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授　与　学　位	博士（工学）
学位授与年月日	平成２７年３月２５日
学位授与の根拠法規	学位規則第４条第１項
研究科，専攻の名称	東北大学大学院工学研究科（博士課程）ナノメカニクス専攻
学位論文題目	Design of Sealing Surface of Mechanical Seal for Ventricular Assist Device （補助人工心臓用メカニカルシールの接触面設計に関する研究）
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論 文 内 容 要 旨

Ventricular assist device (VAD) assists role of human heart as a blood pump by rotating impeller at servo-controlled certain rotating frequency. Mechanical seal, which consists of seat ring (Silicon Carbide) and seal ring (Carbon), is installed in order to seal introduced blood against internally-circulated purified water. Low leakage of sealed fluid, low and stable friction and high load capacity are required for mechanical seal of VAD. It was reported that introduced blood causes unstable driving property and high electric energy consumption in a few cases. Therefore, in order to realize more stable driving property and lower electric energy consumption for the sake of next-generation VAD, which improves patient's quality of life more and more, friction force generated at sealing surface of mechanical seal in VAD must be stabilized and decreased. However, neither fundamental friction property of mechanical seal in blood had not been investigated nor friction mechanism of mechanical seal had not been clarified yet. In addition, design guideline for sealing surface does not exist due to lack of understood friction mechanism. This thesis aims to clarify friction property and sealing property of mechanical seal at similar condition of VAD, and to clarify friction mechanism of mechanical seal in blood. Finally, design concept of sealing surface of mechanical seal for next-generation VAD will be proposed.

In chapter 1, background of the thesis are described and aims of the thesis are stated.

In chapter 2, experimental apparatus to clarify friction property and sealing property were designed based on actual structure of conventional VAD. Schematic of the test apparatus is shown in Fig. 1 and typical friction property of mechanical seal in

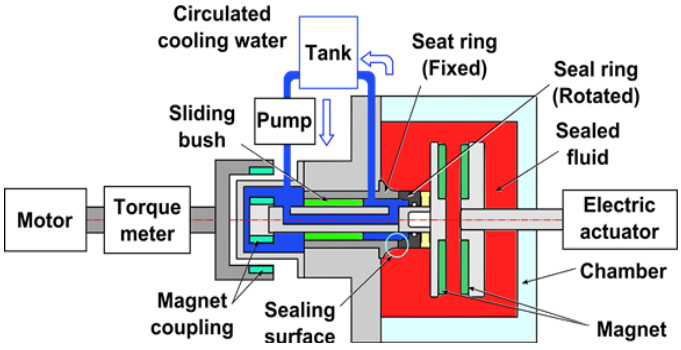


Fig. 1　Friction apparatus for mechanical seal.

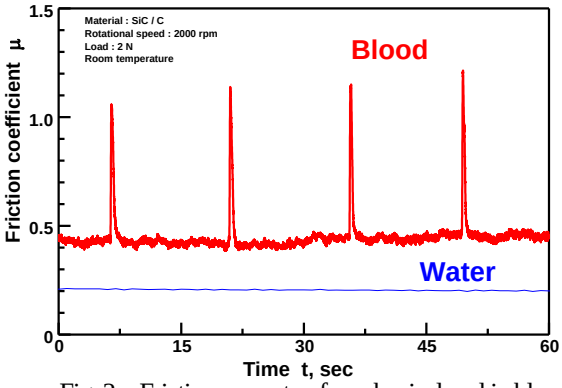
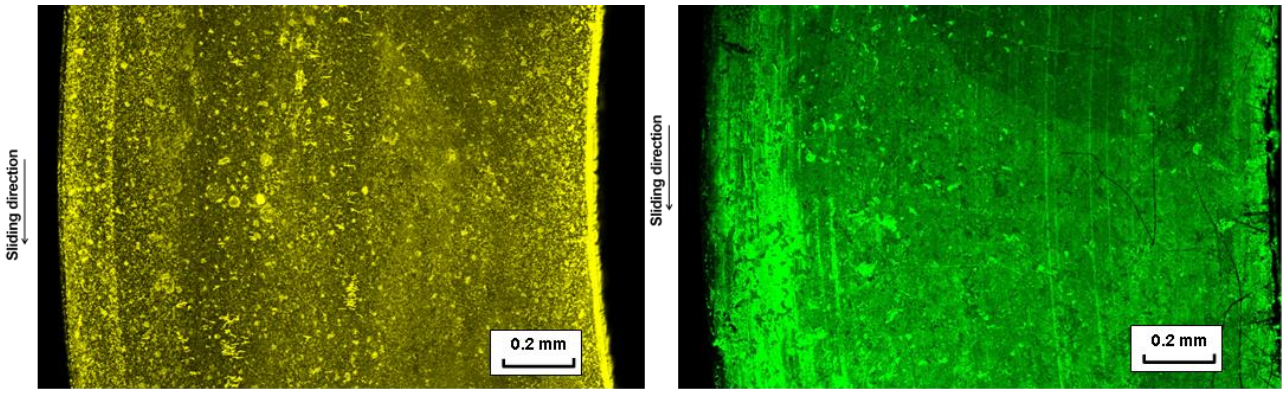


Fig. 2　Friction property of mechanical seal in blood.

blood is shown in Fig. 2. It was clarified that friction of mechanical seal shows high and unstable behavior in blood while low and stable friction coefficient was observed in water. Average friction coefficient and standard deviation in blood were 0.71 and 0.15 while those of in water were 0.17 and less than 0.01, respectively. In addition, periodic frictional peak appears from baseline of friction coefficient in blood. As examined friction property of mechanical seal shows similar tendency to monitored behavior of servo-controlled current in VAD, it is estimated that current is increased by servo-controller to maintain certain rotating frequency against frictional torque generated at sealing surface of mechanical seal. It was also found that whole sealing surface is covered by blood-derived protein film after friction test in blood by means of immunofluorescence staining method, as shown in Fig. 3. Furthermore, it was clarified that existence of formed protein film contributes generation of periodic frictional peak and high baseline of friction coefficient. On the other hand, it was found that protein film decreases leakage of sealed fluid. Therefore, in order to satisfy requirements for mechanical seal, formation of protein film must be controlled.

In chapter 3, in order to clarify formation mechanism of protein film on sealing surfaces of mechanical seal, static protein adsorption on substrate is investigated by means of quartz crystal microbalance (QCM) while protein adsorption with friction and effect of plasma protein on friction property are investigated by pin/disk tribometer. As a result of experiments investigated by QCM, it was found that protein begins to adsorb on sputtered SiC and C statically as soon as substrates are exposed to plasma and adsorbed mass increases by biological protein aggregation. Further, faster adsorption speed was detected on C than on SiC. On the other hand, as a result by pin/disk tribometer, protein film was formed from concaves of disk in friction region of disk and thickness of the protein film was found to be 200 nm approximately while less protein adsorption was detected in non-frictional region. It was also clarified that denatured and aggregated protein, whose secondary structure shows β-sheet, consists of protein film. These results indicate that protein molecules is denatured by high temperature and high pressure caused around contact point between sealing surface and aggregation of protein molecules occur.

In order to pursue formation process of protein film, artificial concaves were fabricated by means of pico-second laser processing and effect of concaves on formation process of protein film was investigated. It was clarified that denatured and aggregated protein molecule is caught in front section of concaves along sliding direction of sliding counterpart and accumulation of aggregated protein occurs at every friction cycle. Further, protein film is begun to be formed on plateau region



(a) seat ring (b) seal ring

Fig. 3 Protein film formed on (a) seat ring and (b) seal ring.

as concaves are completely filled by aggregated protein. Relationship between friction coefficient in plasma and coverage of protein film is shown in Fig. 4. It is obvious that coverage of protein film slightly increases until 80 cycles and tendency of friction coefficient corresponds to the coverage. This relationship indicates that higher friction is generated by protein film formed on plateau region while lower friction force was generated during aggregated protein is kept in concaves.

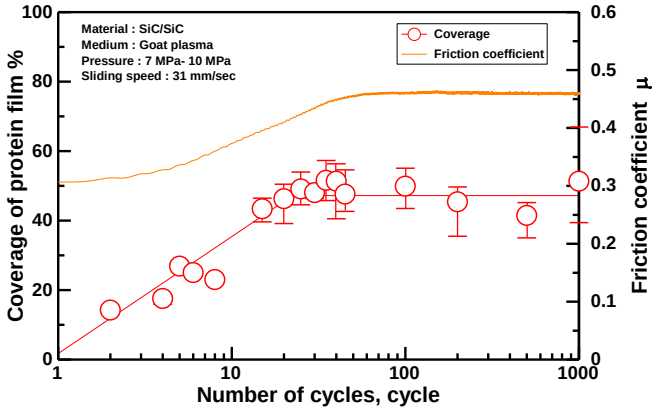


Fig. 4 Relationship between friction coefficient and coverage of protein film.

In chapter 4, in order to clarify generation of mechanism of periodic frictional peak, the effect of concaves on friction property of mechanical seal in blood was examined and material combinations of polished SiC / polished SiC and Initial SiC / Polished SiC were employed. As a result, friction coefficient of polished mechanical seal did not show apparent periodic frictional peak while mechanical seal which consists of initial SiC / polished SiC showed periodic frictional peak in blood. This comparison indicates that concaves on sealing surfaces contribute generation of periodic frictional peak. Rotational frequency of mechanical seal was utilized and it was found that dominant periodicity of frictional peak is independent from rotational frequency but depends on temperature of circulated water and on concentration of sealed fluid. Further, introduction of anionic surfactant, which suppresses hydrophobic interaction between protein molecules by terminating hydrophobic group of protein molecule, demolished generation of periodic frictional peak gradually. These results suggest that periodic frictional peak is generated by biological aggregation of protein molecules in concaves.

Based on formation mechanism of protein film on substrate friction mechanism of mechanical seal in blood was proposed as shown schematic in Fig. 5. Total friction force generated at sealing surface is defined as summation of friction force generated at contact area between plateau region of seat ring and protrusion of seal ring and of adhesion force between aggregated protein molecules from concaves on seat ring against sliding counterpart. Furthermore, generation mechanism of periodic frictional peak is proposed as repetition of aggregation of denatured protein in concaves, adhesion against sliding counterpart and delamination of aggregated protein by friction force. This mechanism suggests that it is effective to prevent protein adsorption on sealing surface in order to achieve low and stable friction of mechanical seal in blood by creation of hydrophilic surface, which suppresses static protein adsorption caused by hydrophobic interaction.

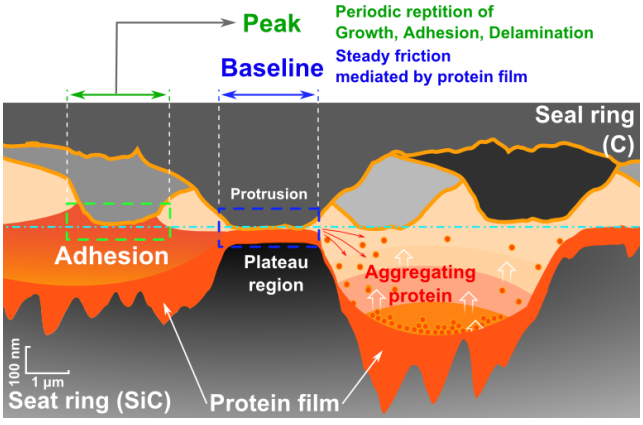


Fig. 5 Friction mechanism of mechanical seal in blood.

In chapter 5, material combination of mechanical seal was replaced from SiC/C to polished SiC/polished SiC and pre-sliding in water was carried out to achieve hydrophilic

sealing surface with an aim to prevent protein adsorption based on previous investigation. Load, rotating frequency and duration during pre-sliding were 2 N, 2000 rpm and 24 hours, respectively. Effect of pre-sliding on friction coefficient is shown in Fig. 6. Average friction coefficient and standard deviation of friction coefficient were decreased to 0.02 and less than 0.1, respectively. Furthermore, formation of protein film was suppressed though smoothed sealing surface showed sudden increase of friction force at 13 N, compared to lower load than that of original material combination of mechanical seal, as shown in Fig. 7. For the sake of actual use of mechanical seal in VAD, further higher load carrying capacity in blood must be ensured to prevent sudden increase of friction force against impact load.

In chapter 6, small concaves, which approximate diameter and depth are 5 μm and 500 nm, are fabricated on sealing surface of smoothened SiC by means of wet blast treatment in order to ensure higher load carrying capacity and diamond-like carbon (DLC), which shows bio-inertness and suppresses static protein adsorption, was coated. Further, formation of protein film on plateau region is expected to achieve lower leakage late of sealed fluid. Effectiveness of DLC on protein adsorption with/without friction were verified by immersing test and by pin/disk tribometer, respectively, and it was proved that DLC suppresses protein adsorption on plateau region and in concaves on sealing surface of mechanical seal. As a result of creation of DLC-coated small concaves on sealing surface, generation of periodic frictional peak was suppressed and stable friction was achieved in blood as shown in Fig. 6. Average friction coefficient and standard deviation of friction coefficient showed 0.17 and 0.01, respectively. Further, load carrying capacity in blood was extended to 16 N as shown in Fig. 7. In addition, leakage of sealed fluid was decreased to 12 nl/min while original mechanical seal (SiC/C) showed 48 nl/min.

In chapter 7, general conclusions of the thesis are summarized and design concepts for sealing surface of mechanical seal for next-generation VAD is proposed.

In this thesis, fundamental friction property of mechanical seal for VAD was clarified and friction mechanism of mechanical seal in blood were clarified. Based on clarified friction mechanism, low and stable friction, higher load carrying capacity and lower leakage were realized by controlling protein adsorption on sealing surface of mechanical seal. Proposed design concepts enables development of mechanical seal which shows ideal properties for next-generation VAD.

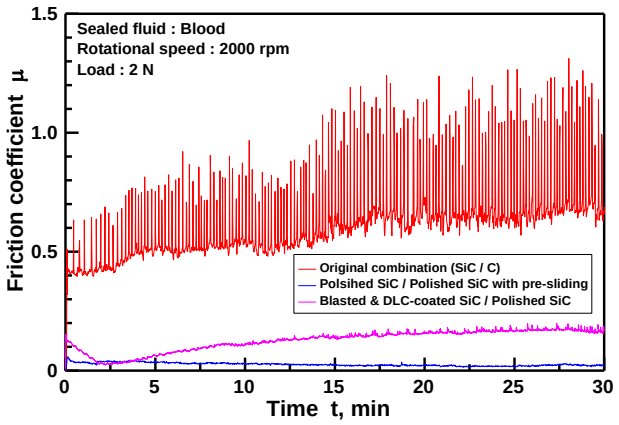


Fig. 6 Effect of creation of smooth and hydrophilic surface on friction coefficient in blood.

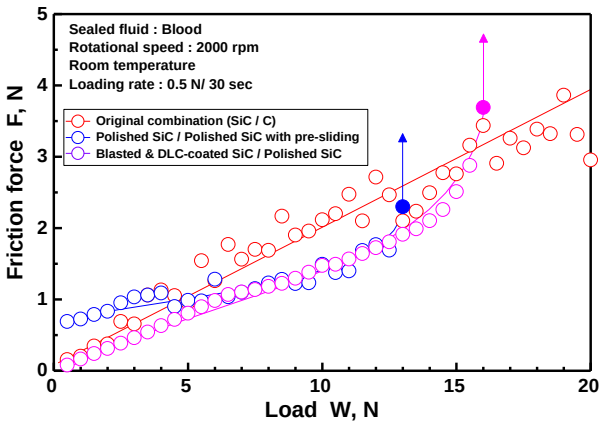


Fig. 7 Effect of creation of smooth and hydrophilic surface on load carrying capacity in blood.

論文審査結果の要旨

血液用メカニカルシールの摩擦は，体内埋込型補助人工心臓における信頼性と低消費電力の鍵を握るものであり，より低く安定した摩擦の実現は，低消費電力で高信頼・高性能が求められる次世代補助人工心臓実現の重要な技術課題である。しかしながら，実機において発生する摩擦特性に関する情報はほとんど存在せず，その低摩擦化のための技術開発は現場での経験によるところが多い。

このような背景のもとに本論文は，補助人工心臓用メカニカルシールの摩擦を再現できる摩擦試験機を開発し，摩擦面に形成される血液由来のたんぱく膜に起因した摩擦の発生機構およびメカニカルシール接触面でのたんぱく膜の形成機構を実験的に明らかにした。さらにたんぱく膜形成の抑制および制御の観点から低く安定した摩擦が要求される次世代の補助人工心臓用メカニカルシール接触面のための設計指針を提案した。本論文は，これらの研究成果をまとめたものであり，全編7章からなる。

第1章は緒論であり，本研究の背景，目的および構成を述べている。

第2章では，補助人工心臓用メカニカルシールの摩擦・密封特性を評価することが可能な摩擦試験機を設計試作し，摩擦面に形成される血液由来のたんぱく質の膜が支配するメカニカルシールの摩擦・密封特性を明らかにしている。これは，補助人工心臓用メカニカルシールの接触面設計をする上において必要不可欠な有益な知見である。

第3章では，メカニカルシールの接触面をモデル化した2つの実験により，非摩擦部および摩擦部に形成されるたんぱく膜の形成機構およびたんぱく膜が血液用メカニカルシールにおいて発生する摩擦に及ぼす影響を実験的に明らかにしている。これらは，安定した低摩擦を実現するメカニカルシール設計のために重要な知見である。

第4章では，血液用メカニカルシールにおいて発生する周期的摩擦の増減現象が，摩擦表面に存在する数十マイクロメートルオーダの凹部に起因することを世界で初めて明らかにしている。これは，安定した低摩擦を実現するメカニカルシールを設計するために有効かつ重要な知見である。

第5章では，第3，4章において得られた知見に基づき，血液用メカニカルシールの摩擦を支配する摩擦面のたんぱく膜の抑制を目的とした材料選定とその表面の幾何学的形状および表面自由エネルギーの制御により，実機より一桁低い摩擦が発生することを実証している。これは，血液用メカニカルシールの接触面設計のための重要な成果である。

第6章では，密封特性および耐荷重性を考慮した血液用メカニカルシールのために，摩擦面に形成されるたんぱく膜を任意に制御することを目的とした表面の幾何学的形状と表面自由エネルギーの制御により，低い摩擦，高い密封特性および十分な耐荷重性を同時に満足させ得ることを実証している。これは，次世代の補助人工心臓用メカニカルシールの接触面設計のための重要な成果である。

第7章は結論である。

以上要するに本論文は，補助人工心臓用メカニカルシールにおける摩擦機構を明らかにするために，実機の現象を再現できる摩擦試験機を開発し，摩擦面に形成される血液由来のたんぱく膜に起因した摩擦機構および摩擦下でのたんぱく膜の形成機構を明らかにするとともに，たんぱく膜形成の制御の観点から低く安定した摩擦が要求される次世代の補助人工心臓用メカニカルシール接触面のための設計指針を提案したものであり，ナノメカニクスおよび機械工学の発展に寄与するところが少なくない。

よって，本論文は博士(工学)の学位論文として合格と認める。