Friction Properties of Polymer Gel with High Mechanical Strength

1. Introduction

Polymer gels each comprise a polymer skeleton of chemically bonded three-dimensional network structure and a significant liquid component (solvent). Compared with other polymer materials such as plastic and rubber, conventional polymer gels have often been brittle materials featuring lower mechanical strength and lower elastic modulus. This may be readily understood when considering examples of familiar polymer gels such as jelly and tofu soy bean curd.

The swelling degree of a polymer gel will vary, owing to external stimuli such as temperature, pressure, hydrogen ion index (pH value), light, and electric field, whereby the volume of polymer gel is altered. Focusing on this feature, applications of polymer gels as functional and intelligent materials have been researched and commercially adopted for sensors and actuators.

Recent developments regarding polymer gels include high-strength gel types known as double network gels \(^1\) and nano-composite gels \(^2\) as well as high-tenacity gel called topological gel \(^3\), whereby brittleness-induced quality issues with polymer gels resulting from poor handling have been much alleviated. Consequently, the scope of utilization of polymer gels in industrial applications seems to expand.

Incidentally, though being solid matter, polymer gels consist primarily of liquid: as such it is known that they feature significantly low friction coefficients and unique friction behavior. Their unique friction mechanism \(^4\) has been increasingly elucidated. However, many examples thus far reported on friction properties of polymer gels have been associated with combinations of water-solvent polymer gels and smooth-surfaced glass plates.

Now, let us consider utilizing the excellent low-friction features of polymer gels in order to reduce friction loss that occurs between machine components. In this case, the solvent for gel is needed, for example, not to corrode nearby metal parts, solidify, evaporate, or boil in a wider temperature range.

In this technical paper, we hereafter report the friction properties of water solvent-free double network gel (refer to \(\text{Fig. 1};\) hereinafter referred to as DN gel) \(^1\) used in conjunction with a steel (bearing steel: SUJ2) under various conditions.

2. Overview of the DN gel

A polymer constituting a skeletal component in a polymer gel has a three-dimensional network structure when situated in a solvent. A polymer gel in which the bonds of polymer chains forming the network (that is, cross-links) are covalent is known as chemical gel; if the bonds are not covalent bonds, the polymer gel is called physical gel.

Because cross-links in a chemical gel are formed by covalent bonds, the network structure once formed is...
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It is estimated that the DN gel has a unique structure such as a one shown in Fig. 3, known as the sea-island structure. Incidentally, Fig. 3 provides a schematic diagram, and actual DN gel possesses nonuniformity in density of cross-links.

The sea phase, which is a matrix, consists of a mixed phase comprising a fine network (high cross-linking density) and a coarse network (low cross-linking density), while the island phase virtually comprises soft gel alone. The most outstanding feature of DN gel is a ratio of hard gel to soft gel: more specifically, the composition of DN gel is designed such that the volume of soft gel is about 20 times as great as that of hard gel.

Having such a unique structure, the DN gel has compressive strength in a range of 10 to 50 MPa. This strength is 10 to 100 times as great as that of each gel constituting the DN gel. Furthermore, as shown in Fig. 1, even when compressed by about 90%, the DN gel does not fail: when the load is removed, it restores its original shape like a rubber material.

The high strength mechanism of the DN gel cannot be explained by using a conventional fracture model that can reflect elasticity alone: this mechanism seems very stable, being insoluble and infusible. In contrast, a physical gel is characterized in that its cross-links are liberated by external factors including temperature, thereby reversibly changing from liquid to solid or vice versa. Our DN gel is a type of polymer gel that belongs to a chemical gel category.

For a given chemical gel, a uniform cross-linking reaction is difficult to achieve, and as a result, a certain degree of nonuniformity in density with cross-links occurs in a process of cross-linking reaction (see Fig. 2, red circles represent higher density cross-links).

A network structure involving nonuniform density with cross-links will exhibit differences in the degree of "elongation (tensile force)" on the individual polymer chains across cross-links. Also, there will be difference in degree of transformation (elastic modulus) between the higher cross-linking density area and the lower cross-linking density area. Because of these reasons, when an external force is applied to a gel having nonuniformity in density of cross-links, the force is not evenly exerted onto the polymer links, and fracture will occur at a particular polymer link where a greatest tension takes place.

Any gel material consists primarily of liquid, and contains only a small amount of skeleton component that is responsible for mechanical strength, and accordingly, it is essentially a low-strength material. In addition, while an external force is being applied to a chemical gel, such fracture of polymer chains continuously occurs, and, consequently, chemical gel is a very brittle, low-strength material.

Fig. 1 Compression property of DN gel


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Fig. 2 Schematic drawing of polymer structure of chemical gel

Sea phase: mixed phase consisting of hard gel and soft gel

Island phase: phase primarily consisting of soft gel
to derive from the unique structure of the DN gel comprising both hard gel and soft gel rather than from mechanical properties of each material\(^5\).

### 3. DN gel specimen

As previously mentioned, the DN gel comprises two polymer gel types, each having a polymer network structure independent of the other: since much of the soft gel exists in a mixed phase comprising hard gel and soft gel, the DN gel boasts greater mechanical strength.

The specimen used is the DN gel whose hard gel is poly (2-acrylamido-2-methylpropanesulfonate) gel (hereafter simply referred to as PAMPS gel) and whose soft gel is polyacryl amide gel (hereafter simply referred to as PAAm gel), wherein the cross-linking agent used is methylene bis-acryl amide (hereafter simply referred to as MBAAm).

Having such composition, the DN gel is hydrophilic and the solvent usually used is water. For this technical paper, ethylene glycol was used as the solvent that features a solidifying point lower than that of water and a boiling point higher than that of water.

Table 1 summarizes the composition of the DN gel specimen used, and Fig. 4 shows the chemical structures of the raw materials used.

The compression properties of the DN gel specimen using ethylene glycol solvent are provided in Table 2.

### 4. Friction properties of DN gel

#### 4.1 Test conditions

The test rig adopted for the sliding test is schematically illustrated in Fig. 5. The block portion in this diagram represents the DN gel test piece, and the disk portion stands for a metal mating material. The substance of the metal mating material used is SUJ2.

The load was applied onto the test piece, and the mating material was turned, thereby the sliding test was performed. The friction coefficient was calculated by dividing the friction force detected with a load cell by the test load.

The test conditions applied are summarized in Table 3, which gives the parameters about friction properties for the DN gel and SUJ2 material. The parameters include the load, sliding velocity, surface roughness of the mating material, and test temperatures. This technical report hereafter describes the effects of these parameters.

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<table>
<thead>
<tr>
<th>Table 1 Composition of DN gel</th>
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<tbody>
<tr>
<td><strong>Polymer composition ratio</strong></td>
</tr>
<tr>
<td>Hard gel</td>
</tr>
<tr>
<td>Soft gel</td>
</tr>
<tr>
<td>Solvent</td>
</tr>
</tbody>
</table>

※Boiling point of ethylene glycol: 198˚C, solidifying point: -13˚C

<table>
<thead>
<tr>
<th>Table 1 Compression property of DN gel</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Test piece dimensions</strong></td>
</tr>
<tr>
<td>5 × 5 × 15.6</td>
</tr>
<tr>
<td><strong>Load</strong></td>
</tr>
<tr>
<td>2.0, 4.9, 9.8, 24.5</td>
</tr>
<tr>
<td><strong>Sliding velocity</strong></td>
</tr>
<tr>
<td>0.5, 1.0, 1.4, 3, 4, 6, 7, 9, 10</td>
</tr>
<tr>
<td><strong>Mating material</strong></td>
</tr>
<tr>
<td><strong>Sliding track dia.</strong></td>
</tr>
<tr>
<td>Φ23</td>
</tr>
<tr>
<td><strong>Time</strong></td>
</tr>
<tr>
<td>5-30 (per time setting)</td>
</tr>
<tr>
<td><strong>Temperature</strong></td>
</tr>
<tr>
<td>25, 50, 80</td>
</tr>
<tr>
<td><strong>Atmosphere</strong></td>
</tr>
</tbody>
</table>

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Fig. 4 Chemical structures of the materials

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![Fig. 5 Sliding test method](image-url)
4.2 Test result (part 1)
-Effects of surface roughness of mating material, load, and velocity-

Fig. 6 shows the effects of surface roughness of mating material, load, and velocity on the friction behavior of the DN gel having ethylene glycol solvent (temperature 25˚C).

For the test, several mating material types, each having a unique degree of surface roughness were used, whereby the surface roughness ranged from a very smooth surface equivalent to that of a rolling contact bearing raceway surface to the mating material surface of a sliding bearing.

In any case, the kinetic friction coefficient $\mu_k$ is less than about 0.01, and under the test conditions, the DN boasts very low friction coefficient. However, when the surface roughness of mating material is as high as 0.4 μmRa, the kinetic friction coefficient of the DN gel is somewhat higher.

When an ordinary plastic material is allowed to slide...
relative to SUJ2 under similar conditions, a lubrication state known as boundary lubrication usually occurs and the resultant kinetic friction coefficient \( \mu_k \) is approximately 0.1. From this fact, it should be understood that the DN gel having ethylene glycol solvent features unconventional low friction properties when sliding relative to SUJ2.

Incidentally, the DN gel having ethylene glycol solvent exhibits unique friction behavior, that is, slow increase in kinetic friction coefficient \( \mu_k \). Usually, this trend is experienced in the hydrodynamic lubrication state.

### 4.3 Test result (part 2)

- **Effects of temperature, load, and velocity**

Fig. 7 illustrates the effects of load and sliding velocity onto the friction behavior of the DN gel having ethylene glycol solvent with several atmospheric temperature settings (25˚C, 50˚C and 80˚C) (surface roughness of mating material: 0.05 µmRa).

At 25˚C, the DN gel exhibits excellent friction coefficients, that is, kinetic friction coefficient \( \mu_k \) as low as 0.01 or smaller in a very wide range of load and sliding velocity. However, at the higher temperature range, the kinetic friction coefficient \( \mu_k \) of the DN gel tends to significantly increase at low-speed, high-load condition settings.

Though the kinetic friction coefficient of the DN gel, \( \mu_k \), is high at a limited region in low sliding velocity range, the \( \mu_k \) slowly increases as the sliding velocity increases. Under any test temperature setting in the regions where the kinetic friction coefficient of the DN gel, \( \mu_k \), increases, the DN gel boasts excellent low friction quality, that is, a kinetic friction coefficient \( \mu_k \) of 0.01 or smaller.

### 5. Discussion

The DN gel having ethylene glycol solvent was allowed to slide against a mating material (SUJ2), and the kinetic friction coefficient, \( \mu_k \), was thereby determined. At the same time, with a similar test rig, a variety of polymer materials including conventionally used plastic materials were tested to determine their kinetic friction coefficients. Consequently, it has been learned that the kinetic friction coefficient, \( \mu_k \), of the DN gel is very low; about 1/10 as low as that of the other examined polymer materials. Beginning with this finding, we have attempted to estimate a possible lubrication state that can occur with the DN gel having ethylene glycol solvent under the test conditions adopted for the present research. The method for estimation is described below.

Let us assume that under the test conditions mentioned above, the DN gel takes a hydrodynamic lubrication state by the so-called wedge effect and that the DN gel in this state can be expressed with the model in Fig. 8.

With this model, the DN gel test piece is assumed to be a fixed piece and the mating material (SUJ2) is assumed to move to the right in the model shown in this page at a sliding velocity \( U \). A liquid film of ethylene glycol is assumed to be present between the DN gel test piece and the mating material (SUJ2), whereby the thickness of the liquid film at the inlet side is taken as \( h_1 \) and that at the outlet side as \( h_2 \) (\( h_1 > h_2 > 0 \)).

The pressure \( P \) working on the DN gel test piece in this model (the so-called bearing load capacity) can be expressed by expression ① derived from the Reynolds equation. Where, \( \eta \) is the viscosity of ethylene glycol, and \( a \) is a coefficient determined by the bearing surface length \( B \) and \( h_1 \) and \( h_2 \):

\[
P = \frac{\eta U a}{h_2^2} \tag{1}
\]

![Fig. 8 Hydrodynamic lubrication model of DN gel](image)

At the same time, the friction force \( F \) working on the DN gel test piece can be defined by expression ②, where \( A \) is the area of the sliding surface of the DN gel test piece, \( \beta \) is a coefficient determined by the bearing surface length \( B \), \( h_1 \), and \( h_2 \):

\[
F = \frac{\eta U A}{h_2^2} \beta \tag{2}
\]

When the test load is \( W \), the contact surface pressure \( P = W/A \), and kinetic friction coefficient, \( \mu_k = FW \), then from expressions ① and ②, the kinetic friction coefficient \( \mu_k \) can be expressed by expression ③, where \( \gamma \) is a coefficient determined by \( h_1 \) and \( h_2 \):

\[
\mu_k = \left( \frac{\eta U}{P} \right)^{1/2} \gamma \tag{3}
\]
Consequently, from expression (3), it appears that the kinetic friction coefficient $\mu_k$ in the case of fluid lubrication state is proportional to $\frac{\eta U}{P}$ to the 1/2 power.

The results of Figs. 6 and 7 have been reorganized, taking $\frac{\eta U}{P}$ along the X axis. The result of reorganization is plotted in Fig. 9. Incidentally, for comparison purpose, this diagram also includes the plotting of friction coefficient values obtained from a series of measurements that used polyether etherketone (PEEK) resin (a super-engineering plastic) in place of the DN gel.

In this diagram, two broken lines having inclination of the 1/2 power are indicated. In a region where $hU/P$ is greater than about $1 \times 10^{-9}$ [m], the inclination of $\frac{\eta U}{P}$ is nearly proportional to the “1/2 power” broken lines: in this situation, the test pieces are under a lubrication state equivalent to fluid lubrication. Incidentally, a fluid lubrication state never occurs with a conventional material under such load-velocity conditions: this finding is supported by the friction behavior of PEEK resin test pieces that were subjected to test conditions identical to those for our DN gel test pieces. In summary, we believe that our DN gel is a very unique material that is capable of low-friction sliding in a fluid lubrication state, beginning at an unconventionally low $\frac{\eta U}{P}$ region, whereby this gel material boasts a high degree of physical strength which conventional gel materials lack.

6. Conclusion

We have proven that our DN gel, having ethylene glycol solvent, exhibits extremely low friction coefficients under various conditions when used in conjunction with a steel species which is commonly used as a mating material for a sliding member.

In addition, it has been suggested that, compared with other polymer materials, the DN gel enables easy achievement of fluid lubrication state by controlling contact surface pressure, sliding velocity and solvent viscosity.

We wish to utilize this technology to help further reduce friction loss occurring on machine components.

References