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1. Introduction

Human activities have always consumed energy. In ancient times, human beings started cultivation to increase the amount of energy taken in via food consumption, as a result, humans had time to invent culture and technology. That is to say, the surplus of energy made for an affluent society. This trend was dramatically accelerated by the Industrial Revolution. This era initiated the consumption of fossil fuels in every aspect of our life, even in cultivation. This means that our life is supported by energy consumption.

Fossil fuels are formed from dead, buried organisms and it takes millions of years to generate fossil fuels, meaning that our consumption rate is too fast as a sustainable energy resource. One of the discussions about this issue was written by the Club of Rome (Meadows et al., 1972). Evaluation of the technology from the perspective of energy consumption is one of the most important discussions within engineering.

Because fossil fuels are limited, the amount of energy which we can use will have to peak at some point (Hubbert, 1956). Predicted oil peaks range between 2010 and 2040 according to the review by Richard (Kerr, 2007). The International Energy Agency (IEA) reports in the World Energy Outlook 2010 that the peak of conventional oil came on 2007. On the other hand, rapid economic growth of developing nations means that global demand for energy is growing. These situations indicate that the energy issue will become more serious in the near future. So, reduce, reuse and recycling (3R’s) are more and more important for the resource problem.

Supercritical fluid (SCF) is expected to be one of the technologies used for the recycling of plastics because of its reactivity. On the other hand, there are general discussions on the energy consumption of the recycling of plastics, which indicate that the recycling can be wasteful of energy in some cases (Takeda, 2008). If much more energy is consumed for recycling than to make virgin material, recycling cannot save resources. This is especially the case when using SCF; energy consumption should be considered because large amounts of supercritical water or methanol require large amounts of energy. A phase diagram of methanol is shown in Fig. 1. It indicates that with SCF, the temperature and the pressure of the materials are higher than critical points. Critical points of water and methanol are shown in Table 1. Critical temperatures of the alcohol and water are higher than 250°C. Figure 2 shows the internal energy of methanol (Reuck & Craven, 1993) which increases with temperature at 10 MPa, which means that making such a high temperature and pressure state requires large amounts of energy.
So, a technology which can minimize the amount of SCF was required for the recycling process using SCF to avoid wasting energy.
It was reported that the extruder can be used as a feeder and a reactor for the supercritical alcohol (Goto et al., 2005). Supercritical fluid can be injected into the polymer because the extruder can pressurize the polymer, which means that such process can minimize the amount of supercritical fluid and energy consumption. Here, LCA (Life Cycle Assessment) and EPR (Energy Profit Ratio or EROI: Energy Return on Investment) is applied to evaluate the energy consumption of this technology.

General knowledge of the LCA and SCF was explained first. Then, chemical reaction for the recycling of Si-XLPE in the supercritical methanol (Goto et al., 2008; Goto et al., 2001) was shown. After that, the technology which uses the extruder for the chemical reaction in supercritical fluid to develop the continuous process for industrialization (Goto et al., 2011) was introduced. The obtained recycled PE properties satisfied the requirements of Japan’s industrial standard of insulation for 600V cross-linked polyethylene cable (600V XLPE cable) (Goto et al., 2006). Then energy consumption to make recycled PE is compared with that of virgin PE by LCA as one example of the evaluation of energy consumption for recycling technology.

2. Life Cycle Assessment (LCA) for evaluation of recycling technology

The final aim of LCA is the evaluation of the influence of human activity on the environment and society to select the best combination of products, services or technology. So, it is useful for the development of technology to take an objective view. LCA is quantitative analysis from raw material extraction through to processing, distribution, use, maintenance, recycling and disposal (Ito et al., 2007). It is useful for engineers, who tend to take the seeds oriented approach, to take a wider view of the technology. The procedure is standardized by ISO 14040 and 14044 in four distinct phases as follows:

1. **Goal and Scope**

   It is impossible to gather all of information which is required for LCA. It is required to consider which raw materials and process of works affect the goal, to determine the boundaries of investigation. In this phase, function unit, system boundaries, assumptions, the allocation methods are defined.

2. **LCI**

   Next, the scenario of life cycle is assumed from and to nature system. The data which cannot be directly measured can be compensated for by the database of unit base for LCA.

3. **LCIA**

   The effect of the obtained results are discussed with regard to the goal and scope, such as effect on global warming, acid rain or resource problem. Normalization by weighting is conducted depending on goal and scope. But weighting will also make the meaning of the obtained data obscure in some cases. So, when it comes to studying energy consumption, it is preferable not to use weighting.

4. **Interpretation**

   The key purpose of this phase is checking the credibility and accuracy of data, and sensitivity of parameters. It is also evaluated if the goal and scope is achieved.

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3. Energy Profit Ratio (EPR)

The concept of energy profit ratio is one of the methods for the interpretation of the results of LCA. EPR originates from the idea of the ‘rabbit limit’ which is based on the case of rabbit hunting shown in Figure 3 (Gunther).

![Image of rabbit hunting](image)

**Fig. 3. Rabbit hunting**

When you have to chase a rabbit to obtain food, you need to consume energy for hunting. If the energy taken for hunting the rabbit is greater than that of the energy obtainable from eating the rabbit, rabbit hunting is not sustainable. EPR is determined using the following equation (Amano, 2008):

\[
\text{EPR} = \frac{\text{yielded energy}}{\text{consumed energy}}
\]

EPR must be more than 1 if the activity is sustainable. The larger the amount of EPR means the greater the efficiency of the energy resource. So, EPR is also convenient for consideration of sustainatbility. This method was originally used to evaluate the value of energy resource such as oil, coal and so on. Here, we adopt the concept of EPR for recycling. The final aim of the recycling technology is to contribute to the sustainability of human life, that is to say, evaluation of recycling technology requires the concept of the rabbit limit. In the case of recycling, it must be evaluated if consumption of energy decreases. Here, energy consumption ratio (ECR) is defined as follows (Goto et al., 2009):

\[
\text{ECR} = \frac{\text{Consumed energy for virgin product}}{\text{Consumed energy for recycled products}}
\]

In this case, it is assumed that the recycled product is as valuable as the virgin product, such as in the case of horizontal recycling. This concept can also be adopted in the case of the improvement of industrial processes, distribution of products or supply chain to compare before and after improvement.
4. Supercritical fluid for recycling of plastics

A comparison of properties between gas, liquid and SCF is shown in Table 2. The density of SCF is 1/10-1/2 of liquid. On the contrary, viscosity close to the gas and diffusion constant is in the middle of gas and liquid. As a result, SCF has strong chemical activity. So, SCF is expected to be the method for decomposition of harmful chemical materials (Sako et al., 1998). Moreover, dielectric constant decreases around the critical point, which means that the properties as solvent are change around critical points (Saito, S.,1996). For example, water can be mixed with oil above the critical point of water.

When you need to use the reaction between water and polymer which is not dissolved in water, supercritical water can be used without other solvents. Another merit of using supercritical fluid is the ability of diffusion. Cross-linked polymers are difficult to stir to accelerate the chemical reaction because cross-linking of polymers robs the liquidity of polymer. The high diffusion constant of SCF is positive effect for water or methanol to diffuse into the polymer without stirring. All these properties lead us to expect that SCF has advantages for the chemical recycling of cross-linked polymers.

<table>
<thead>
<tr>
<th>Properties</th>
<th>Gas</th>
<th>Supercritical fluid</th>
<th>Liquid</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density (kg/m³)</td>
<td>0.6~1</td>
<td>200~900</td>
<td>1000</td>
</tr>
<tr>
<td>Diffusion Constant (m²/s)</td>
<td>10⁻⁵</td>
<td>10⁻⁷~10⁻⁸</td>
<td>10⁻⁹</td>
</tr>
<tr>
<td>Viscosity (Pa⋅s)</td>
<td>10⁻⁵</td>
<td>10⁻⁵~10⁻⁴</td>
<td>&lt;10⁻³</td>
</tr>
</tbody>
</table>

Table 2. Properties of supercritical fluid, liquid and gas

5. Application of chemical reaction in supercritical fluid

Here, a general analysis of chemical reaction in supercritical fluid is given. Chemical reaction in SCF is basically the same as chemical reaction at room temperature. SCF is useful if you need to accelerate the chemical reaction kinetics. Most of the research of chemical reaction of supercritical water and alcohol is transesterification. For example, decomposition of carbonate (Arai, 2002), and also applied for the siloxane bond such as decomposition of silica gel (Kitahara, 1969; Kitahara et al., 1970) or silicon rubber. All of this research takes advantage of acceleration of chemical reaction in supercritical fluid.

On the other hand, the disadvantages of SCF are difficulties in the development of the process for commercialization because it is difficult to put solid material into the SCF and take materials from the SCF. This difficulty is mainly derived from the properties of SCF which is close to the highly pressurised gas. So, development of a continuous process for SCF is discussed in the following section.

6. Recycling technology for Si-XLPE using supercritical fluid

Silane cross-linked polyethylene (silane-XLPE) is widely used as an insulation material for wires and cables (Maruyama, 2003). Recycling of cross-linked polymers poses one of the most difficult problems in the recycling of polymer wastes. Most of industrial waste silane-XLPE is buried in landfills or burned as fuel, because silane-XLPE is difficult to process due to its low fluidity derived by cross-linking elements formed by the siloxane bond. The research for recycling of silane-XLPE as polymer is as follows:

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1. Clashed XLPE pellet is mixed with virgin polyethylene using a twin screw extruder (Tokuda et al., 2003),
2. Powdered XLPE is used as a filler of polymer (White, et al., 2000; Voldner, 2000).

These technologies require further investigation of the application for recycled products and the development of the market due to the apparent difficulty in using directly the recycled product as insulation of wire or cables (horizontal recycling).

6.1 Chemical reaction for recycling of Si-XLPE

Chemical reaction in supercritical alcohol is shown in Figure 4. Siloxane bond (Si-O-Si) can be decomposed into the alkoxide group (-OR) or the hydroxyl group (-OH) by supercritical alcohol selectively.

The condition for selective decomposition of cross-linking bond is investigated. The gel fraction and molecular weight of products against decomposition temperature is shown in Figure 5. The gel fraction vanished at over 300°C when methanol was used as supercritical fluid. Using water, a temperature over 370°C was necessary to decline the gel components to 0wt%. These results indicate that the cross-linking element was completely decomposed at over 300°C using methanol and at 370°C using water. Molecular weight was decreasing at over 340°C using water and methanol. This result agreed with the general knowledge about the thermal decomposition temperature of PE.

By using water, the molecular weight of the products would be lower than raw PE whenever the gel fraction of the products decreased to 0%. It is indicated that the reaction in supercritical water was not selective only for the cross-linking element, but also for the main chain of PE.

In the case of methanol, at temperatures between 300°C – 340°C and pressure more than critical pressure, the cross-linking element was completely decomposed and the molecular weight of the recycled PE was kept the same as virgin PE before being cross-linked. It means that using supercritical methanol is necessary to obtain thermoplastic PE which has the same molecular weight as virgin PE.

Fig. 4. Chemical reaction of crosslinking bond in supercritical alcohol
6.2 Process design

One of the most important issues with supercritical fluid technology is the development of a continuous industrial process for the solid material. The conventional process for SCF is compared with the process using an extruder that is developed for the recycling of cross-linking polyethylene in Table 3. The most fundamental method is the batch process, but in using this process, the reactor should be cooled to take out the product and put raw materials in the reactor. That is to say, efficiency of heat energy is low. In the case of a semi-continuous process, heat efficiency is better than that of the batch process, but the amount of water or alcohol required is about the same as the batch process because the pressure is applied by the materials used as SCF. The amount of SCF cannot be saved in this process, which means improvement on the efficiency of energy is small. In the general continuous process for the supercritical fluid, the solid material was crushed into small particles and dispersed in the solvent to make a slurry. Then, a liquid pump was used to feed the slurry, but this kind of process also has a problem for industrialization as follows. It is very expensive to crush the materials into particles to make powder for slurry. Moreover, the process requires much more solvent than the solid material itself. Ordinary, 10 of solvent required for one of solid material to make slurry. To solve these problems, we developed a new continuous process for the supercritical fluid using an extruder. Extruder is used as the processing machine for polymers. Pressure and heat can be applied to the polymer at 400°C and 50 MPa with the extruder. The polyethylene which is crushed into 5 mm in diameter can be fed to extruder. This means that the new process using extruder does not require making powder of solid material. Furthermore, a minimum amount of supercritical fluid is required for the chemical reaction into pressurized polymer.

![Gel fraction and number molecular weight of recycled product made by supercritical methanol and water. Black circle: made by methanol, White circle: made by water](image-url)
Table 3. Comparison of the process for supercritical fluid

<table>
<thead>
<tr>
<th>Process Type</th>
<th>Necessity of break into pieces</th>
<th>Heat Efficiency</th>
<th>Amount of Solvent</th>
<th>Continuous process</th>
<th>Productivity</th>
</tr>
</thead>
<tbody>
<tr>
<td>Batch</td>
<td>Not necessary</td>
<td>× Low</td>
<td>× Large</td>
<td>× Impossible</td>
<td>× Low</td>
</tr>
<tr>
<td>Pump</td>
<td>Necessary</td>
<td>Δ</td>
<td>× Large</td>
<td>Δ Possible</td>
<td>Δ</td>
</tr>
<tr>
<td>Semi Continuous</td>
<td>Not necessary</td>
<td>Δ</td>
<td>× Large</td>
<td>× Impossible</td>
<td>Δ</td>
</tr>
<tr>
<td>Extruder</td>
<td>Not necessary</td>
<td>× High</td>
<td>× Small</td>
<td>× Possible</td>
<td>× High</td>
</tr>
</tbody>
</table>

The process was designed to make recycled PE from crushed Si-XLPE waste continuously and automatically. A scheme of the continuous process is shown in Figure 6. The twin screw extruders are made by Japan Steel Works and were used as equipment for the chemical reaction (Ext-Chem) and degassing (Ext-Degas). Methanol was fed using a high pressure pump from the methanol tank and heated to supercritical state by the heater before it was injected to the Ext-Chem. The tubular reactor was attached to the Ext-Chem to keep Si-XLPE and methanol for more than 15 min at 10 MPa, 335°C. The pressure control valve was connected to the tubular reactor. Ext-Degas was mounted to the outlet of the pressure control valve to separate the recycled PE and the gas. Pelletizer was prepared to cut the recycled PE strand into the pellets. The whole electric power demand for this process was measured using the attached ammeter to evaluate the energy consumption.
The Si-XLPE pellet with 30% in the degree of gel fraction was gathered from the factory for the production of 600V XLPE cable. The methanol used here was made by WAKO Chemicals. 

40kg/hour of Si-XLPE was charged to the extruder from the hopper, then 10 phr of alcohol was injected to the Ext-Chem. The injected supercritical alcohol was kept at over 300°C at 10 MPa. The reactor was kept at 330°C and 10 MPa. The Ext-Degas was set at 200°C to extrude the recycled PE.

Electric power demand for this process is shown in Figure 7. Power demand is stable at around 160A 200V in 10 hours, which means that the process was well controlled. The average electric power demand in 10 hours is 159.3A, 200V.
6.3 Properties of the products

The appearance of cable extrusion of the recycled PE and the 600V rating XLPE cable (cross section of conductor 38mm², thickness of insulation:1.2mm) is shown in Figure 8. The processability of the recycled PE was good enough to be used as cable insulation.

![Cable extrusion](image)

(a) Cable extrusion  (b) Recycled 600V rating XLPE cable

Fig. 8. Trial for cable recycling

The properties of the recycled cable are shown in Table 4. The initial mechanical and electrical properties of the cable satisfied JIS C3605.

<table>
<thead>
<tr>
<th>Items</th>
<th>Recycled Cable</th>
<th>Required</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tensile Strength at break(MPa)</td>
<td>20</td>
<td>&gt;10</td>
</tr>
<tr>
<td>Elongation(%)</td>
<td>500</td>
<td>&gt;200</td>
</tr>
<tr>
<td>Volume Resistance(MΩ/km)</td>
<td>10×10⁵</td>
<td>&gt;1.5×10³</td>
</tr>
<tr>
<td>Breakdown Voltage(kV)</td>
<td>28</td>
<td>-</td>
</tr>
<tr>
<td>Heat Deformation(%)*</td>
<td>0.9</td>
<td>&lt;40</td>
</tr>
</tbody>
</table>

* : At 120°C under the force of 20N

Table 4. Properties of the recycled cable

7. Life cycle assessment of recycling of silane-XLPE

600V XLPE cable is the target application of this technology. The structure of 600V XLPE cable is shown in Figure 9. The conductor is covered by insulation and sheath.

![Structure of 600V XLPE cable](image)

Fig. 9. Structure of 600V XLPE cable.

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Figures 10 and 11 illustrate the process for production of 600V XLPE cable using virgin PE and recycled PE as insulation respectively. The same copper (Cu) and polyvinyl chloride (PVC) can be applied to the virgin and the recycled product as the conductor and sheath respectively.

It is reported that recycled PE can be used the same as the virgin PE (Goto et al., 2006), which means that energy used for the cable processing should be the same in each product. So, the system boundary was determined as the hatched box in Figures 10 and 11. Energy consumption to produce the recycled PE was compared with that of the virgin PE in this study. The life cycle energy assessment of the whole process for the production of cable is reported in another paper (Goto, 2004).

![Fig. 10. Process for the the virgin products.](image1)

![Fig. 11. Process for the recycling products.](image2)

Si-XLPE was assumed to be gathered from the cable factory and recycled at that site. Energy unit bases of virgin PE, methanol, electric power and steel were represented in Table 5 (Amano, 2006). These values were converted to the energy per 1kg of PE for calculation.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Unit base</th>
</tr>
</thead>
<tbody>
<tr>
<td>PE</td>
<td>$U_{PE}$</td>
</tr>
<tr>
<td>Methanol</td>
<td>$U_{MeOH}$</td>
</tr>
<tr>
<td>Electric power</td>
<td>$U_{ele}$</td>
</tr>
<tr>
<td>Steel</td>
<td>$U_{Fe}$</td>
</tr>
</tbody>
</table>

Table 5. Energy unit base

Energy for the recycled PE (Er) was represented as following equation.
\[ E_r = U_{\text{MeOH}} + E_1 + E_2 \]  

(1)

\( E_1 \) and \( E_2 \) are the energy consumption for crushing Si-XLPE and for the supercritical processing respectively for 1kg of PE. Energy consumption for each unit process is represented as follows:

\[ E_n = E_n' + E_n'' \]  

(2)

\( E_n' \) is electric energy consumption per 1kg of PE, which was estimated equal to the volt-ampere(VA) per 1kg of PE. Measured electric demand shown in Figure 6 was used for evaluation of that of the supercritical processing. On the other hand, electric demand for the crusher was estimated as 30% of the electric capacity of crusher which can treat 40kg/hour of the Si-XLPE waste.

\( E_n'' \) is energy requirement to make the equipment in the unit process per 1kg of PE, which is represented as following equation.

\[ E_n'' = \frac{E_n'''}{V_n \times D_n \times Y_n \times R_n} \]  

(3)

- \( E_n''' \): Energy requirement to make the equipment (kcal)
- \( V_n \): Productivity of PE (kg/hour)
- \( D_n \): Operation time in the year (hour/year)
- \( Y_n \): Life span of the equipment (year)
- \( R_n \): Occupation rate of equipment for the product(-)

Equation (4) was used to estimate:

\[ E_n'' = \sum_i U_i W_i \]  

(4)

\( W_i \) is weight of the part of equipment (kg). \( U_i \) (kcal/kg) is unit base of the materials of which part of equipment consist. Equipment used for crusher and for the supercritical treatment mainly consist of steel. So, we calculate the energy consumption for the production of the equipment as following equation instead of equation (4).

\[ E_n'' = U_{Fe} W_n \]  

(5)

\( W_n \) is total weight of the equipment for the unit process (kg). \( U_{Fe} \) is unit base of steal (kcal/kg).

The found energy consumption is shown in Table 6. The required energy for the recycling of Si-XLPE was smaller than that of virgin PE. These results indicate that the recycling of Si-XLPE using supercritical alcohol can save energy consumption.

Moreover, further information about the recycling process is revealed as follows. Energy required for the construction of the equipment is smaller than that of the electric energy used for the operation of the equipment. Electric energy was mainly used as the energy source for the heaters and for the motors. So, these results indicate that the heat insulation and the efficiency of the motors are important matters in saving energy rather than the size reduction of the equipment.

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The general aim of recycling is saving resources. Energy is the most fundamental resource because every human activity, including mining, requires energy. So, LCA on energy is a useful tool for the development and evaluation of the meaning of recycling technology.

<table>
<thead>
<tr>
<th></th>
<th>Recycled PE (kcal/kg)</th>
<th>Virgin PE (kcal/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_{\text{MeOH}}$</td>
<td>244.4</td>
<td>-</td>
</tr>
<tr>
<td>$E_{\text{PE}}$</td>
<td>-</td>
<td>7054.0</td>
</tr>
<tr>
<td>$E_1$</td>
<td>0.6</td>
<td>-</td>
</tr>
<tr>
<td>$E_1'$</td>
<td>103.7</td>
<td>-</td>
</tr>
<tr>
<td>$E_2$</td>
<td>17.0</td>
<td>-</td>
</tr>
<tr>
<td>$E_2'$</td>
<td>2240.2</td>
<td>-</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td>2605.9</td>
<td>7054.0</td>
</tr>
</tbody>
</table>

Table 6. Energy analysis of the recycled PE.

ECR is calculated from the data shown in table as following equation.

$$ECR = \frac{7054.0}{2605.9} = 2.71$$

The obtained ECR is 2.71 which means that recycling of Si-XLPE is the technology that can contribute for saving energy. This example also indicates that the LCA and EPR methods are useful for evaluation of recycling technology. Moreover, these methods will be effective in the development of the recycling process to look at the final target.

8. Conclusion

The energy consumption for the recycling of Si-XLPE was studied. It is indicated that energy can be saved if the extruder is applied as a feeder and a reactor for the reaction in supercritical methanol. A life cycle energy assessment can give an answer to the criticism of wasting of energy because of the high pressure and temperature of supercritical methanol. Moreover, the results give us a principle in the development of a more efficient process.

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This book deals with several aspects of waste material recycling. It is divided into three sections. The first section explains the roles of stakeholders, both informal and formal sectors, in post-consumer waste activities. It also discusses waste collection programs for recycling. The second section discusses the analysis tools for recycling system. The third section focuses on the recycling process and optimal production. I hope that this book will convey both the need and means for recycling and resource conservation activities to a wide readership, at both academician and professional level, and contribute to the creation of a sound material-cycle society.

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