Electronic Supplementary Material (ESI) for Soft Matter.

Electronic Supplementary Information

A self-oscillating gel system with complex dynamic behavior based on a time delay between the oscillations

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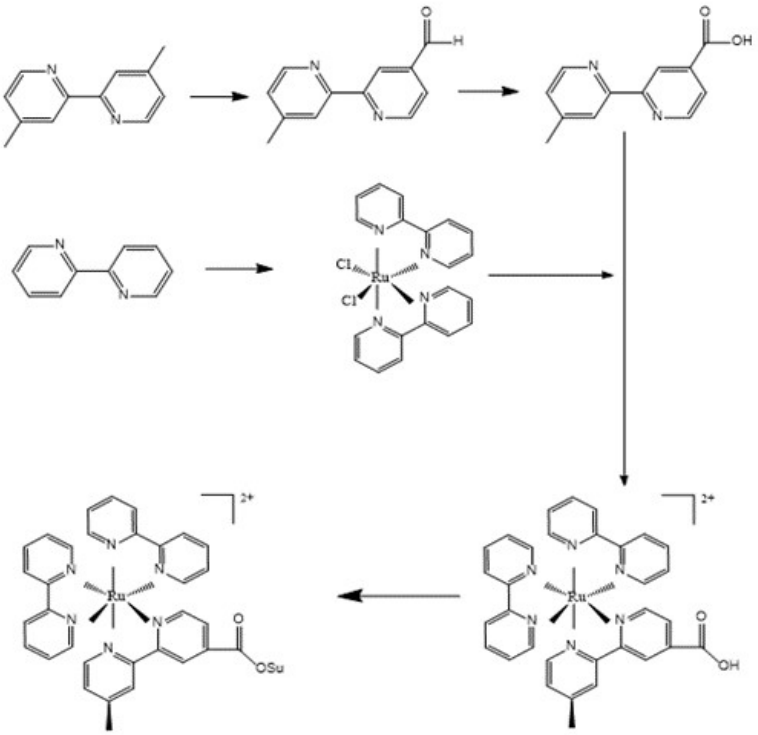
1.Materials

N-Isopropylacrylamide (NIPAAm), N-3-(Aminopropyl) methacrylamide (NAPMAm), Ruthenium (III) chloride hydrate ($RuCl_3 \cdot 3H_2O$, 99 %), 4,4'-dimethyl-2,2'-dipyridyl, 2,2'-bipyridine, lithium chloride (LiCl), 1,4-Dioxane, Sodium Pyrosulfite (NaS_2O_5), N,N'-Dicyclohexylcarbodiimide (DCC), 2,5-Pyrrolidinedione, tetramethylenediamine (TEMED), ammonium persulfate (APS), sodium carbonate (Na_2CO_3), sodium bromate ($NaBrO_3$) and malonic acid (MA) were purchased from Aladdin Bio-Chem Technology Co., Ltd (Shanghai, China). Selenium dioxide (SeO_2), N, N'-Methylenebisacrylamide (MBAAm) was purchased from Macklin Biochemical Co., Ltd (Shanghai, China). Dimethyl sulfoxide (DMSO), HNO_3 aqueous solution was purchased from Kelong Chemicals (Chengdu, China). Ethyl acetate ($C_4H_8O_2$), petroleum ether, dichloromethane (CH_2Cl_2), ethanol (C_2H_6O), NaOH, hydrochloric acid (HCl), acetic Acid (CH_3COOH), Acetonitrile (CH_3CN), Dimethyl formamide (DMF), Iso-Propyl alcohol and acetone (CH_3COCH_3) were purchased from Guanghua Sci-Tech Co., Ltd (Guangdong, China). Ammonium hexafluorophosphate ($NH_4[PF_6]$) and hexafluoro phosphoric acid (HPF_6 in water, 60%) were purchased from Meryer Sci-Tech Co., Ltd (Shanghai, China).

2.Synthesis of Bis(2,2'-bipyridine) (1-(4'-methyl-2,2'-bipyridine-4-carbonyloxy)-2,5-pyrrolidinedione) ruthenium (II) bis(hexafluorophosphate) (Abbreviated as $Ru(bpy)_3$–NHS)



It was synthesized according to previous report. [1]

3. Preparation of poly (NIPAAm-co-NAPMAm-co-$Ru(bpy)_3$NAPMAm) gel

NIPAAm, NAPMAm, MBAAm, and TEMED were dissolved in distilled water. (Table.1) After the pre-gel solution was cooled in iced water under a nitrogen

atmosphere for 30 min, the initiator APS was added. Then the pre-gel solution was injected into the mould (Fig.1a) and radical copolymerization was conducted at 4℃. After gelation, the prepared gels were thoroughly washed with distilled water to remove unreacted monomers for two days. The gels were immersed in DMSO to exchange solvent. After that, the gels were immersed in DMSO solution of 70 mM $Ru(bpy)_3$–NHS containing triethylamine for 24 h in the mould (Fig.1b) to conjugate $Ru(bpy)_3$ to amino group of the gels. Then, the gels were thoroughly washed with DMSO to remove unreacted reagents. Prepared gels were preserved in distilled water.

Table1. Structural profiles of the SOGs

	G1	G2	G3	G4	G5	G6
NIPAAm/mg	113	113	113	113	113	113
NAPMAm/mg	17	17	17	17	17	17
MBAAm/mg	1.70	1.36	1.02	0.68	2.04	2.38
TEMED/mg	3.1	3.1	3.1	3.1	3.1	3.1
APS/mg	1.25	1.25	1.25	1.25	1.25	1.25
H_2O/ml	1	1	1	1	1	1

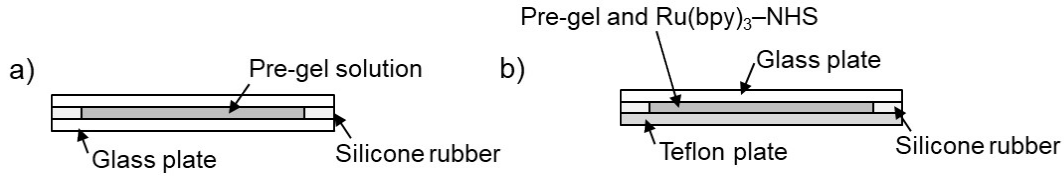


Fig.1 Preparation of the (NIPAAm-co-NAPMAm-co-$Ru(bpy)_3$NAPMAm) gel undergoing anisotropic contraction.

References

[1] B. M. Peek, G. T. Ross, S. W. Edwards, G. J. Meyer, T. J. Meyer and B. W. Erickson, *Int. J. Protein Res.*, 1991, **38**, 114