Title	エチレンプロピレン共重合体合成におけるMg(OEt)2型Ziegler-Natta触媒構造の影響
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論　　文　　題　　目	Comprehension of Mg(OEt)2-based Ziegler-Natta Catalyst Structure in Heterophasic Polypropylene Characteristics
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論文の内容の要旨

Ziegler-Natta catalyst is one of the important catalysts in polyolefin production. According to its complex structure over multi-length scale and its heterogeneity, a precise recipe for achieving required catalyst performances and polymer properties has not been established.

Focusing on $Mg(OEt)_2$-based Ziegler-Natta catalyst, the morphology regulation is an intensive examined issue. At present, many parameters are known that they had effects on the morphology of $Mg(OEt)_2$ and upcoming catalyst. However, it is still unclear that how the parameters influence on the morphology. The influences of Mg source were investigated. The results showed $Mg(OEt)_2$ was developed through three steps; (i) seed generation, (ii) seed growth and isolation, and (iii) particle growth. Also, large Mg source could give large particle size of $Mg(OEt)_2$. Afterwards, large-size $Mg(OEt)_2$ was used for high impact propylene copolymerization. According to the system, the amount of produced polypropylene (PP) could not be observed, although it was an important for determining copolymer content. Thus, the equation from the correlation between flow rate and yield was used. The results showed that the equation had sufficient accuracy as seen in well consistent with decane extraction results. Finally, the development of PP particle and its performances as a reactor granule for high impact polypropylene was examined. The results showed that pore volume and its distribution were important factors for determining rubber capacity and dispersion.

This research showed inclusive investigation throughout the process of polyolefin production. The results showed an effective alternative to design the catalyst through the modification of precursor. Also, the importance of polymer characteristics was stressed. Thus, it is high possibility to apply this finding to modify the catalyst structure for achieving required polymer properties.

Keywords: Magnesium ethoxide, High impact polypropylene, Reactor granule, Particle development,

Rubber dispersion.

論文審査の結果の要旨

Ziegler-Natta Catalysts have long been keeping their industrial and academic importance for olefin polymerization. Especially, they produce almost 100 million tons of polyolefins per year in all over the world.
However, the structure of Ziegler-Natta catalysts have never been clarified according to their complexity over multi-length scale and their heterogeneity. Though the great improvements of the industrial catalysts have been achieved, a precise recipe for achieving required catalyst performances and polymer properties has not been established yet.

Focusing on Mg(OEt)2-based Ziegler-Natta catalyst, the morphology regulation is an intensive issue for research. At present, many parameters are known that they affect the morphology of Mg(OEt)2 and upcoming catalyst. However, it is still unclear that how the parameters influence the morphology. The influences of Mg source were first investigated. The results showed Mg(OEt)2 was developed through three steps; (i) seed generation, (ii) seed growth and isolation, and (iii) particle growth. Also, large Mg source could give large particle size of Mg(OEt)2. Afterwards, large- size Mg(OEt)2 was used for making the catalyst to produce high impact propylene copolymer. According to the system, the amount of produced polypropylene could not be determined, although it is important for determining copolymer content. Thus, the equation from the correlation between monomer flow rate and polymer yield was used. The results showed that the equation had sufficient accuracy as seen in well consistent with decane extraction results. Finally, the development of polypropylene particle and its performances as a reactor granule for high impact polypropylene was examined. The results showed that pore volume and its distribution were the important factors for determining rubber capacity and dispersion.
This research showed inclusive investigation throughout the process of polyolefin production. The results showed an effective alternative to design the catalyst through the modification of precursor. Also, the importance of polymer characteristics was stressed. Thus, it is high possibility to apply this finding to develop the catalyst having the structure for achieving required polymer properties.

As mentioned above, this dissertation greatly contributes to the understanding of the specific structures and features of the most important industrial olefin polymerization catalyst. Therefore, this dissertation can be recommended for the doctor degree of JAIST in the field of materials science.