screw speeds and barrel temperatures at 6.0 kg/h for LLDPE compound with 10 vol % calcium carbonate (Multiflex). [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

pound  $0.417 \text{ W}/(\text{m K})$  and  $0.216 \text{ m}^2/\text{s}$ , respectively, as shown in Table II.

At the barrel temperature of  $130^\circ\text{C}$  and at the screw speed of 50 rpm, the melting of the aluminum flake compounds was initiated first, and melting completion was earlier than the calcium carbonate compound as shown in Figure 25. This could be caused by the higher thermal diffusivity. But at higher screw speed, the effect of heat dissipation by small particle competes with heat conduction. At 150 and 250 rpm, these two compounds have the same melting length as shown in Figure 25.

#### Effects of filler content

About 10 and 20 vol % of small size of the calcium carbonate (Multiflex,  $0.07 \mu\text{m}$ ) were used in our experiments.

The greater the amount of filler used in a compound, the lower the specific heat capacity of the compound is, because the heat capacity of the filler is smaller than that of the matrix polymer as shown in Table II.

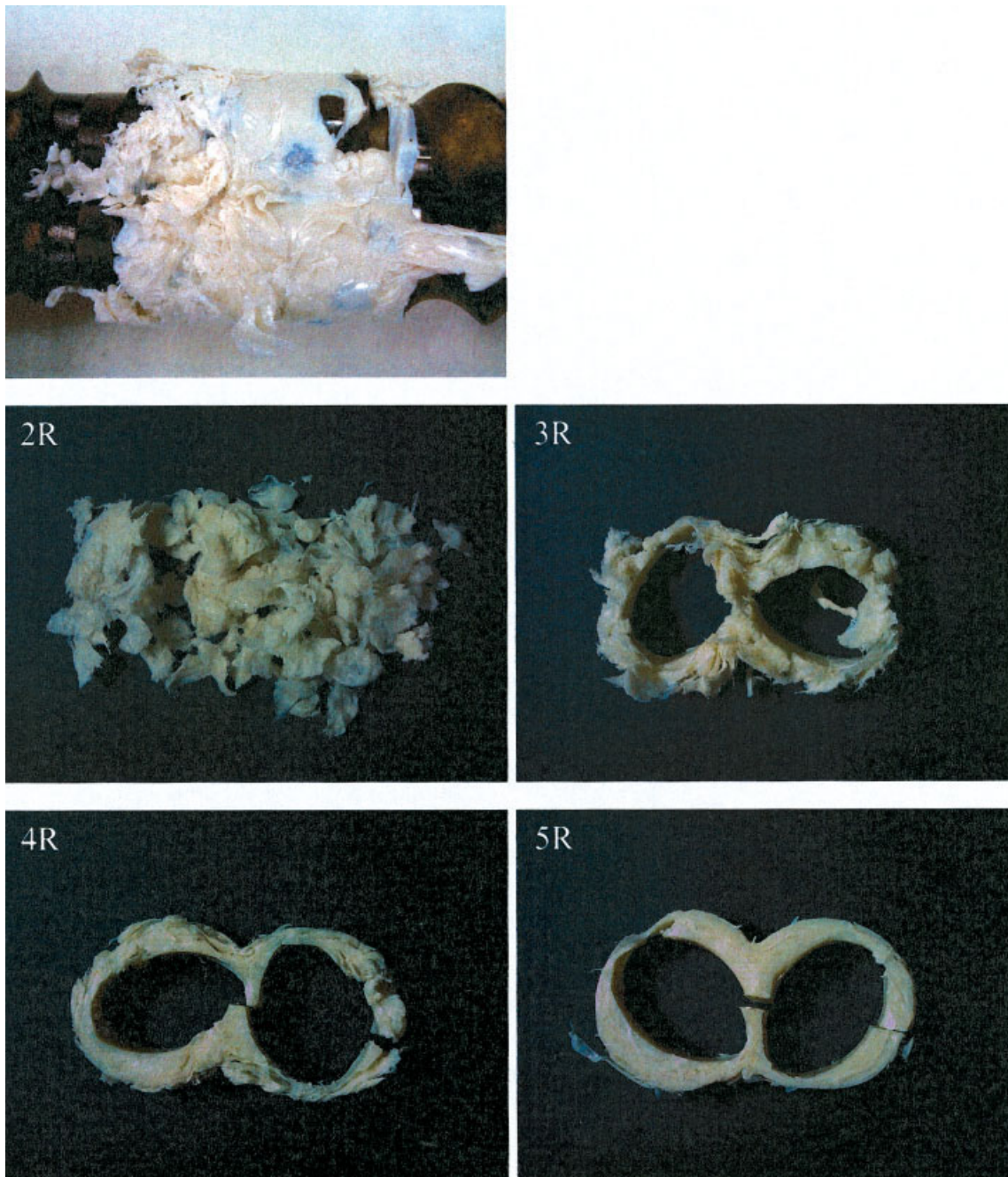
The compound with a filler of 20 vol % completely melted slightly earlier than the compound with a filler of 10 vol %. The shorter melting length would seem to be caused by greater heat dissipation and by less heat

energy needed to melt the compound containing 20 vol % filler.

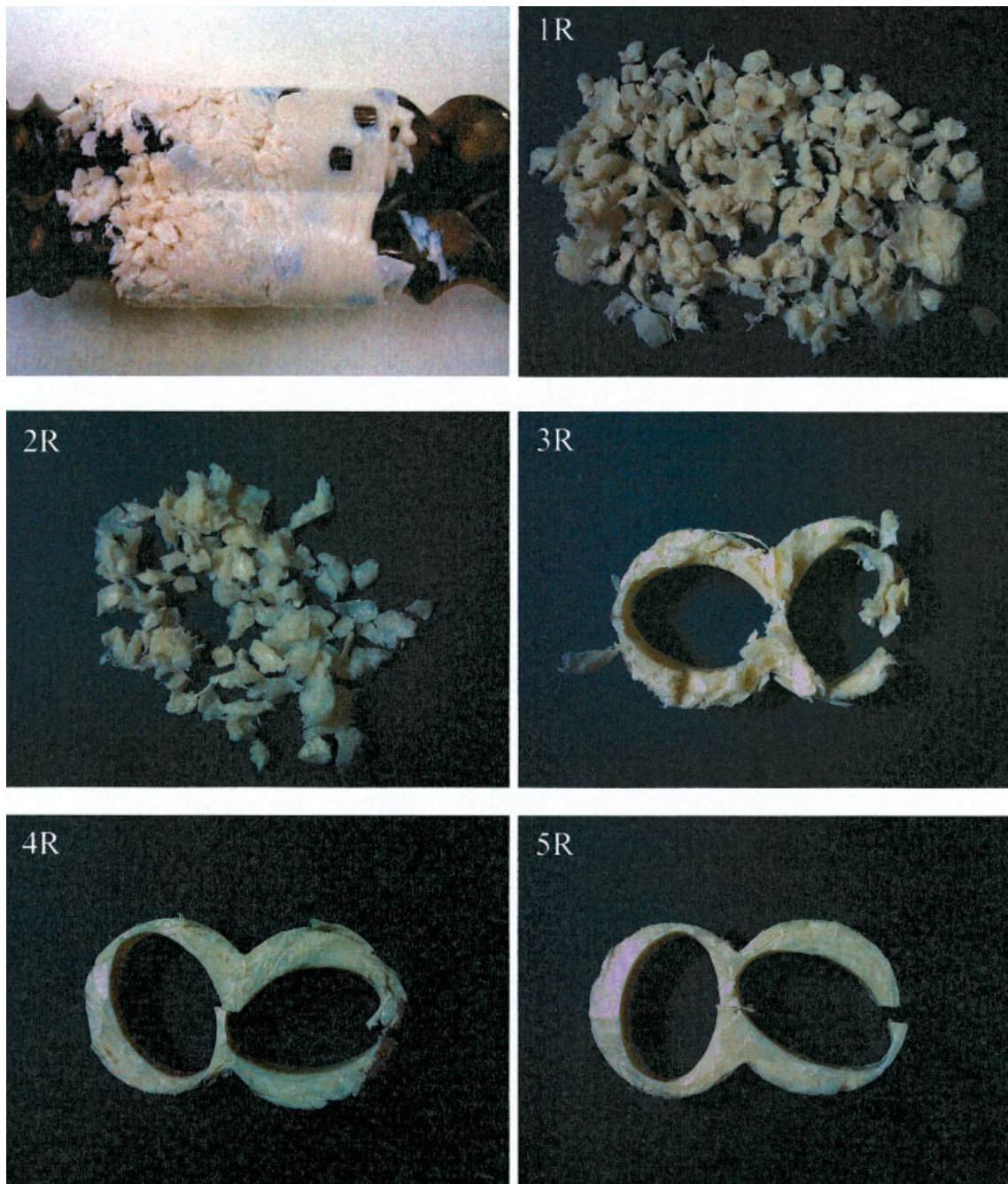
#### Effects of filler size

We used two different calcium carbonates in size, such as  $0.07 \mu\text{m}$  (Multiflex) and  $3.0 \mu\text{m}$  (ViCALity Heavy), and made two compounds with 10 vol % of filler. In this case, we wanted to observe the effects of the size of the particle of two different fillers on melting initiation and propagation.

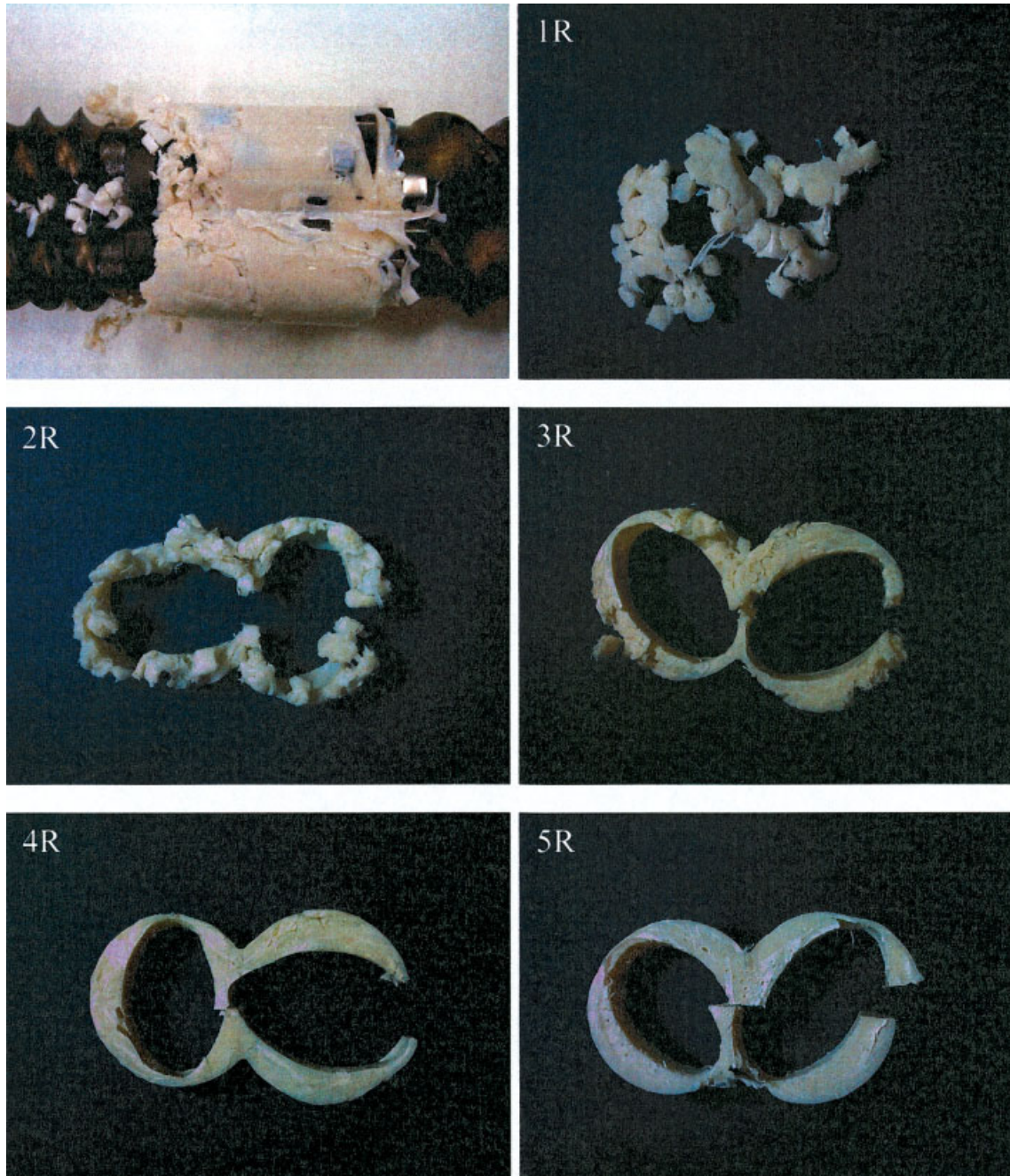
Although the position of melting initiation of these two compounds is the same, there is some difference in the melting length between two compounds filled with small sized (LLDPE + CCl) or large sized calcium carbonate (LLDPE + CC2). As we can see, at  $70^\circ\text{C}$  and 50 rpm (Fig. 23), at  $110^\circ\text{C}$  and 50 or 250 rpm (Fig. 24), and at  $130^\circ\text{C}$  and 250 rpm (Fig. 25), the completion of the melting of the small sized filler compound was usually faster than that of the large sized filler compound. From Figures 2 through 4, we can see that the smaller the particle, the higher the shear viscosity and the more viscous the heat dissipation.



**Figure 19** Sectioned carcasses of master batched compound with 20 vol % of calcium carbonate (Multiflex) at feed rate 6.0 kg/h, barrel temperature 70°C, screw speed 50 rpm. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]



**Figure 20** Sectioned carcasses of master batched compound with 20 vol % of calcium carbonate (Multiflex) at feed rate 6.0 kg/h, barrel temperature 110°C, screw speed 50 rpm. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]



**Figure 21** Sectioned carcasses of master batched compound with 20 vol % of calcium carbonate (Multiflex) at feed rate 6.0 kg/h, barrel temperature 130°C, screw speed 150 rpm. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

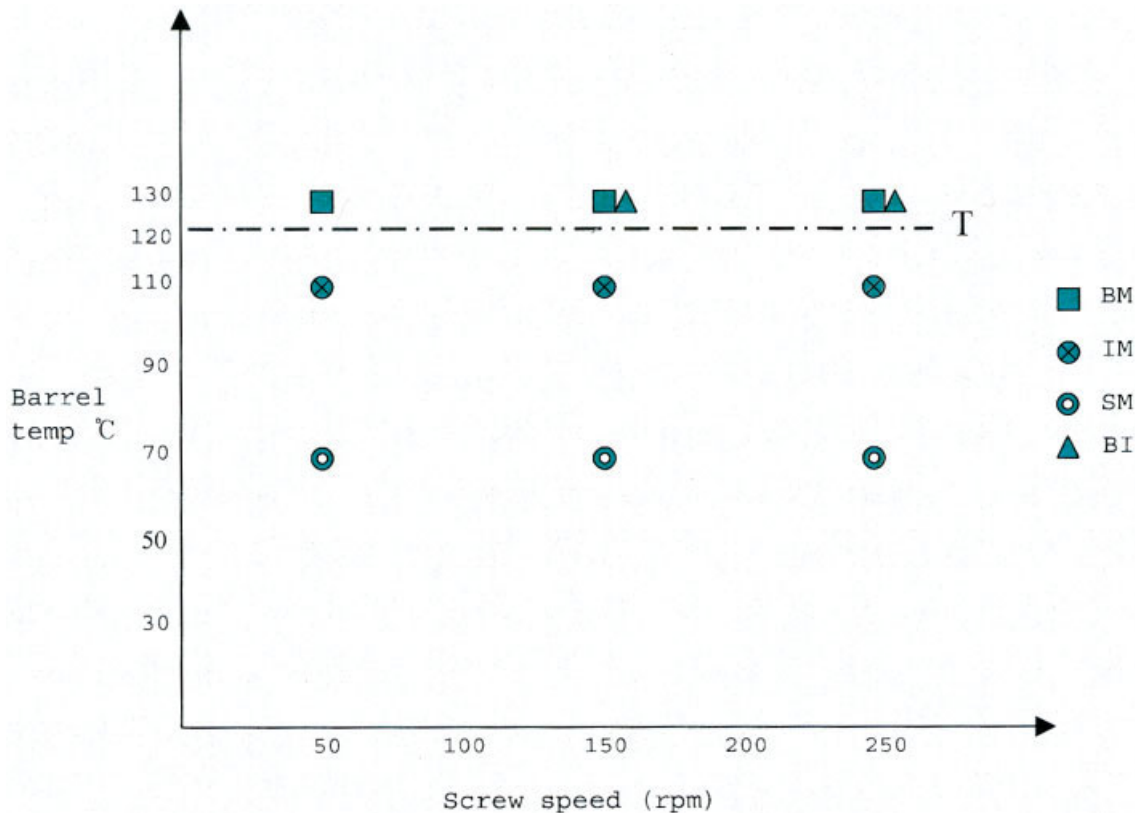


Figure 22 Melting mechanisms at various screw speeds and barrel temperatures at 6.0 kg/h for LLDPE compound with 20 vol % of calcium carbonate (Multiflex). [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

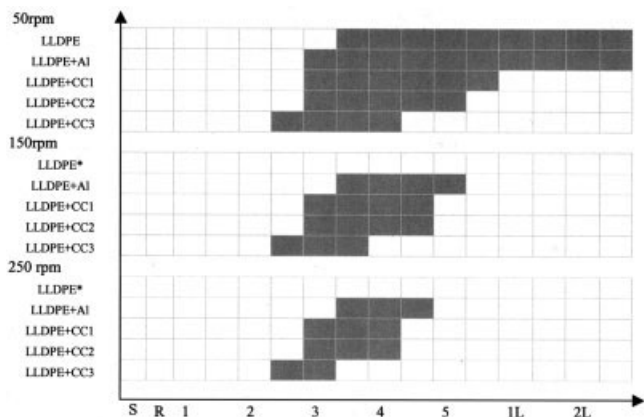


Figure 23 Melt length from a location of melting initiation to a location of full melting for neat LLDPE and its compounds at 70°C barrel temperature, and three screw speeds such as 50, 150, and 250 rpm. (S, screw element; R, ring element; 1–5, disc number on the right-handed kneading disc block element; 1L and 2L, disc number on the left-handed kneading disc block element; LLDPE\*, there is no bar mark for LLDPE, which means the no melting.)

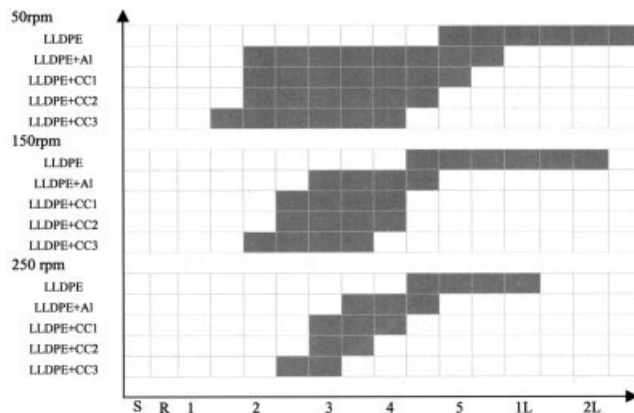
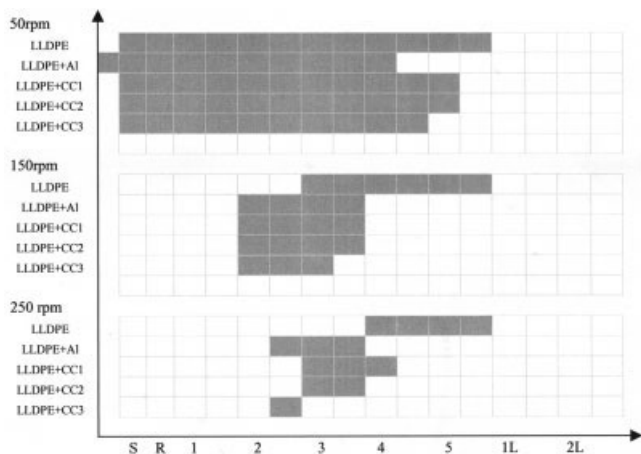


Figure 24 Melt length from a location of melting initiation to a location of full melting for neat LLDPE and its compounds at 110°C barrel temperature and three screw speeds such as 50, 150, and 250 rpm. (S, screw element; R, ring element; 1–5, disc number on the right-handed kneading disc block element; 1L and 2L, disc number on the left-handed kneading disc block element.)



**Figure 25** Melt length from a location of melting initiation to a location of full melting for neat LLDPE and its compounds at 130°C barrel temperature and three screw speeds such as 50, 150, and 250 rpm. (S, screw element; R, ring element; 1–5, disc number on the right-handed kneading disc block element; 1L and 2L, disc number on the left-handed kneading disc block element.)

## CONCLUSIONS

We carried out melting experiments using compounds that contain aluminum flakes, different sizes of calcium carbonate, or different amounts of calcium carbonate individually.

The melting length decreases with increasing screw speed as shown in Figures 19–21.

The presence of fillers enhances the rate of melting, and it begins closer to the hopper.

For compounds mixed with filler, the melting length of barrel melting initiation and propagation mechanism at a low screw speed is greater in melting mechanisms.

There appear to be several heat sources to melt pellets. These heat sources seem to be viscous heat dissipation of

melt surrounding pellets and deformation heat dissipation of pellets and friction heat dissipation between pellets and the screw surface. Viscous heat dissipation is more favorable at high screw speeds.

Even though fillers influence melting, the compounds' melting mechanisms as shown in Figures 6, 10, 14, and 18 follow that of polyolefins. Figures 6, 10, 14, and 18 show that compounds have the same three melting initiation mechanisms and four melting propagation mechanisms, depending on barrel temperature and barrel screw speed as do polyolefins. At temperatures above the crystalline melting point, melting initiates at the barrel and is propagated by viscous heating. At lower temperatures, mechanical deformation of pellets seems to dominate. At still lower temperatures, melting is initiated by friction between the pellets and the screw surface.

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