

**METHOD FOR INJECTION-MOLDING OF RESIN**  
**BACKGROUND OF THE INVENTION**  
**RELATED ART**

The present invention relates to a method for  
5 injection-molding of resin, such as thermotropic liquid  
crystalline polymer (herein below "liquid crystalline  
polymer"). In more detail, the present invention provides  
an injection-molding method with a multi-cavity mold system,  
wherein the molten resin exhibits good flowability and the  
10 waste resin can be reduced. Therefore, the productivity of  
molded articles can be improved and the cost can be  
reduced by the present invention. Further, the present  
invention relates to an injection-molding system, which is  
used for the method of the present invention and a mold  
15 used for the present invention.

**PRIOR ARTS**

A liquid crystalline polymer exhibits good thermal  
stability and good flowability in molten phase as well as  
excellent moldability because of less flash and warpage  
20 development in molding process compared to the other  
thermoplastic resins. Accordingly, consumption of the  
liquid crystalline polymers has been increasing in the fields  
of electric and electronic devices and precision machines,  
high-tech devices such as personal computers and mobile  
25 phones.

However, due to the rapid expansion of consumption and application area, the liquid crystalline resins now face competition in the market not only with super engineering plastics such as PPS (polyphenylene sulfide resin), but also with general-purpose engineering plastics such as PBT (polybutylene terephthalate) and polyamides. Accordingly, market requires cost-reduction even for the molded parts made from liquid crystalline polymers, and there is great demand for further improvement on moldability of said resin.

In terms of improving productivity of the liquid crystalline polymers, various attempts have been carried out from both of the material and process points of view. From the material point of view, for example, Japanese Patent Application Laid Open No. 85733/1984 discloses that productivity in extrusion molding of liquid crystalline polymers can be improved by adding small amount of liquid crystalline compound having low molecular weight (less than 1000) thereto. In addition, Japanese Patent Application Laid Open No. 252457/1991 discloses a technology to improve flowability of a liquid crystalline polymer by adding small amount of oligomer of which primary monomer component is p-hydroxy benzoic acid.

However, the former resin comprising the low molecular weight compound may show unstable metering

behavior in the molding process due to thermal decomposition and gas evolution, and may cause short shot. Therefore, said resin is not suitable for molding an article used in electronic components or the like wherein high dimensional stability is required. In addition, said molded product may invite warpage and blister when subjected to reflow soldering, a recently popular soldering process. On the other hand, the molded articles of the latter resin may be thermal decomposable due to the heat unstable oligomers, and are not suitable for the reflow-soldering process. That is, although the approaches from the material point of view could provide some improvement, it gave negative effect in total partly due to the high molding temperature of liquid crystalline polymers, and no satisfied result has yet achieved.

From the process point of view, for example Japanese Patent Application Laid Open No. 31776/1994 discloses a method for injection-molding of resin using a mold having plurality of cavities characterized in that the diameter of the respective entrance runners and/or gates are different.

Although this molding method can provide slightly improved weld strength to the molded article, there is no significant improvement on the productivity of the resin. That is, no approach from the process point of view

improved the moldability of the liquid crystalline polymers significantly.

Further, it is usual in this art field to optimize molding conditions such as mold temperature and injection speed per resins to improve productivity, however, in fact, significant effect is hardly attained.

As described above, there is no prior art, which can significantly enhance productivity of molded products without impair the characteristics of the resin such as liquid crystalline polymer.

#### SUMMARY OF THE INVENTION

The inventors had carried out extensive studies on injection-molding processes and found that, in a method for injection-molding of thermotropic liquid crystalline polymer, wherein molten resin is injected by a injection machine, flows into the mold through the sprue and fills cavities by way of runners which are connected to the cavities via gates, moldability can be significantly improved by using a resin having certain melt viscosity and a mold having runners with certain cross-sectional area. That is, by employing the resin and mold as above, flowability of the molten resin at injection is increased and therefore, the moldability of the resin such as liquid crystalline polymer is significantly improved. In addition, it has also been surprisingly found that the productivity of above injection-

molding method can be further improved when a multi-cavity system is employed. Further, it has also been found that the productivity of above injection-molding method can be improved further more by adding a certain amount of mold-release agent to the liquid crystalline polymer before  
5     subjecting the polymer to the injection-molding process.

Accordingly, the present invention provides a method for injection-molding of resin, in which a mold comprising a sprue, one or more runners and one or more  
10     cavities is used, characterized in that the cross-sectional area of at least part of the runners is less than  $2.0 \text{ mm}^2$ , and the melt viscosity of the resin is equal to or less than 800 poise when measured at the temperature of  $25^\circ\text{C}$  higher than the melting point of the resin and the shear rate of  
15      $1000 \text{ sec}^{-1}$ .

The method of the present invention is especially effective where the mold has multiple cavities, especially, 4 or more cavities. The method of the present invention is especially effective for injection-molding of a thermotropic  
20     liquid crystalline polymer.

The present invention further provides a method for injection-molding of a liquid crystalline polymer composition, wherein said composition comprises 100 parts by weight of liquid crystalline polymer and 0.01-1 parts by  
25     weight of mold-release agent.

The present invention further provides an injection-molding system for resin which is preferably used in the present invention, comprising an injection-molding machine and a mold comprising a sprue, one or more runners and one or more cavities characterized in that the cross-sectional area of at least part of the runners is less than  $2.0 \text{ mm}^2$ .

The present invention further provides a mold for injection-molding comprising a sprue, one or more runners and one or more cavities characterized in that at the cross-sectional area of at least part of the runners is less than  $2.0 \text{ mm}^2$ .

The injection-molding system and the mold of the present invention having 4 or more cavities are especially useful.

#### BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 is a schematic diagram of the cavity used in the example.

Figure 2 is a schematic diagram of the mold with 8 cavities used in the example showing layout of runners and cavities.

Figure 3 is a schematic diagram of the mold with 16 cavities used in the example showing layout of runners and cavities.

25 Symbols

1: cavity, 2: side gate, 3: opening, 4: runner,  
5: sprue

#### DETAILED DESCRIPTION OF THE INVENTION

According to the present invention, in a method  
5 for injection-molding of resin, wherein molten resin is  
injected from an injection machine then fed into a mold  
through the sprue and fills cavities by way of runners which  
are connected to the cavities via gates, runners having  
certain thin cross-sectional area are employed.

10 In the present specification and claims, runner  
means a channel through which molten resin, which is fed  
into the mold through the sprue, flows towards the product  
parts (cavities) of the mold. Resin is filled into the cavities  
through the gates connected with the runners to provide  
15 shaped product. When the mold has multiple cavities, the  
runners divide the molten resin into each of the cavities.

Sprue means the part from the nozzle of the  
injection cylinder to the runner through which molten resin  
flows.

20 In injection-molding of a general purpose  
engineering plastics such as PBT, it is believed that the  
cross-sectional area of whole of the runners should be more  
than about 5 mm<sup>2</sup>. When said area is less than 5 mm<sup>2</sup>, the  
flowability of the resin is believed to decrease and causes  
25 short-shot, wherein the cavity is not completely filled with

the resin.

On the other hand, in injection-molding of liquid crystalline polymer, a mold having runners of which cross-sectional area is thinner, that is 3-5 mm<sup>2</sup>, has conventionally been used. As is the case of the general-purpose plastics, it has been generally believed that thinner runners may cause deficiency in filling the resin into the cavities.

The present inventors have surprisingly found that by means of very thin runners, that is, those having less than 2.0mm<sup>2</sup> of cross-sectional area, the flowability of molten resin in the molding process is increased and adopted this phenomenon to accomplish the present invention.

Accordingly, the present method for injection-molding of resin using a mold comprising a sprue, one or more runners and one or more cavities, characterized in that the cross-sectional area of at least part of the runners is less than 2.0mm<sup>2</sup> and the melt viscosity of the resin, measured at the temperature of 25°C higher than the melting point of the resin and at the shear rate of 1000 sec<sup>-1</sup>, is equal to or less than 800 poise. The cross-sectional area of the runner may preferably be less than 1.6mm<sup>2</sup>, more preferably, less than 1.3 mm<sup>2</sup>.

When a mold wherein cross sectional area of the

whole runners is more than  $2.0\text{mm}^2$  is used, flowability of molten resin decrease and causes short shot to provide unsatisfactory shaped articles. Such defective tendency as above is significant where a mold having 2 or more cavities, especially, 4 or more cavities is used.

The lower limit of the cross-sectional area of the runner is not limited as long as it is enough for degassification and the resin can flow well. It may preferably be more than  $0.1\text{mm}^2$ , more preferably, more than  $0.2\text{mm}^2$  and most preferably, more than  $0.5\text{mm}^2$ .

The reason why the flowability of molten resin is improved by employing a mold having runners of which cross-sectional area is small has not yet fully described. It is presumable that when a resin of which melt viscosity is less than 800 poise is used, the viscosity of the resin may be decreased in accordance with increase of the flow speed of the molten resin through the runner; and because of the increased flow speed, resistance against the molten resin increases and therefore, the resin will release heat and the viscosity is further reduced due to the heat.

According to the present invention, the cross-sectional shape of the runner is not limited and may be any of known shapes such as circle, semicircle, trapezoid, square, and triangle. The length of the runner is not limited and may be designed in a conventional manner

based on the layout of the cavities.

The cross-sectional areas of the respective runners in a mold may or may not be identical. When the areas are not identical among the runners, the total length  
5 of the runners where the cross-sectional area is in the range of the present invention is more than  $1/4$ , preferably,  $1/3$  and more preferably,  $1/2$  of the whole length of the runners.

In a mold having multiple cavities, the cross-  
10 sectional areas of the respective runners may or may not be identical, and the length of each runner for each cavity may or may not be identical.

Because of the thin runners, total amount of resin fed into the mold is reduced and therefore, the  
15 present invention is advantageous for lowering cost. Especially, liquid crystalline polymers are in general used for manufacturing small-size articles and many of conventional molding systems for liquid crystalline polymers have relatively large part of resin molded in the  
20 runners when compared with the product. The present invention can also remove this problem.

That is, according to the method of the present invention, when the mold is filled with the resin, the ratio of the resin remains in the runners to the total amount of the  
25 resin in the cavities, runners and sprue is preferably less

than 50 wt%, more preferably, less than 25 wt% and most preferably, less than 15 wt%.

By adjusting the volume ratio of the respective parts of the mold as above, the waste of resin can be reduced and therefore, productivity of the molding process is significantly improved.

In the present invention, flowability of resin is improved more significantly when the method is to provide molded products of which individual weight is less than 1g, i.e. where the amount of resin filled in one cavity is less than 1g, and the improvement of productivity is more significant in this condition.

Further, the injection-molding method of the present invention is especially useful for mold having 2 or more, especially, 4 or more cavities. When a conventional mold having multiple cavities and runners of which cross-sectional area is more than 2.0 mm<sup>2</sup>, the filling status of the respective cavities may vary widely. To the contrary, cavities can be uniformly filled by employing the mold of the present invention having the thinner runners.

According to the present invention, the number of the cavities may be one or more and preferably 4 or more. As the number of cavities increases, for example, to 8, 16 or 32 cavities, the productivity of the present method is more enhanced.

When a mold having multiple cavities is used, the layout of the cavities is not limited and any of conventional layouts can be employed. The number of the cavities is not limited as long as they can be arranged, and usually, may be up to around 100.

According to the present method for molding, the linear velocity of the injected molten resin in the runner may be more than 1000mm/sec and preferably, 1500-100000mm/sec. By adjusting the linear velocity of the injected molten resin in the runner to 1000mm/sec or higher, short-shot in filling cavities may be reduced and weld strength of the molded product may be improved, especially in an article with long flow distance, more especially, an article molded by means of a cavity having a large flow distance/thickness ratio.

In the present invention, the resin used in the present method have an melt viscosity, measured with capillary rheometer under the condition at the temperature of 25°C higher than the melting point of the resin and at the shear rate of 1000 sec<sup>-1</sup>, of equal to or less than 800 poise, preferably 10-800 poise, and more preferably 100-600 poise. From moldability and flowability points of view, 100-550poise is most preferable. If the melt viscosity is higher than 800 poise, flowability of the resin decreases and causes insufficiently filling of the cavities to give

unsatisfied product.

As for resins exhibit the above-defined viscosity, liquid crystalline polymers are especially useful.

We discuss below on liquid crystalline polymers, which are useful for the molding method of the present invention.

The liquid crystalline polymer used in the present molding process may be any of those exhibits anisotropic melt phase and called as thermotropic liquid crystalline polymer by the skilled art in this field.

The anisotropic melt phase can be confirmed by means of conventional polarized light system using orthogonal light polarizer. In more detail, the sample on the Leitz's hot stage under nitrogen atmosphere may be observed with Leitz's polarization microscope at 40-magunification. The above-described polymer is optically anisotropic. That is, the polymer transmits light when observed under orthogonal light polarizer. When a sample is optically anisotropic, the sample transmit polarized light even under static state.

Examples of the liquid crystalline polymers used in the present invention may include liquid crystalline polyesters and liquid crystalline polyamides which exhibit anisotropic melt phase. Said polymers may be consisted of one or more monomer units selected from the group

consisting of aromatic hydroxycarboxylic acids, aromatic dicarboxylic acids, aromatic diols, aromatic hydroxyamines, aromatic diamines, aromatic aminocarboxylic acids and the like, especially, from the group consisting of aromatic  
5 dicarboxylic acids, aromatic diols and aromatic hydroxycarboxylic acids.

Examples of the aromatic hydroxycarboxylic acid may include not only aromatic hydroxycarboxylic acids but also alkyl, alkoxy or halogen substituted derivatives thereof  
10 as well as ester forming derivatives thereof and may include 4-hydroxybenzoic acid, 2-hydroxy-3-naphthoic acid, 2-hydroxy-6-naphthoic acid, 2-hydroxy-7-naphthoic acid, 3-methyl-4-hydroxybenzoic acid, 3,5-dimethyl-4-hydroxybenzoic acid, 2,6-dimethyl-4-hydroxy benzoic acid,  
15 2-hydroxy-5-methyl-6-naphthoic acid, 2-hydroxy-5-methoxy-6-naphthoic acid, 3-chloro-4-hydroxybenzoic acid, 2-chloro-4-hydroxybenzoic acid, 2,3-dichloro-4-hydroxybenzoic acid, 3-bromo-4-hydroxybenzoic acid, 2-hydroxy-5-chloro-6-naphthoic acid, 2-hydroxy-7-chloro-6-naphthoic acid, 2-  
20 hydroxy-5,7-dichloro-6-naphthoic acid and 4-hydroxy-4'-biphenyl carboxylic acid. Among the above, 4-hydroxy benzoic acid and 2-hydroxy-6-naphthoic acid are preferable because they provide polymers of which properties and melting point can be easily controlled.

25 Examples of the aromatic dicarboxylic acids may

include not only aromatic dicarboxylic acids but also alkyl, alkoxy or halogen substituted derivatives thereof as well as ester forming derivatives thereof, and may include terephthalic acid, chloro terephthalic acid, dichloro terephthalic acid, bromo terephthalic acid, methyl terephthalic acid, dimethyl terephthalic acid, ethyl terephthalic acid, methoxy terephthalic acid, ethoxy terephthalic acid, isophthalic acid, 4,4'-biphenyldicarboxylic acid, 3,4'-biphenyldicarboxylic acid, 4,4''-terphenyldicarboxylic acid, 2,6-naphthalenedicarboxylic acid, 2,7-naphthalenedicarboxylic acid, 1,6-naphthalenedicarboxylic acid, diphenylether-4,4'-dicarboxylic acid, diphenoxybutane-4,4'-dicarboxylic acid, diphenylethane-4,4'-dicarboxylic acid, diphenylether-3,3'-dicarboxylic acid and diphenylethane-3,3'-dicarboxylic acid. Among the above, terephthalic acid and 2,6-naphthalenedicarboxylic acid are preferable because they provide polymers of which properties such as mechanical property, heat resistance, melting point and moldability can be easily controlled to an adequate level.

Examples of the aromatic diols may include not only aromatic diols but also alkyl, alkoxy or halogen substituted derivatives thereof as well as ester forming derivatives thereof, may include hydroquinone, chloro hydroquinone, methyl hydroquinone, 1-butyl hydroquinone,

phenyl hydroquinone, methoxy hydroquinone, phenoxy hydroquinone, resorcin, 4-chloro resorcin, 4-methyl resorcin, 4,4'-dihydroxybiphenyl, 4,4''-dihydroxyterphenyl, 2,6-naphthalenediol, 1,6-naphthalenediol, 2,7-naphthalenediol, 4,4'-dihydroxydiphenyl ether, bis(4-hydroxyphenoxy)ethane, 3,3'-dihydroxybiphenyl, 3,3'-dihydroxydiphenyl ether and 2,2-bis(4-hydroxyphenyl)methane. Among the above, hydroquinone and 4,4'-dihydroxybiphenyl are preferable in view of their reactivity at the polymerizing process and the characteristics of the obtainable liquid crystalline polymer.

Examples of the aromatic hydroxyamines, aromatic diamines, aromatic aminocarboxylic acids may include aromatic hydroxyamines such as 4-aminophenol, N-methyl-4-aminophenol, 3-aminophenol, 3-methyl-4-aminophenol, 4-amino-1-naphthol, 4-amino-4'-hydroxydiphenyl, 4-amino-4'-hydroxydiphenyl ether, 4-amino-4'-hydroxydiphenyl methane, 4-amino-4'-hydroxybiphenyl sulfide and 4,4'-diaminobiphenyl sulfone; aromatic diamines such as 1,4-phenylenediamine, N-methyl-1,4-phenylenediamine, N,N'-dimethyl-1,4-phenylenediamine, 4,4'-diaminodiphenylsulfide (thiodianiline), 2,5-diamino toluene, 4,4'-ethylenedianiline, 4,4'-diaminodiphenoxy ethane, 4,4'-diaminodiphenyl methane (methylene dianiline) and 4,4'-diaminodiphenyl ether (oxydianiline); and aromatic amino carboxylic acid such as 4-aminobenzoic acid, 2-

amino-6-naphthoic acid and 2-amino-7-naphthoic acid, and ester derivatives thereof.

The liquid crystalline polymer used in the present invention may be the one obtained by copolymerizing the  
5 above described monomer units with one or more other monomer components to the extent that the object of the present invention is not impaired. The other monomer component may be selected from the group consisting of alicyclic dicarboxylic acids, aliphatic diols, alicyclic diols,  
10 aromatic mercaptocarboxylic acids, aromatic dithiols and aromatic mercaptophenols.

Examples of the alicyclic dicarboxylic acids, alicyclic diols and aliphatic diols may include hexahydrotelephthalic acid, trans-1,4-cyclohexanediol, cis-  
15 1,4-cyclohexanediol, trans-1,4-cyclohexanedimethanol, cis-1,4-cyclohexane dimethanol, trans-1,3-cyclohexanediol, cis-1,2-cyclohexane diol, trans-1,3-cyclohexanedimethanol, and linear or branched chain aliphatic diols such as ethylene glycol, 1,3-propanediol, 1,4-butanediol and neopentylglycol,  
20 and ester forming derivatives thereof.

Examples of the mercaptocarboxylic acids, aromatic dithiols and aromatic mercaptophenols include, for example, 4-mercaptobenzoic acid, 2-mercapto-6-naphthoic acid, 2-mercapto-7-naphthoic acid, benzene-1,4-dithiol,  
25 benzene-1,3-dithiol, 2,6-naphthalene-dithiol, 2,7-

naphthalene-dithiol, 4-mercaptophenol, 3-mercaptophenol, 6-mercaptophenol and 7-mercaptophenol, and ester forming derivatives thereof.

The liquid crystalline polymers consisting of the above monomer components may include both of those give anisotropic melt phase and those do not, depending on components of the polymer and the ratio thereof, and the sequence distribution. The liquid crystalline polymers used in the present are limited to those exhibit anisotropic melt phase.

Examples of the preferred liquid crystalline polymers are consists of the following monomer components;

4-hydroxybenzoic acid/ 2-hydroxy-6-naphthoic acid copolymer;

4-hydroxybenzoic acid/ terephthalic acid/ 4,4'-dihydroxy biphenyl copolymer;

4-hydroxybenzoic acid/ terephthalic acid/ isophthalic acid/ 4,4'-dihydroxy biphenyl copolymer;

4-hydroxybenzoic acid/ terephthalic acid/ isophthalic acid/ 4,4'-dihydroxy biphenyl/ hydroquinone copolymer;

4-hydroxybenzoic acid/ terephthalic acid/ hydroquinone copolymer;

4-hydroxybenzoic acid/ 2-hydroxy-6-naphthoic

- acid/ terephthalic acid/ 4,4'-dihydroxy biphenyl copolymer;  
 4-hydroxybenzoic acid/ 2-hydroxy-6-naphthoic  
 acid/ terephthalic acid/ hydroquinone copolymer;  
 4-hydroxybenzoic acid/ 2,6-naphthalene  
 5 dicarboxylic acid/ 4,4'-dihydroxy biphenyl copolymer;  
 4-hydroxybenzoic acid/ terephthalic acid/ 2,6-  
 naphthalene dicarboxylic acid/ hydroquinone copolymer;  
 4-hydroxybenzoic acid/ 2,6-naphthalene  
 dicarboxylic acid/ hydroquinone copolymer;  
 10 4-hydroxybenzoic acid/ 2-hydroxy-6-naphthoic  
 acid/ 2,6-naphthalene dicarboxylic acid/ hydroquinone  
 copolymer;  
 4-hydroxybenzoic acid/ terephthalic acid/ 2,6-  
 naphthalene dicarboxylic acid/ hydroquinone/ 4,4'-dihydroxy  
 15 biphenyl copolymer;  
 4-hydroxybenzoic acid/ terephthalic acid/ 4-  
 aminophenol copolymer;  
 4-hydroxybenzoic acid/ terephthalic acid/ 4,4'-  
 dihydroxy biphenyl/ 4-aminophenol copolymer;  
 20 2-hydroxy-6-naphthoic acid/ terephthalic acid/ 4-  
 aminophenol copolymer;  
 4-hydroxybenzoic acid/ terephthalic acid/ 2,6-  
 naphthalene dicarboxylic acid/ hydroquinone/ 4,4'-dihydroxy  
 biphenyl copolymer;  
 25 4-hydroxybenzoic acid/ 2-hydroxy-6-naphthoic

acid/ terephthalic acid/ 4-aminophenol copolymer;

4-hydroxybenzoic acid/ 2-hydroxy-6-naphthoic acid/ terephthalic acid/ 4,4'-dihydroxy biphenyl/ 4-aminophenol copolymer;

5 2-hydroxy-6-naphthoic acid/ terephthalic acid/ hydroquinone copolymer;

2-hydroxy-6-naphthoic acid/ terephthalic acid/ 4,4'-dihydroxy biphenyl copolymer;

4-hydroxybenzoic acid/ terephthalic acid/ ethylene glycol copolymer;

4-hydroxybenzoic acid/ terephthalic acid/ 4,4'-dihydroxy biphenyl/ ethylene glycol copolymer;

4-hydroxybenzoic acid/ 2-hydroxy-6-naphthoic acid/ terephthalic acid/ ethylene glycol copolymer;

15 4-hydroxybenzoic acid/ 2-hydroxy-6-naphthoic acid/ terephthalic acid/ 4,4'-dihydroxy biphenyl/ ethylene glycol copolymer.

Preferred examples of liquid crystalline polymers used in the present invention are those comprising 2-hydroxy-6-naphthoic acid residue. By employing liquid crystalline polymer having 2-hydroxy-6-naphthoic acid residue, the productivity for providing the molded articles can be further improved.

The method for preparing the liquid crystalline polymer, which can be molded in the present method, is not

limited and any known method can be employed. For example, conventional polyester polycondensation method to give ester bondings among the monomer components in the above combination such as molten acidolysis method and slurry polymerization method may be employed.

The molten acidolysis method is characterized in that monomers selected from the group consisting of aromatic hydroxycarboxylic acids, aromatic dicarboxylic acids, aromatic diols, aromatic hydroxyamines, aromatic hydroxydiamines, aromatic aminocarboxylic acids and the like are reacted without the presence of heatexchange fluid. In this method, the monomers are firstly heated to give molten solution of the reactants and then the solution is reacted to give the molten polymer. The final step of this method may be carried out under vacuum to facilitate removal of volatile by-products such as acetic acid or water.

The slurry polymerization method is characterized in that monomers are reacted in a heatexchange fluid to give polymer as suspension in the fluid.

In either of the molten acidolysis method or the slurry polymerization method, the monomers having hydroxyl group and/or amino group may be preliminary esterified with lower aliphatic acid (preferably, in a manner to provide acetate) before subjecting the same to the reaction. Alternatively, the polymerization process may be

carried out under the presence of lower aliphatic anhydride, preferably acetic anhydride. The lower aliphatic acid may have 2-6, preferably 2-4 carbon atoms, and acetic acid is most preferable.

5. In either of the molten acidolysis method or the slurry polymerization method, catalysts may optionally be added to the reaction.

Examples of the catalysts which may optionally be used include organic tin compounds such as dialkyl tin oxide (ex. dibutyl tin oxide) and diaryl tin oxide; organic titanium compounds such as titanium dioxide, alkoxy titanium silicate and titanium alkoxide; antimony trioxide; alkaline or alkaline earth metal salt of carboxylic acid such as potassium acetate, sodium acetate and zinc acetate; and  
10 gaseous acid catalysts such as Lewis acid (ex.  $\text{BF}_3$ ) and  
15 halogenated hydrogen (ex.  $\text{HCl}$ ).

The amount of the catalyst added to the reaction per 10 parts by weight of the monomers may be 0.001-2 parts by weight, preferably 0.01-1 parts by weight.

- 20 In light of the balance of heat resistance and moldability, the melting point (determined by means of differential scanning calorimetry) of the liquid crystalline polymer used in the present invention is preferably in the range of 250-400 °C, more preferably, in the range of 270-  
25 350 °C.

In order to enhance the effect of the present invention, mold-release agent may be added to the liquid crystalline polymer. In general, a liquid crystalline polymer exhibits good flowability and therefore good releasability without releasing agent. However, by adding mold-release agent to the polymer, the effects of the present invention can be enhanced synergistically, i.e. thinner runners can be used and the number of the cavities can be increased.

The mold-release agent may be added to the liquid crystalline polymer used in the present invention by means of a mixing machine, such as extruder, which is usually used in melt kneading process. Alternatively, the mold-release agent may be added to liquid crystalline polymer pellets so that the agent adhere to the surface of the pellet, and the pellets may be subjected for the injection-molding process.

In the embodiment wherein the mold release agent is admixed with the polymer, 0.001-1 part by weight, preferably, 0.05-0.5 parts by weight of mold-release agent may be added to 100 parts by weight of liquid crystalline polymer. In the embodiment wherein the mold-release agent is adhered to the surface of the polymer pellet, 0.001-0.05 parts by weight, preferably, 0.01-0.03 parts by weight of the mold-release agent may be used per 100 parts by weight of liquid crystalline polymer pellets.

The mold-release agent used in the present invention may be any of the conventional agents for molding resin, which does not significantly decompose at the molding temperature of the liquid crystalline polymer.

5 Examples of such mold-release agents as above may include fluorocarbon resins, polysiloxanes, higher aliphatic acids and metal salts, esters and amides thereof, higher aliphatic alcohol and esters thereof, amide of higher aliphatic amine, fluorocarbon surface active agent and

10 polyolefin wax. Among them, alkaline metal salt such as sodium or potassium salt, alkaline earth metal salt such as calcium or barium salt, magnesium salt and aluminum salt of higher aliphatic acid; esters between higher aliphatic acid and ethyleneglycol, butanediol, glycerin, trimethylol

15 propane and pentaerythritol, especially, full esters containing no free hydroxy group; amides between higher aliphatic acid and ethylenediamine, methylenediamine, hexamethylenediamine, p-phenylenediamine and p-xylilenediamine are preferable.

20 In the present specification and claims, higher aliphatic acid represents saturated hydrocarbon acid having 12-40 carbon atoms, preferably more than 15 carbon atoms and more preferably, more than 17 carbon atoms. Examples of the higher aliphatic acids may include lauric

25 acid, myristic acid, palmitic acid, stearic acid, arachic acid,

behenic acid and montanic acid.

The liquid crystalline polymers used in the present invention may be admixed with other resin component and reinforcing agents and/or fillers of fibrous, lamellar or particulate materials as long as they do not impair the object of the present invention.

Examples of the other resin components include polyamides, polyesters, polyphenylene sulfide, polyether ketone, polycarbonate, polyphenylene ether and modified compounds thereof, thermoplastic resins such as polysulfone, polyethersulfone and polyether imide; thermosetting resins such as phenol resin, epoxy resin and polyimide resin. One or more of those resins may be blended with the liquid crystalline polymer.

Examples of the reinforcing agents and/or fillers of fibrous, lamellar or particulate materials may include glass fiber, silica-alumina fiber, alumina fiber, carbon fiber potassium titanate whisker, aramid borate whisker, titanium oxide whisker, alomido fiber, talc, mica, graphite, calcium carbonate, dolomite, clay, wollastonite, glass beads, glass flakes, barium sulfate and titanium oxide. They may be used solely or in combination of the two or more. Among them, glass fiber is preferably used because of its good balance of physical property and cost.

The reinforcing agents and/or fillers of fibrous,

lamellar or particulate materials may be added to the liquid crystalline polymer in an amount of 0-60wt%, especially 10-50wt%, when the total amount of the liquid crystalline polymer composition comprising the agents and/or fillers is counted as 100 wt%,

The liquid crystalline polymer molded according to the present invention may be further admixed with one or more conventional additives for example colorant such as dyes and pigments, antioxidant; thermal stabilizer; UV absorbent; antistatic agent and surface active agent.

The manner to add the above-described additives, i.e. other resin components, reinforcing agent and/or fillers or others to the liquid crystalline polymer molded in the present invention is not limited.

For example, liquid crystalline polymer, reinforcing agent and/or filler, and if desired, mold-release agent and thermal stabilizer may be fed respectively into a kneading machine and mixed there, or those raw components may be mixed preliminarily in a mixer such as Henschel mixer, super mixer, ribbon blender or tumbler and then, the resulting mixture may be added to the a kneading machine to provide resin composition to be molded. The kneading machine used in this process may be any of conventional machines such as single screw extruder, twin screw extruder, tandem extrude, Banbury mixer or the like.

The melt viscosity of the resin molded in the present invention may vary depending of the additives such as other resin component, reinforcing agent and/or fillers and others, and any of resin compositions of which melt viscosity is less than 800 poise may preferably be used in the present molding method.

The injection-molding machine used in the present invention is not limited and may be any of conventionally used machines. For example, the clumping unit of the machine may be either of straight-hydraulic type (oil pressure type) or toggle type. The injection unit may be those driven by screw, such as screw pre-plasticizing type or in-line screw type, or plunger such as plunger pre-plasticizing type or single plunger type. The direction of the cylinder in the injection unit may be either transverse or vertical. The drive unit may be not only of hydraulic (oil pressure) type but also electric motor-driven type or combined type.

Optimal injection condition may be determined based on the type of the resin, size of the injection-molding machine, shape of the mold and number of the cavities. The molding condition may be as follows: the cylinder temperature may preferably be between the melting point of the resin and melting point + 80°C, more preferably, between the melting point + 10°C and the melting point +

50°C; the mold temperature may preferably be between 20°C and 150°C.

In order to control the linear speed of the molten resin, the molding condition, especially, injection speed of the resin may be adjusted so that the desired speed is obtained. The condition may be selected based on the resin and mold. As the runner becomes thinner, the more linear speed increases. The linear speed may also vary depending on the viscosity of the resin.

The molded products obtained by the present invention are preferably parts of known electric and electronic devices or precision devices such as connectors, relays, tact switches, coil bobbins, volume parts, motor parts such as commutators and separators, coils, quartz oscillators and IC tips.

The injection-molding system for resin comprising an injection-molding machine and a mold comprising a sprue, one or more runners and one or more cavities, characterized in that the cross-sectional area of at least part of the runners is less than 2.0 mm<sup>2</sup> is also within the range of the present invention. The system having 4 or more cavities is preferable.

The mold for injection-molding of resin comprising a sprue, one or more runners and one or more cavities, characterized in that the cross-sectional area of at

least part of the runners is less than  $2.0 \text{ mm}^2$  is also within the range of the present invention. The mold having 4 or more cavities is preferable.

#### EXAMPLE

5           The method for injection-molding of resin of the present invention is described below by way of examples. The present invention is not limited to those examples.

Box type cavity (1) shown in Figure 1 comprising side gate(2) was used. Three types of runners (4) having  
10 cross-sectional areas of  $2.5 \text{ mm}^2$ ,  $1.5 \text{ mm}^2$  and  $0.8 \text{ mm}^2$  respectively were used to construct molds having 8 cavities (Fig. 2) and 16 cavities (Fig. 3). The size of the cavity (1) used in this example was as follows:

inner dimension =  $15 \text{ mm} \times 10 \text{ mm} \times 10 \text{ mm}$

15 wall thickness =  $0.15 \text{ mm}$

side gate =  $0.3 \text{ mm} \times 0.4 \text{ mm}$  square

Thermotropic liquid crystalline polymers (LCP-1 and LCP-2, see below) and the other resins were subjected to injection-molding using injection-molding machine (PS40-  
20 5VSE, NISSEI Plastic Industrial Co., Ltd.; Clumping force: 40tonf) and the above mold. The injection-molding was carried out under the condition, i.e. cylinder temperature, mold temperature, and injection speed, shown below.

The obtained molded product was evaluated  
25 according to the criteria shown below. In case where a

satisfied product was obtained, the shortest molding cycle was determined. Results are shown in table 1 below.

### Resins

5 LCP-1: p-hydroxybenzoic acid/ 2-hydroxy-6-naphthoic acid/ terephthalic acid/ hydroquinone copolymerized liquid crystalline polymer composition comprising 30 wt% of glass fibers (melting point: 325°C, melt viscosity: 350poise)

10 LCP-2: p-hydroxybenzoic acid/ 2-hydroxy-6-naphthoic acid copolymerized liquid crystalline polymer composition comprising 30wt% of glass fibers (melting point: 280°C, melt viscosity: 530 poise)

15 PA46: Polyamide (melting point: 290°C, melt viscosity: could not be measured since the product decomposed at 25°C higher than its melting point) (Stanyl™ TS250F6; Japan Synthetic Rubber Co., Ltd.)

PA66: Polyamide(melting point: 260°C, melt viscosity: 2200 poise) (Leona® FG173; Asahi Kasei Corporation)

20 PBT: Polybutylene terephthalate (melting point: 225°C, melt viscosity: 3400 poise) (Duranex™ 3370; Polyplastics Co. Ltd.)

PPS: Polyphenylene sulfide (melting point: 285°C, melt viscosity: 3500 poise) (Fortron™ 1140A Polyplastics Co. Ltd.)

### Molding Temperature

25 Cylinder temperature

LCP-1: 350°C

LCP-2: 330°C

PA46: 310°C

PA66: 285°C

5 PBT: 250°C

PPS: 310°C

Mold Temperature

LCP-1: 50°C

LCP-2: 50°C

10 PA46: 60°C

PA66: 60°C

PBT: 60°C

PPS: 130°C

Evaluation Criteria

15 ○: All cavities were filled completely

△: small short-shot

×: short-shot

□: flash was occurred

Table 1

|       |                                                | Evaluation (molding cycle: sec.) |        |        |     |       |       |
|-------|------------------------------------------------|----------------------------------|--------|--------|-----|-------|-------|
| mold  | cavities                                       | 8                                |        |        | 16  |       |       |
|       | runner cross-sectional area (mm <sup>2</sup> ) | 2.5                              | 1.5    | 0.8    | 2.5 | 1.5   | 0.8   |
|       | resin composition in the runners (wt%)         | 45                               | 33     | 21     | 34  | 23    | 14    |
| resin | LCP-1                                          | O(17)                            | O(9.2) | O(8.5) | △   | O(12) | O(11) |
|       | LCP-2                                          | O(16)                            | O(8.7) | O(8.0) | △   | O(11) | O(10) |
|       | PA46                                           | O(18)                            | O(12)  | △      | □   | △     | ×     |
|       | PA66                                           | O(20)                            | △      | △      | △   | ×     | ×     |
|       | PBT                                            | O(20)                            | △      | ×      | △   | ×     | ×     |
|       | PPS                                            | O(21)                            | □      | ×      | □   | △     | ×     |

The results shown in table 1 support the fact that the method for injection-molding of resin of the present invention can be carried out in short molding cycle with a mold having multiple cavities, and therefore, the productivity of the molding process is improved. In addition, because of the thin runners, the amount of waste resin, that is, the material molded in the runners, can be reduced.

#### Effect of the invention

The present method for injection-molding of resin can be carried out in a short molding cycle using a mold having multiple cavities, and can reduce waste of resin. The present invention attained improved productivity and reduced cost for manufacturing electronic, electric or precise parts.

## WHAT IS CLAIMED IS

1. A method for injection-molding of resin using a mold comprising a sprue, one or more runners and one or more cavities, characterized in that the cross-sectional area of at least part of the runners is less than 2.0mm<sup>2</sup> and the melt viscosity of the resin, measured at the temperature of 25°C higher than the melting point of the resin and at the shear rate of 1000 sec<sup>-1</sup>, is less than 800 poise
2. The method of claim 1, wherein the ratio of the length of at least part of the runners where the cross sectional area is less than 2.0 mm<sup>2</sup> to the whole length of the runners is 1/4 or more.
3. The method of claim 1 or 2, wherein the cross-sectional area of the runner is less than 1.6 mm<sup>2</sup>.
4. The method of claim 1 or 2, wherein the cross-sectional area of the runner is less than 1.3 mm<sup>2</sup>.
5. The method of any one of claims 1-4, which provides molded products of less than 1g.
6. The method of any one of claims 1-4, wherein the ratio of the amount of resin filled in the runners to the total amount of resin filled in the cavities, runners and sprue is less than 50 wt%.
7. The method of any one of claims 1-4, wherein the mold has 4 or more cavities.



8. The method of any one of claims 1-4, wherein the linear speed of the molten resin flows in the runner at injection is 1000mm/sec or higher.

9. The method of any one of claims 1-4, wherein the resin is a thermotropic liquid crystalline polymer.

10. The method of claim 9, wherein the thermotropic liquid crystalline polymer comprises 2-hydroxy-6-naphthoic acid residue as a monomer component.

11. The method of claim 9, wherein 100 parts by weight of the thermotropic liquid crystalline polymer is admixed with 0.001-1 parts by weight of a mold-release agent before injection.

12. The method of claim 9, wherein 100 parts by weight of thermotropic liquid crystalline polymer pellets are admixed with 0.001-0.05 parts by weight of the mold-release agent before injection so that the agent adheres to the surface of the pellets.

13. The method of claim 9, wherein thermotropic liquid crystalline polymer is a polymer composition comprises 1-50 wt % of reinforcing agent and/or fillers.

14. An injection-molding system for resins comprising an injection-molding machine and a mold comprising a sprue, one or more runners and one or more cavities, characterized in that the cross-sectional area of at least part of the runner is less than 2.0mm<sup>2</sup>.



15. The system of claim 14, wherein the ratio of the length of at least part of the runners where the cross sectional area is less than  $2.0 \text{ mm}^2$  to the whole length of the runners is  $1/4$  or more.

5 16. The system of claim 14 or 15, wherein the mold comprises 4 or more cavities.

10 17. A mold for injection-molding of resin comprising sprue, one or more runners and one or more cavities, characterized in that the cross-sectional area of at least part of the runners is less than  $2.0 \text{ mm}^2$ .

18. The mold of claim 17, wherein the ratio of the length of at least part of the runners where the cross sectional area is less than  $2.0 \text{ mm}^2$  to the whole length of the runners is  $1/4$  or more.

15 19. The mold of claim 17 or 18, which comprises 4 or more cavities.

