Tough and conductive polymer hydrogel based on double network for photo-curing 3D printing

Xueyuan Ding, Runping Jia, Zuzhong Gan, Yong Du, Dayang Wang and Xiaowei Xu

Abstract
Conductive hydrogels (CHs) have attracted significant attention in wearable equipment and soft sensors due to their high flexibility and conductivity. However, CHs with high-strength and free-structure still need to be further explored. Herein, 3D printing high-strength conductive polymer hydrogels (CPHs) based on a double network was prepared. Firstly, PHEA-PSS hydrogels were prepared by copolymerizing 2-Hydroxyethyl acrylate (HEA) with 4-Vinylbenzenesulfonic acid (SSS) using a photo-curing 3D printer. Then 3, 4-Ethylenedioxythiophene (EDOT) was in situ polymerized in the network of PHEA-PSS to obtain the PHEA-PSS/PEDOT hydrogels. It can not only satisfy the printing of complex spatial structures, but also has high mechanical and electrical properties. When the content of EDOT is 12 wt%, the tensile strength of the PHEA-PSS/PEDOT hydrogels is close to 8 MPa, the electrical conductivity reach to 1.2 S cm$^{-1}$ and the elasticity remain unchanged. Due to the presence of hydrogen and coordination bonds, CPHs have certain self-heal ability. In addition, the resistance of the hydrogel is sensitive to the changes of external pressure. The results show that CPHs can be used as a 3D printing material for flexible sensors.

1. Introduction

Hydrogels, a polymeric material, whose chains have abundant hydrophilic groups for retaining a significant fraction of water, have been widely used in various fields such as soft robotics and regenerative medicine. The applications of traditional hydrogels are often severely limited by their low mechanical [1–4]. Fortunately, With the deepening of research, highly stretchable hydrogels [5], conductive hydrogels [6], 3D printable hydrogels [7] and self-healing hydrogels [8] are emerging continuously. Specifically, with the growing demand of biomaterials and wearable equipment, electrically conductive hydrogels (CHs) with electrical conductivity and flexible mechanical properties have attracted great interest [9]. CHs have many excellent properties, such as strong adhesion, high porosity, good swelling and biocompatibility, which can transform external stimuli into recorded electrical signals [10–12]. Therefore, CHs have been widely used in the preparation of human motion sensor. Simultaneously, CHs, as a wound dressing, can promote the regeneration of infected skin with the help of the human body’s endogenous bioelectric systems [13]. With the development of 3D printing, CHs having a specific spatial structure become possible. Currently, printable CHs are mostly ionic conductive with a free-moving ion [14]. Due to the simple preparation process and large deformation, it is very suitable for 3D printing.

However, ionic conductivity is sensitive to the change in gel moisture [15]. As the skeleton, the flexible chain has a large deformation, but the single network structure cannot withstand large stress, resulting in a low mechanical strength. Thus, the printed hydrogel is broken easily. It is reported that the combination of conductive polymer (polypyrrole (PPy) [16], polystyrene (PTh) [17] and polyaniline (PANI) [18]) and hydrogel can form interpenetrating polymer network (IPN) or double network (DN) structure, which can improve the mechanical properties of hydrogel [19, 20]. The conductive polymer not only provided electrical
properties but also act as a stiff chain to improve strength. The hydrogel acted as a flexible chain to disperse the stress. The conductive polymer hydrogels (CPHs) are usually synthesized via a two-step polymerization method. After the first network was polymerized, it was immersed and swelled in the monomer of the second network. The monomer of the second network entered into the first network and polymerized to form IPN structure [21]. But the molding time (1–2 days) is far from meeting the requirements of 3D printing rapid prototyping. Furthermore, the subsequent processing after polymerization is also difficult due to the insoluble and infusible characteristic [22].

Here, based on the two-step method, the first step in hydrogel synthesis was carried out by 3D printing. Through digital light processing (DLP) photo-curing printing, using trimethyl benzoyl diphenyl phosphine oxide (TPO) as a photo-initiator to initiate free radical polymerization of 2-hydroxyethyl acrylate (HEA) and 4-vinylbenzenesulfonic acid (SSS), the poly (2-hydroxyethyl acrylate-4-vinylbenzenesulfonic acid) (PHEA-PSS) hydrogel with 3, 4-ethylenedioxythiophene (EDOT) monomer was obtained as a flexible chain. Then under the action of oxidant and dopant, poly(3,4-ethylenedioxythiophene) (PEDOT) was obtained by in situ polymerization of EDOT in the hydrogel, which acted as stiff chain. The CPHs poly (2-hydroxyethyl acrylate-4-vinylbenzenesulfonic acid) / poly(3,4-ethylenedioxythiophene) (PHEA-PSS/PEDOT) eventually formed. The characterization (including chemical composition and morphology), printing effect, mechanical properties, conductivity, swelling behavior and pressure responsiveness of hydrogels were systematically studied and discussed. The results show that the processability of the CPHs could be improved by 3D printing. The tensile stress-strain curve and cyclic tensile curve suggested their good mechanical strength and certain repair properties. The hydrogel had a porous structure from PHEA-PSS and high electrical conductivity from PEDOT. In addition, the potential pressure sensor application of these CPHs in human motion monitoring were demonstrated. This study provided inspiration for the research and application about 3D printing of high-strength CPHs in the future.

2. Experimental

2.1. Synthesis of hydrogels
Firstly, PHEA-PSS hydrogel was synthesized from HEA (2-Hydroxyethyl acrylate) and SSS (4-Vinylbenzenesulfonic acid) monomer with the initiation of the TPO (trimethyl benzoyl diphenyl phosphine oxide). Then, the PHEA-PSS hydrogel containing the EDOT (3, 4-Ethylenedioxythiophene) monomer was put into the FeCl₃ and APS (Ammonium persulfate) oxidant solution for in situ polymerization. Then, the PHEA-PSS/PEDOT hydrogels were finally obtained. Specifically, a certain amount of HEA monomer (6.5 g), SSS monomer (1 g) and different amount of EDOT monomer were dissolved in 20 ml of deionized water. The mixture was placed in a flask and stirred for one hour under ultrasound condition. When the liquid became clear, a certain amount of TPO was added. Ultrasonic stirring was then continued for 1 h to obtain the liquid photo-curing hydrogel resin. The hydrogel was printed by the as-prepared resin through DLP 3D printing system. After washing with alcohol and dried in an oven at 60 °C for 1 h, the printed hydrogel was placed into the oxidizer solution to prepare the PHEA-PSS/PEDOT. Characterizations of the PHEA-PSS/PEDOT were carried out after standing for 24 h in constant temperature and humidity environment.

The type of the DLP is XunShi moonRAY. The CAD model required for printing was created by computer. The wavelength of the printer is 405 nm. The printing accuracy of per-layer is 50 μm. The base layer curing time is 40 s and the number of layers is 5. Each layer’s curing time is 25 s and the compensation time is 3 s. As the amount of EDOT increases, the curing time will increase relatively. When the hydrogel was printed, it was post-cured for 30 min in an UV curing chamber (370 nm, 100 W).

2.2. Characterization
FTIR (Thermo Nicolet iN10) was performed to determine the chemical structure of PHEA-PSS hydrogel samples. The polymerization of PEDOT in the hydrogel was tested by XPS (Thermo ESCALAB 250Xi). After the hydrogel was lyophilized, the freeze-dried sample was slit. The cross-sectional morphology of the hydrogel was observed by scanning electron microscopy (FEI Quanta200 FEG) under an electron beam with an accelerating voltage of 7 kV. For the mechanical strength test, the stress-strain curve was obtained using a universal testing machine (CARDANO SUN500). The dimensions of the sample were 20 mm × 4 mm dumbbell with thicknesses of 2 mm, and the displacement rate was 100 mm min⁻¹. The electrical conductivity was measured via the four-probe method (SX1934 (SZ-82)) at room temperature by inserting the four probes into cuboidal samples. To demonstrate the sensing capability of the hydrogel, the current variations in the hydrogel were recorded by electrochemical workstation (CHI 760e) using the amperometric i-t curve. Moreover, the hydrogels can act as conductor to connect electrical circuit and light up a 5 mm diameter round light-emitting diode (LED) using a 3 V battery. The difference in swelling of the hydrogel under different compositions was observed. After
immersing in deionized water for a certain time, the samples were taken out and weighed quickly. The valid data were obtained by taking the average of four samples for each component.

The swelling ratio was according to the following equation:

$$SR = \frac{(W_s - W_d)}{W_d} \times 100\%$$

$W_s$ and $W_d$ were the weights of swollen hydrogel and the nonswollen hydrogel. Each sample was tested four times and then averaged.

3. Results and discussion

3.1. Design strategy and preparation

The schematic diagram of the overall fabrication process of CPHs was shown in figures 1(a) and (b). The DLP printing requires that the printing resin should have a low viscosity before photo-polymerization to ensure that each layer of resin could be quickly and fully replenished. The initial viscosity of the resin prepared by the one-pot reaction was 100 mPa·s. The uneven dispersion of the slightly soluble EDOT can seriously affect the performance of the hydrogel. The ionic bond formed between SSS and EDOT, and the oil-water structure formed by HEA allow the EDOT to be successfully dispersed in aqueous solution [23]. The oil phase also included the photo-initiator TPO. When irradiated by the printer’s UV light, the TPO was cleaved into two free radicals, and the double bond was polymerized to produce PHEA-PSS hydrogel [24]. In addition to increasing the solubility of EDOT, SSS monomer also acted as templates for the polymerization of PEDOT. Then, EDOT was well fixed on the PHEA-PSS chain. The transparent hydrogel gradually turned blue-black color after placing in the oxidizer solution. In contrast, pure PHEA hydrogel was unable to immobilize free EDOT monomer, leading to the polymerization of EDOT in the solution (figure S1 is available online at stacks.iop.org/MRX/7/055304/mmedia). Due to the slow absorption of the oxidant by the hydrogel, it avoided the vigorously polymerization of EDOT between the two phases and uneven internal polymerization. Meanwhile, after entering into the water molecule, the flexible chain was elongated, which was favorable for the linear growth of EDOT to improve the conductivity. When the PEDOT was formed, the hydrogel was taken out and dried. Contact areas between the PEDOT chains increased after the re-curlling of the molecular chains, which created more electrically conductive paths. Finally, through the coordination bond of Fe$^{3+}$, hydroxyl, and sulfonate, the 3D printing CPHs form an interpenetrating double network structure with PHEA-PSS as the flexible chain and PEDOT as the stiff chain.

3.2. FT-IR Spectrum of the PHEA-PSS

The FTIR spectra of HEA, SSS and the printed PHEA-PSS hydrogel were conducted to determine the chemical structure of PHEA-PSS (figure 2(a)). The peak appeared at 1727 cm$^{-1}$ was attributed to C=O stretching vibration of HEA [25]. The absorption peaks of C–O in original alcohols appeared at 1080 cm$^{-1}$, and C–O–C appeared at 1297 cm$^{-1}$ and 1199 cm$^{-1}$. In the characteristic peaks of SSS, the peaks at 1600 and 1500 cm$^{-1}$ correspond to the in-plane vibration of aromatic ring, and the absorption peak at 835 cm$^{-1}$ ascribed to the p-disubstitution of aromatic ring [26]. The peaks at 1185 and 1040 cm$^{-1}$ were attributed to $\text{SO}_3^–$ symmetric stretching [27]. The characteristic peaks of each functional group in HEA and SSS can be found in PHEA-PSS, indicating that the reaction between HEA and SSS is successful. Then, three adjacent peaks at 1297, 1199, and 1185 cm$^{-1}$ eventually formed a broad peak in the range of 1350 $\sim$ 1150 cm$^{-1}$ of PHEA-PSS. The peak at 1637 cm$^{-1}$ was attributed to the C=C stretching caused by terminal acrylate group, and the peak at 1400 cm$^{-1}$ was ascribed to the in-plane vibration of CH$_2$ from $-\text{C}=\text{CH}_2$. Both HEA and SSS had these two peaks, which
disappeared after the reaction [28]. It indicated that the polymerization was complete. In summary, the PHEA-PSS hydrogel was successfully prepared.

3.3. XPS analysis of PHEA-PSS/PEDOT

Since many of the PEDOT infrared peaks are covered by PHEA-PSS, XPS was used to demonstrate the formation of PEDOT in hydrogel. The S 2p core-level XPS spectrum of the sample was shown in figure 2(b). In agreement with previously reported studies [29], two peaks appeared at 168.6 and 164 eV corresponded to the sulfur signals of sulfonate (in PHEA-PSS) and thiophene (in PEDOT). The peak at 168.6 eV can be decomposed into two peaks of S 2p$_{1/2}$ and S 2p$_{3/2}$ with a relative intensity ratio of 2:1 at 168.3 eV and 169.6 eV, respectively [30].

3.4. Microstructure of the hydrogel

To investigate the microstructure, the cross-sectional morphology of the freeze-dried PHEA-PSS and PHEA-PSS/PEDOT (5 wt%) hydrogel samples were observed. The internal structure of the two samples is significantly different, and the microstructure is closely related to mechanical properties, swelling ratio and electrical properties. Before the polymerization of EDOT (figure 2(c)), the PHEA-PSS hydrogel exhibited a fluffy, porous three-dimensional network structure. The pores distributed uniformly with an average size of about 0.2 μm. The structure ensured that the PHEA-PSS possessed high swelling ratio and good flexibility. Under the action of external force, there is enough space for tolerating the volume deformation. When EDOT was in situ polymerized in the hydrogel (figure 2(d)), the original network structure was wrapped by PEDOT, which was formed along the PHEA-PSS chain. The DN structure mentioned above was finally formed. Compared with figure 2(a), some of the pores are blocked, thereby reducing the expansion rate of the hydrogel. However, the mechanical strength was increased due to the compactness of the DN structure and the interconnection between the two networks. Although the pores were decreased, the network structure still existed, except that the diameter of the pores is reduced to 0.5~1 μm. (figure S2).

3.5. Photo-polymerization and fast gelling

As an additive manufacturing technology, DLP based on layer-by-layer solidification is from down to top. Each exposure can cure a layer of resin, greatly improving the printing speed [31]. However, the requirements for curing degree have increased. If one layer is not fully cured, it will cause all subsequent printing failure. The
The curing time of each layer determined the total printing time. In order to meet the need of fast printing, the exposure time should be as short as possible without affecting the molding. In order to obtain the shortest curing time, we recorded the time required for the solution to reach a certain viscosity under the UV light (table 1). When the amount of EDOT is not large, increasing EDOT has little effect on the curing rate. When a certain amount (>20 wt%) of EDOT is exceeded, the polymer chain was too less to form a gel structure. The whole structure was broken and viscous, and has no fixed shape. In the actual situation, the time in the table 1 would be subtracted by 5~10 s to improve the printing rate (EDOT ≤ 10 wt%). The printed samples were shown in the figure S3. The EDOT content of the printed hydrogels was 12 wt%. It was proved that the liquid resin of PHEA-PSS could meet the printing of complex spatial structures. Compared with previous studies on single-layer printing and extrusion printing, the accuracy and efficiency of PHEA-PSS hydrogel printed by DLP have been greatly improved [32, 33].

### 3.6. Mechanical properties

PHEA-PSS/PEDOT hydrogels exhibited outstanding mechanical properties. The PHEA-PSS hydrogel itself was highly flexible. It could be stretched to four times its initial length. But the tensile strength was low and easy to be broken. The mechanical properties of the hydrogel were improved after the introduction of PEDOT, which was in situ polymerized in oxidant solution. The sample was approximately 8 MPa at EDOT content of 12 wt%. Figure 3(a) was the typical tensile stress-strain curve of the hydrogel at different amount of EDOT. As the EDOT content increases, the tensile strength of the sample increased from 0.72 to 7.8 MPa, while the fracture strain decreased obviously. It was because that PEDOT as a stiff chain could sustain the stress well [34]. The mechanical strength of the CPHs increased with the increasing of stiff chain content. Due to the π-electron conjugated structure, PEDOT is so difficult to rotate. Therefore, when PEDOT is added to the hydrogel, the deformation of the hydrogel is reduced. But the PHEA-PSS as a flexible chain could well distribute the stress [35]. Compared with the previous pure PEDOT material [36], the tensile strain of the PHEA-PSS/PEDOT hydrogel is up to 173%, implying its high flexibility.

With the increase of EDOT, the content of oxidant (FeCl₃) increases. As a hybrid network, the coordinative bond formed between iron ions and PHEA-PSS further enhanced the mechanical strength [37]. In the control experiment (figure 3(b)), the amount of EDOT was unchanged, while FeCl₃ was increased, and the stress-strain curve showed the same trend: tensile strength increased and strain at break decreased. The coordinative bond could serve as physical cross-linked points and enhance the entanglement between molecular chains. When the chain suffered the stress, it was difficult to be stretched and spread out. The hydrogel samples showed certain rigidity under low deformation condition. When the stress was applied continuously, the coordinative bond broke firstly. The previously curled chains were expanded and the deformation enlarged. The stress-strain curve shows a significant turning point. Iron ions also lead to the encapsulation of some hydrophilic groups on the molecular chain, resulting in a decrease of swelling ratio.

The cyclic tensile test was carried out on CPHs with PEDOT content of 5 wt%, and the PHEA-PSS/PEDOT hydrogel showed obvious hysteresis phenomenon (figure 3(c)). The areas between the loading and unloading curves of the samples showed the energy dissipation per unit volume. The results show that hydrogel can dissipate energy effectively and maintain space network structure. At the same time, due to the stress distribution caused by the breakage of coordination bond, the main chain structure of hydrogel was still integrated. After removing the stress, the coordination bond can be quickly recovered. Therefore, compared with pure PHEA-PSS hydrogel, the hydrogel containing PEDOT demonstrated smaller permanent deformation [5, 38].

There were a large number of hydroxyl groups in the PHEA-PSS chains. The coordinative bonds and hydrogen bonds formed between the chains enabled the CPHs good repair properties [39, 40]. The two damaged halves of the CPHs were repaired soon. The sample can be reconnected after 24 h under a certain pressure. The reconnected hydrogel could be subjected to a secondary stretching (figure 3(d)). Although the mechanical

| Table 1. Photo-curing time of the resin at different EDOT contents. |
|------------------|------------------|
| EDOT content (wt%) | T(s) (viscosity > 100 Pa-s) |
| 0                | 25~30            |
| 2                | 30~40            |
| 5                | 30~40            |
| 8                | 35~45            |
| 12               | 75~90            |
| 15               | 120~200          |
| Above 20         | ∞                |
properties of the material decreased under the secondary stretching, the hydrogel still exhibited certain repair properties.

### 3.7. Electrical conductivity

The ratio of oxidant (APS), dopant (FeCl₃) and monomer (EDOT) plays a key role in the polymerization and electrical properties of the electrically conductive polymer. In order to find the optimal ratio of FeCl₃, APS, and EDOT, the conductivity of different PHEA–PSS/PEDOT hydrogels obtained at different ratios was tested under the condition of fixed EDOT content of 12 wt% (figure 4(a)). Ions from the FeCl₃ and APS were adsorbed inside the hydrogel. It was difficult to remove them completely. The electrical conductivity of dry gel was ascribed to π-electron conjugated polymer of PEDOT. For ruling out the influence of ions, the evaluation of PEDOT was based on the conductivity of the dry sample. Compared to only one oxidizer APS, Fe³⁺ as electron acceptor took electrons from π-electron conjugated polymer of PEDOT to form carrier in the chains. When the FeCl₃ content was low, few carriers led to a poor electrical conductivity. When FeCl₃ was excessive, too much oxidizing points led to a decrease in the molecular weight of PEDOT, which weakened the contact of electrically conductive phases. At the same time, peroxidation also destroyed the conjugated structure of PEDOT, which caused the electrical conductivity decline. When the ratio of EDOT, APS and FeCl₃ was 1:1:1, the electrical conductivity of dry gel is the largest (0.5 S cm⁻¹). Combined with the tensile data, the decrease in electrical conductivity in the dry state indicated that the stiff chain does not increase under the excessive FeCl₃. However, the tensile strength was increased. These results supported that the coordinative bonds reinforced the hydrogel. Meanwhile, the watery sample was tested after its mass reached to a constant value at room temperature and 60% relatively humidity. Due to the existence of free water, the increase of ion concentration leads to the significant increase of electrical conductivity. The hydrogel had a large deformation under this water swelling state. Using a four-point probe, the electrical conductivity was greatly affected by the pressure. The error was expanded by
nearly two orders of magnitude compared to dry gel. In the absence of free water, the movement of the ions is impeded, and in the dry state, the electrical conductivity is reduced. Therefore, the electrical conductivity of PHEA-PSS/PEDOT comes from the synergistic effect of ionic conductivity and electrical conductivity.

After using EDOT: FeCl₃: APS = 1:1:1 as a fixed ratio, the electrical conductivity of PHEA-PSS/PEDOT with different EDOT concentration in wet state (60% relatively humidity, RT) was tested (figure 4(b)). At low concentration, an increase of EDOT had little effect on conductivity because the electrically conductive chains are far apart. When the content of EDOT reached to 8 wt%, the electrical conductivity was significantly improved due to the formation of electrically conductive paths. When the EDOT content exceeded 15 wt%, the free EDOT would escape from the hydrogel interior and polymerized in the oxidizer solution. This is because that the amount of EDOT immobilized on PHEA-PSS chains was limited, resulting in constant electrical conductivity. Although the electrical conductivity of 1.2 S/cm for PHEA-PSS/PEDOT was much lower than that of pure PEDOT (780 S cm⁻¹) [43], it had been improved compared with ionic conductive hydrogel (0.104 S cm⁻¹ [15], 0.029 S cm⁻¹ [14]) or other CPHs (PPy: 0.003 S cm⁻¹ [16], PEDOT: 0.01 S cm⁻¹ [44]). The hydrogel was printed into the shape of batman (figure 4(c)), which could illuminate the LED. In summary, the PHEA-PSS/PEDOT hydrogel is a synergistic structure of electronic and ionic conduction.

3.8. Swelling performance
The swelling performance of the printing hydrogel showed reduced swell ability with increasing the content of EDOT (figure 4(d)). The swelling rate decreases from high to low until it reaches a constant rate. Combined with the SEM image and mechanical properties mentioned above, on one hand, the in situ polymerization of PEDOT on PHEA-PSS hydrogel blocked some small pores. On the other hand, the coordination bond formed by iron ions encapsulated a lot of hydrophilic groups in the chains.

![Figure 4. (a) Electrical Conductivity of hydrogel in swollen state and dry state with different oxidant ratios respectively with EDOT content of 12 w%. (b) Conductivity at different concentrations of EDOT with a fixed ratio of FeCl₃: APS = 1:1. (c) Photograph of printing hydrogel lighting the LED. (d) Swelling performance of hydrogel showing a decline with the addition of EDOT.](image-url)
3.9. Sensor application
The application of the PHEA-PSS/PEDOT hydrogel pressure sensors for monitoring human activity is investigated. The hydrogel was tied to the finger in nitrile gloves and the two ends of hydrogel were attached to the working electrode and counter electrode of electrochemical workstation. The $\Delta I/I$ value was recorded by i-t curve to detect the sensitivity of the sensor (figure 5(a)). Bending the finger rapidly within 10 s to 25 s, the hydrogel could convert fast changeful mechanical signal into electrical signal. Different pressures could also be reflected. In 40–60 seconds, the fingers bend a certain angle and stay for a period of time. No fracture was observed even after multiple folds, verifying the excellent durability and stretchability of the printed hydrogel. When a 100 g weight was loaded on the sample, a platform of $\Delta I/I$ (20%) was found after a short delay deformation (figure 5(b)).

4. Conclusion
In summary, we successfully prepared a conductive hydrogel resin by one-pot reaction and used it for photo-curing 3D printing. The resin not only satisfied fast printing requirements, but also produced flexible materials with various shape. Complex structures such as hollow stereoscopic shapes could also be printed to improve the printing effect. Moreover, the printed PHEA-PSS hydrogel was act as a template in the synthesis of PEDOT, which could improve the conductivity of the hydrogel. The PHEA-PSS/PEDOT hydrogel prepared by in situ polymerization had DN structure with physical cross-linked points. It not only inherited the high deformation of hydrogel (173% of tensile strain, 5 wt% of EDOT), but also maintained the merits of self-healing and conductivity of PEDOT (1.2 S cm$^{-1}$, 12 wt% of EDOT). At the same time, the mechanical strength of PEDOT (8 MPa, 12 wt% EDOT) was increased. Further, the role of iron ions in the system as oxidant, dopant, and ion crosslinking point was deeply discussed. It was demonstrated that hydrogels could be used as pressure sensors to monitor human activity. This study provided a new idea for 3D printing conductive hydrogel, which could be used in stretchable electrical devices and soft robotics applications.

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ORCID iDs

Runping Jia @ https://orcid.org/0000-0002-6199-8607

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