

Supporting Information

Natural Product Gelators and General Method for Obtaining Them from Organisms

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Table of Contents

1. Experimental Section.....	3
a. Materials	3
b. General Information	3
c. Extraction and Isolation	4
d. Gelation Tests	5
e. Rheology Measurements	5
f. Identification of Compounds 1-7.....	5
2. Supporting Tables.....	8
Table S1. Information and Gelation Results for 63 Organism Extracts	8
Table S2. Gelation Results for Compounds 1-6 in 48 Solvents and Morphology for Natural Product Gels	11
3. Supporting Figures	14
Figure S1. SEM Images of 16 Extracts Gels.....	15
Figure S2. Structure of the compound 7	15
Figure S3. Spectra of Compound 1-7	28
REFERENCES.....	28

1. Experimental Section

a. Materials

Reagents were purchased from Sigma Aldrich, Fluka, and Alfa Aesar and used as received, without further purification, unless otherwise stated. All solvents used were of analytical grade.

Poria cocos (Schw.) Wolf, *Euchresta japonica*, *Salvia chinensis*, *Taraxacum mongolicum* Hand.-Mazz., *Hizikia fusiforme*, *Dendranthema indicum* (L.) Des Moul., *Pyrrosia lingua* (Thunb.) Farwell, *Wrightia laevis* Hook. f., *Arisaema heterophyllum* Blume, *Antenoron filiforme* (Thunb.) Rob. et Vaut., *Akebia quinata* (Houtt.) Decne., *Akebia trifoliata* (Thunb.) Koidz., *Actinidia arguta* (Sieb. & Zucc) Planch. ex Miq., *Aconitum carmichaelii* Debx., *Mirabilis jalapa* L., *Cynanchum paniculatum* (Bunge) Kitagawa, *Belamcanda chinensis* (L.) Redouté *Verbena officinalis* L., *Eriobotrya japonica* (Thunb.) Lindl., *Vaccaria segetalis*, *Dioscorea nipponica* Makino., *Fraxinus chinensis* Roxb., *Curcuma longa* L., *Caesalpinia sappan* Linn., *Schefflera venulosa* (Wight et Arn.) Harms, *Trichosanthes rosthornii* Harms, *Paris polyphylla*, *Pseudostellaria heterophylla* (Miq.) Pax, *Alisma plantago-aquatica* Linn., *Acanthopanax gracilistylus* W. W. Smith, *Ecklonia kurome* Okam, *Zingiber officinale* Rosc., *Xanthium sibiricum* Patr. ex Widder, *Chaenomeles sinensis* (Thouin) Koehne, *Cyrtomium fortunei* J. Sm., *Terminalia chebula* Retz., *Coix lacryma-jobi* L., *Iphigenia indica* Kunth, *Fagopyrum dibotrys* (D. Don) Hara, *Semiaquilegia adoxoides* (DC.) Makino, *Ceasostrea gigas* Thunberg, *Pleurotus ostreatus*, *Andrographis paniculata* (Burm. f.) Nees, *Impatiens balsamina* L., *Stellaria uliginosa* Murr., *Dendranthema morifolium* (Ramat.) Tzvel., *Gastrodia elata* Bl., *Cirsium setosum* (Willd.) MB., *Lilium brownii* var. *viridulum* Baker, *Panax pseudoginseng* Wall. var. *notoginseng* (Burkill) Hoo et Tseng, *Glycyrrhiza uralensis* Fisch., *Cirsium japonicum* Fisch. ex DC., *Nelumbo nucifera*, *Polygonum tinctorium* Ait., *Liquidambar formosana*, *Salvia miltiorrhiza* Bunge, *Pinus bungeana* Zucc. ex Endl., *Omphalia lapidescens* Schroet., *P. umbellatus*, *Abelmoschus esculentus* (Linn.) Moench, *Dianthus superbus* L., *Lasiosphaera seu calvatia*, *Hedyotis diffusa* Willd., *Oroxylum indicum* (L.) Kurz were purchased from Sankesong herbal medicine market, which were identified by Professor Zhenyu Wang who majored in Chinese Medicine classification, School of Chemistry and Chemical Engineering, Harbin Institute of Technology, Harbin, China. A voucher specimen (No. 201401-64) is deposited at the School of Chemistry and Chemical Engineering, Harbin Institute of Technology, P. R. China.

b. General Information

IR spectra was recorded on a Perkin Elmer Spectru FT-IR spectrophotometer as KBr pellets and the absorption frequencies were expressed in reciprocal centimeters (cm^{-1}). Mass spectral studies were carried out on a Hewlett-Packard 5989B MS (ESI) and Agilent 6890N GC-MS (EI). 1D NMR experiments were performed on a Bruker DRX-400 at 400MHz. Silica gel, Reversed phase silica gel, Sephadex LH-20, Macroporous resin were used for column chromatography. TLC were performed on silica gel GF254; chromatograms were visualized with UV light (254). Polarizing optical micrographs were recorded with a BS series microscope at room temperature. Scanning Electron Microscopy images of gels were recorded on a Quanta 200FEG SEM spectrometer at 20 kV, 10 mA, SE mode and spot size 3.5. Prior to observation, the xerogel was sputter coated with a thin layer of gold. HPLC system: The Agilent 1100 series HPLC. Chromatographic column: TC-C18 Analytical 4.6 x 250 mm 5-Micron column; UV detector; Column temperature: 30°C; Sample volume: 10 μL ;

c. Extraction and Isolation

Extraction of organism. Air-dried organism was homogenized and extracted with 95% ethanol ($\times 3$) under reflux for 2 h to obtain alcohol extract, which was evaporated to dryness and then suspended in distilled water. The suspension was extracted with chloroform ($\times 3$). The chloroform layers were evaporated to dryness under reduced pressure to obtain extract.

Extraction and isolation of six extracts

The air-dried the fruiting body of *Poria cocos* were powdered and extracted with 95% ethanol. The extractive solution was evaporated to dryness and the residues were dissolved in distilled water. The aqueous solution was extracted with chloroform to obtain the total extract. The total extract was subjected to vacuum pressure liquid chromatography on a D101 macroporous resin column (EtOH/H₂O, 0-100 %) to yield 7 fractions. Jelly-active testing confirmed that the gel fraction was present in the fraction 3 and 5. Fraction 3 was further chromatographed on a silica gel column eluted with a gradient of CH₂Cl₂/MeOH (0-100 %) to afford 3 fractions. Fraction 3a was separated by Sephadex LH-20 column eluted with CHCl₃/MeOH (1:1) to yield compound **1**. Fraction 5 was separated by using a silica gel column (n-hexane/ethyl acetate, 0-100 %) to afford 5 fractions. Fraction 5a was then repeatedly purified by chromatography on a reverse-phase RP-C18 column (MeOH/H₂O, 0-100 %), a Sephadex LH-20 column (CHCl₃/MeOH, 1:1) to give compound **2**.

The air-dried the root of *Taraxacum mongolicum* Hand.-Mazz. were homogenized and extracted with 95% EtOH-H₂O for two times with reflux, then the extractive solution was evaporated under reduced pressure to obtain the residue, which was dissolved in distilled water and extracted with chloroform for three times to obtain the extract. Then extract was subjected to a D101 macroporous resin column eluted with EtOH/H₂O step gradient system (0-100 %, v/v) to yield five fractions (Fr.1–Fr.5). Gel-active testing confirmed that the active fraction was present in the fraction 5. Fraction 5 was then repeatedly purified by chromatography on a silica gel column (Petroleum ether/ethyl acetate, 0-100%), a Sephadex LH-20 column (CHCl₃/MeOH, 1:1) to give **3**.

The air-dried the root of *Dioscorea nipponica* Makino were homogenized and extracted with 95% EtOH-H₂O for two times with reflux, then the extractive solution was evaporated under reduced pressure to obtain the residue, which was dissolved in distilled water and extracted with chloroform for three times to obtain the extract. The extract was subjected to a D101 macroporous resin column eluted with EtOH/H₂O step gradient system (0-100 %, v/v) to yield five fractions (Fr.1–Fr.4). Gel-active testing confirmed that the active fraction was present in the fraction 4. Fraction 3 was then repeatedly purified by chromatography on a silica gel column (petroleum ether/ethyl acetate, 0-100%) to give **4**.

The air-dried the fruiting body of *Pleurotus ostreatus* were homogenized and extracted with 95% EtOH-H₂O for two times with reflux, then the extractive solution was evaporated under reduced pressure to obtain the residue, which was dissolved in distilled water and extracted with chloroform for three times to obtain the extract. The extract was subjected to a D101 macroporous resin column eluted with EtOH/H₂O step gradient system (0-100 %, v/v) to yield five fractions (Fr.1–Fr.3). Gel-active testing confirmed that the active fraction was present in the fraction 3. Fraction 3 was then repeatedly purified by chromatography on a silica gel column (Petroleum ether/ethyl acetate, 0-100%), a Sephadex LH-20 column (CHCl₃/MeOH, 1:1) to give **5**.

The air-dried the tuber of *Gastrodia elata* Bl. were homogenized and extracted with 95% EtOH-H₂O for two times with reflux, then the extractive solution was evaporated under reduced pressure to obtain the residue, which was dissolved in distilled water and extracted with chloroform for three times to obtain the extract. The extract was subjected to a D101 macroporous resin column eluted with EtOH/H₂O step gradient system (0-100 %, v/v) to yield five fractions (Fr.1–Fr.5). Gel-active testing

confirmed that the active fraction was present in the fraction 1. Fraction 1 was then repeatedly purified by chromatography on a reverse-phase RP-C18 column (MeOH/H₂O, 0-100 %), a silica gel column (ethanol/ ethyl acetate, 0-100%) to give **6**.

The air-dried the fruit of *Chaenomeles sinensis* (Thouin) Koehne (10 kg) were homogenized and extracted with 95% EtOH-H₂O for two times with reflux, then the extractive solution was evaporated under reduced pressure to obtain the residue, which was dissolved in distilled water and extracted with chloroform for three times to obtain the extract. The extract was subjected to a D101 macroporous resin column eluted with EtOH/H₂O step gradient system (0-100 %, v/v) to yield five fractions (Fr.1–Fr.6). Gel-active testing confirmed that the active fraction was present in the fraction 2. Fraction 2 was then repeatedly purified by chromatography on a silica gel column (Petroleum ether/ethyl acetate, 0-100%), a RP-C18 column (MeOH/H₂O, 0-100%), Sephadex LH-20 column (CHCl₃/MeOH, 1:1) to give **7**.

d. Gelation Tests

Gelation tests of extracts: In a test tube (1.5 mL volume, 10 mm diameter), 50 mg sample was mixed with 0.5 mL solvent and the mixture was heated until the solid dissolved as much as possible. The resulting solution was cooled to 20 °C and then allowed to stand for 24 h at this temperature. When the test tube could be inverted without change of shape of its content, it was identified as a gel (G). If slow change of shape of its content was detectable when the test tube was inverted, then the sample was designated as a viscous (V). Test tube with only liquid was referred to as a solution (S). When a precipitate appeared after the gelator dissolved in a liquid or could not be dissolved, the designations were precipitate (P) and insoluble (I), respectively. All samples concentrations were expressed in % as the ratio of gelator weight (mg) to liquid volume (mL). Ethanol and water mixtures were expressed as volume ratios.

Gelation tests of responsible compounds: Except that the amount of sample used is 10mg, the rest are the same as the extract test method.

The critical gelator concentrations (CGC) are the lowest gelator concentrations which produced gels when samples are prepared by the above method.

e. Rheology Measurements.

The measurements were performed using a Kinexus pro+ Rheometer, equipped with a temperature controller and parallel stainless steel plates (20 mm diameter, 0.5 mm gap). A hot solution of sample in solvent was placed in the shearing gap of the rheometer and was allowed to incubate for 0.5 h at 15 °C. Subsequently it was submitted to a frequency sweep or stress sweep experiment. To minimize solvent evaporation, a liquid trap device was placed over the sample chamber.

f. Identification of Compounds 1-7

Compound **1** was obtained as a white amorphous; powder purity, 97.94% (HPLC); HRESIMS m/z 521.3251 (calc. 521.3237) [M+Na]⁺ (C₃₁H₄₆O₅). The IR spectrum displayed absorption bands for hydroxyl (at 3288 cm⁻¹), carbonyl (at 1708 cm⁻¹) and vinyl (at 1643 cm⁻¹). ¹H-NMR (C₅D₅N, 400MHz) δ 5.35(1H, s, H-11), 5.30(1H, s, H-7), 4.99(1H, s, H-31a), 4.86(1H, s, H-31b), 4.84(1H, s, H-28a), 4.78(1H, s, H-28b), 4.53(1H, dd, H-16), 2.95(1H, dd, H-20), 2.86(1H, dd, H-17), 1.75, 1.51, 1.11, 1.04, 1.00, 0.99(6 × CH₃); ¹³C-NMR(C₅D₅N, 100MHz) δ 36.19(C-1), 30.03(C-2), 176.46(C-3), 149.10 (C-4),

50.58(C-5), 28.39(C-6), 117.80(C-7), 141.66(C-8), 137.31(C-9), 38.65(C-10), 120.11(C-11), 36.80(C-12), 45.44(C-13), 49.13(C-14), 43.63(C-15), 76.24(C-16), 57.43(C-17), 48.27(C-20), 178.47(C-21), 31.18(C-22), 33.04(C-23), 155.85(C-24), 33.90(C-25), 21.85(C-26), 21.69(C-27), 112.00(C-28), 22.09(C-29), 24.67(C-30), 106.87(C-31). Compound **1** was identified as poricoic acid A by comparison of its spectroscopic data with this reported in the literature¹.

Compound **2** was obtained as a white needle crystal (chloroform); purity, 99.40% (HPLC); HRESIMS m/z 455.3511 (calc. 455.3520) $[M+H]^+$ ($C_{30}H_{46}O_3$). The IR spectrum displayed absorption bands for hydroxyl (at 3467 and 3230 cm^{-1}), carbonyl (at 1687 cm^{-1}) and vinyl (at 1643 cm^{-1}). 1H -NMR (C_5D_5N , 400MHz) δ 5.62(1H, d, H-7), 5.38(1H, d, H-11), 5.34 (1H, d, H-24), 3.46(1H, d, H-3), 2.67(1H, dd, H-20), 1.67, 1.64, 1.23, 1.14, 1.09, 1.08, 1.02(7 \times CH_3); ^{13}C -NMR(C_5D_5N , 100MHz) δ 36.34(C-1), 28.68(C-2), 78.00(C-3), 39.35 (C-4), 49.73(C-5), 23.55(C-6), 121.33(C-7), 142.80(C-8), 146.62(C-9), 37.85(C-10), 116.61(C-11), 36.02(C-12), 44.26(C-13), 50.48(C-14), 31.61(C-15), 27.30(C-16), 48.17(C-17), 16.28(C-18), 22.99(C-19), 48.98(C-20), 178.55(C-21), 33.29(C-22), 26.76(C-23), 124.85(C-24), 131.78(C-25), 25.81(C-26), 17.75(C-27), 28.85(C-28), 16.65(C-29), 25.87(C-30). Compound **2** was identified as dehydrotrametenolic acid by comparison of its spectroscopic data with this reported in the literature².

Compound **3** was obtained as white amorphous powder; purity, 95.3% (HPLC); EIMS m/z : 426 $[M]^+$ ($C_{30}H_{50}O$). The IR spectrum displayed absorption bands for hydroxyl (at 3350 cm^{-1}) and vinyl (at 1640 cm^{-1}). 1H -NMR (C_5D_5N , 400MHz) δ 4.91(1H, s, H-29a), 4.76(1H, s, H-29b), 3.49(1H, dt, H-3), 2.51(1H, dt, H-19), 1.77, 1.27, 1.08, 1.07, 1.01, 0.91, 0.85(7 \times CH_3); ^{13}C -NMR(C_5D_5N , 100MHz) δ 151.49(C-20), 110.40(C-29), 78.56(C-3), 56.33(C-5), 51.23(C-9), 49.06(C-18), 48.72(C-19), 43.67(C-17), 43.51(C-14), 41.59(C-8), 40.69(C-22), 40.00(C-1), 39.76(C-4), 38.78(C-13), 37.95(C-10), 36.27(C-16), 35.17(C-7), 30.60(C-21), 29.14(C-2), 28.79(C-23), 28.25(C-15), 26.02(C-12), 21.59(C-11), 19.93(C-30), 19.27(C-6), 18.65(C-28), 16.93(C-25), 16.84(C-26), 16.67(C-24), 15.20(C-27). Compound **3** was identified as lupeol by comparison of its spectroscopic data with this reported in the literature³.

Compound **4** was obtained as white crystal; purity, 92.3% (HPLC); EIMS m/z : 415.3207 $[M+H]^+$ ($C_{27}H_{42}O_3$). The IR spectrum displayed absorption bands for hydroxyl (at 3319 cm^{-1}) and vinyl (at 1658 cm^{-1}). 1H -NMR ($CDCl_3$, 400MHz) δ 5.35(1H, m, H-6), 3.34(1H, m, H-3), 1.83, 1.11, 1.03, 0.96 (4 \times CH_3); ^{13}C -NMR($CDCl_3$, 100MHz) δ 14.30(C-21), 16.06(C-18), 17.01(C-27), 19.19(C-19), 20.65(C-11), 28.58(C-24), 30.10(C-25), 31.20(C-23), 31.40(C-2), 31.63(C-15), 32.01(C-8), 32.03(C-7), 36.43(C-10), 37.01(C-1), 39.57(C-12), 40.05(C-13), 41.39(C-4), 42.05(C-20), 50.01(C-9), 56.29(C-14), 62.01(C-17), 66.63(C-26), 71.47(C-3), 80.20(C-16), 109.57(C-22), 121.29(C-6), 140.29(C-5). Compound **4** was identified as diosgenin by comparison of its spectroscopic data with this reported in the literature⁴.

Compound **5** was obtained as colorless needle crystal; purity, 95.3% (HPLC); EIMS m/z : 396 $[M]^+$ ($C_{28}H_{44}O$). The IR spectrum displayed absorption bands for hydroxyl (at 3412, 3324 cm^{-1}) and vinyl (at 1772, 1656, 1604 cm^{-1}). 1H -NMR ($CDCl_3$, 400MHz) δ 5.57(1H, d, H-6), 5.38(1H, d, H-7), 5.20 (2H, m, H-22, 23), 3.62(1H, m, H-3), 1.03, 0.94, 0.91, 0.84, 0.82, 0.63 (6 \times CH_3); ^{13}C -NMR($CDCl_3$, 100MHz) δ 141.49(C-8), 139.92(C-5), 135.71(C-22), 132.12(C-23), 119.73(C-6), 116.43(C-7), 70.59(C-3), 55.87(C-17), 54.70(C-14), 46.39(C-9), 42.97(C-13, C-24), 40.93 (C-4), 40.57(C-20), 39.23(C-12), 38.52(C-1), 37.17(C-10), 33.23(C-25), 32.12(C-2), 28.43(C-16), 23.14(C-15), 21.25(C-11, C-21), 20.10(C-26), 19.79(C-27), 17.75(C-28), 16.42(C-19), 12.19(C-18). Compound **5** was identified as Ergosterol by comparison of its spectroscopic data with this reported in the literature⁵.

Compound **6** was obtained as white crystal; purity, 98.3% (HPLC); FABMS m/z : 309 $[M+Na]^+$ ($C_{13}H_{18}O_7$). The IR spectrum displayed absorption bands for hydroxyl (at 3443 cm^{-1}) and phenyl (at 1610, 1590, 1515 cm^{-1}). 1H -NMR ($DMSO-d_6$, 400MHz) δ 7.21(2H, d, H-2, 6), 6.97(2H, d, H-3, 5),

4.96-5.26(4H, m, OH-2', 3', 4', 6'), 4.82(1H, d, H-1'), 4.53(1H, t, ArCH₂OH), 4.41(2H, d, ArCH₂OH), 3.38-3.69(2H, m, H-6'), 3.05-3.32 (4H, m, H-2', 3', 4', 5'). ¹³C-NMR(DMSO-d₆, 100MHz) δ 60.6(C-6'), 62.4 (ArCH₂OH), 69.6(C-4'), 73.1(C-2'), 76.5(C-5'), 76.8(C-3'), 100.3(C-1'), 115.8(C-3, 5), 127.4(C-2, 6), 135.6(C-1), 156.0(C-4). Compound **6** was identified as gastrodin by comparison of its spectroscopic data with this reported in the literature⁶.

Compound **7** was obtained as white amorphous powder; purity, 92.3% (HPLC); HRESIMS m/z: 479.3504[M+Na]⁺ (C₃₀H₄₈O₃). The IR spectrum displayed absorption bands for hydroxyl (at 3527cm⁻¹), carboxyl and vinyl (at 1716 cm⁻¹). ¹H-NMR (DMSO-d₆, 400MHz) δ 11.94(1H, brs, H-28), 5.12 (1H, t, H-12), 3.00 (1H, brs, H-3), 2.01(1H, d, H-18), 1.04, 0.90, 0.87, 0.81, 0.75, 0.67 (6×-CH₃); ¹³C-NMR(DMSO-d₆, 100MHz) δ 178.26 (C-28), 138.19 (C-13), 124.56 (C-12), 76.82 (C-3), 54.79 (C-5), 52.37 (C-18), 47.02 (C-9), 46.81 (C-17), 41.63 (C-14), 38.50 (C-4), 38.45 (C-19), 38.37 (C-8), 38.24 (C-1), 36.52 (C-22), 36.32 (C-10), 32.71 (C-7), 30.20 (C-21), 28.25 (C-23), 27.54 (C-15), 26.99 (C-2), 23.81 (C-11), 23.27 (C-27), 22.85 (C-16), 21.08 (C-30), 18.00 (C-6), 17.01 (C-26), 16.90 (C-29), 16.07 (C-24), 15.22 (C-25). Compound **2** was identified as ursolic acid by comparison of its spectroscopic data with this reported in the literature⁷.

2. Supporting Tables

Table S1. Information and Gelation Results for 63 Organism Extracts

No.	Name	Family	Part researched	Extraction rate(%)	C ₆ H ₁₄ ^a	CHCl ₃	CH ₃ OH	CH ₃ OH - H ₂ O ^b
1	<i>Euchresta japonica</i>	Leguminosae	root	0.83	P	P	P	P
2	<i>Salvia chinensis</i>	Labiatae	aerial parts	1.12	P	S	S	V
3	<i>Taraxacum mongolicum</i> H and.-Mazz.	Compositae	root	2.49	P	S	G	-
4	<i>Hizikia fusiforme</i>	Sargassaceae	whole plant	0.07	P	P	P	P
5	<i>Dendranthema indicum</i> (L.) Des Moul.	Compositae	flower	2.80	P	S	G	-
6	<i>Pyrrosia lingua</i> (Thunb.) Farwell	Polypodiaceae	whole herb	1.31	P	S	P	P
7	<i>Wrightia laevis</i> Hook. f.	Apocynaceae	root	0.21	P	S	P	P
8	<i>Arisaema heterophyllum</i> Blume	Araceae	tuber	0.33	P	P	P	P
9	<i>Antenoron filiforme</i> (Thunb.) Rob. et Vaut.	Polygonaceae	whole herb	0.31	P	P	G	-
10	<i>Akebia quinata</i> (Houtt.) Decne.	Lardizabalaceae	tuber	0.23	P	P	P	P
11	<i>Akebia trifoliata</i> (Thunb.) Koidz.	Lardizabalaceae	tuber	0.98	P	S	P	P
12	<i>Actinidia arguta</i> (Sieb. & Zucc) Planch. ex Miq.	Actinidiaceae	root	0.31	P	P	P	P
13	<i>Aconitum carmichaelii</i> Debx.	Ranunculaceae	whole herb	0.10	P	S	P	P
14	<i>Mirabilis jalapa</i> L.	Nyctaginaceae	root	1.24	P	S	P	-
15	<i>Cynanchum paniculatum</i> (Bunge) Kitagawa	Asclepiadaceae	tuber	1.12	P	S	P	P
16	<i>Belamcanda chinensis</i> (L.) Redout é	Iridaceae	rhizome	1.13	P	P	P	P
17	<i>Verbena officinalis</i> L.	Verbenaceae	aerial parts	0.69	P	S	P	P
18	<i>Eriobotrya japonica</i> (Thunb.) Lindl.	Rosaceae	leaf	1.72	P	P	G	-
19	<i>Vaccaria segetalis</i>	Caryophyllaceae	seed	1.28	P	S	P	P
20	<i>Dioscorea nipponica</i> Makino.	Dioscoreaceae	root	0.99	P	S	G	-
21	<i>Fraxinus chinensis</i> Roxb.	Oleaceae	bark	0.27	P	S	V	P
22	<i>Curcuma longa</i> L.	Zingiberaceae	rhizome	1.10	P	S	P	P
23	<i>Caesalpinia sappan</i> Linn.	Leguminosae	heartwood	0.16	P	P	P	P

24	<i>Schefflera venulosa</i> (Wight et Arn.) Harms	Araliaceae	aerial parts	0.22	P	S	P	P
25	<i>Trichosanthes rosthornii</i> Harms	Cucurbitaceae	peel	0.15	P	P	P	P
26	<i>Paris polyphylla</i>	Liliaceae	aerial parts	1.24	P	P	P	P
27	<i>Pseudostellaria heterophylla</i> (Miq.) Pax	Caryophyllaceae	root	0.29	P	P	P	P
28	<i>Alisma plantago-aquatica</i> Linn.	Alismataceae	tuber	3.93	P	S	S	P
29	<i>Acanthopanax gracilistylus</i> W. W. Smith	Araliaceae	root	1.99	P	S	P	P
30	<i>Ecklonia kurome</i> Okam	Alariaceae	whole plant	0.30	P	P	P	P
31	<i>Zingiber officinale</i> Rosc.	Zingiberaceae	rhizome	1.97	P	S	P	P
32	<i>Xanthium sibiricum</i> Patr. ex Widder	Compositae	aerial parts	11.03	P	S	P	P
33	<i>Chaenomeles sinensis</i> (Thouin) Koehne	Rosaceae	fruit	0.84	P	G	G	-
34	<i>Cyrtomium fortunei</i> J. Sm.	Dryopteridaceae	rhizome	8.70	P	P	P	P
35	<i>Terminalia chebula</i> Retz.	Combretaceae	fruit	0.47	P	S	G	-
36	<i>Coix lacryma-jobi</i> L.	Gramineae	seed	2.74	P	S	P	P
37	<i>Iphigenia indica</i> Kunth	Liliaceae	stem	0.14	P	P	G	-
38	<i>Fagopyrum dibotrys</i> (D. Don) Hara	Polygonaceae	rhizome	0.23	P	S	P	P
39	<i>Semiaquilegia adoxoides</i> (DC.) Makino	Ranunculaceae	root	0.13	P	S	P	P
40	<i>Ceassostrea gigas</i> Thunberg	Ostreidae	shell	0.01	-	-	-	-
41	<i>Pleurotus ostreatus</i>	Pleurotaceae	fruiting body	2.33	G	S	P	-
42	<i>Andrographis paniculata</i> (Burm. f.) Nees	Acanthaceae	aerial parts	0.54	P	S	G	-
43	<i>Impatiens balsamina</i> L.	Balsaminaceae	whole herb	0.19	P	S	P	P
44	<i>Stellaria uliginosa</i> Murr.	Caryophyllaceae	whole herb	0.65	P	S	P	P
45	<i>Dendranthema morifolium</i> (Ramat.) Tzvel.	Compositae	flower	1.02	P	S	G	-
46	<i>Gastrodia elata</i> Bl.	Orchidaceae	tuber	0.45	P	S	G	-
47	<i>Cirsium setosum</i> (Willd.) MB.	Compositae	aerial parts	2.45	P	S	G	-
48	<i>Lilium brownii</i> var. <i>viridulum</i> Baker	Liliaceae	scale leaf	0.57	P	S	G	-
49	<i>Panax pseudoginseng</i> Wall. var. <i>notoginseng</i> (Burkill) Hoo et Tseng	Araliaceae	root	0.07	P	P	P	P
50	<i>Glycyrrhiza uralensis</i> Fisch.	Leguminosae	root	0.66	P	S	V	V

51	<i>Cirsium japonicum</i> Fisch. ex DC.	Compositae	aerial parts	2.21	P	S	P	P
52	<i>Nelumbo nucifera</i>	Nymphaeaceae	flower	1.73	P	S	G	-
53	<i>Polygonum tinctorium</i> Ait.	Polygonaceae	stem and leaf	1.05	P	P	P	P
54	<i>Liquidambar formosana</i>	Hamamelidaceae	fruit	0.75	P	S	G	-
55	<i>Salvia miltiorrhiza</i> Bunge	Labiatae	aerial parts	0.90	P	S	S	P
56	<i>Pinus bungeana</i> Zucc. ex Endl.	Pinaceae	pine cone	5.62	P	S	P	P
57	<i>Omphalia lapidescens</i> Schroet.	Tricholomataceae	scleroti-um	0.79	P	P	P	P
58	<i>P. umbellatus</i>	Polyporaceae	scleroti-um	0.36	P	P	P	P
59	<i>Abelmoschus esculentus</i> (Linn.) Moench	Malvaceae	fruit	1.75	P	P	P	P
60	<i>Dianthus superbus</i> L.	Caryophyllaceae	whole herb	0.77	P	P	P	-
61	<i>Lasiosphaera seu calvatia</i>	Lvcoperdaceae	fruiting body	0.21	P	P	P	P
62	<i>Hedyotis diffusa</i> Willd.	Rubiaceae	whole herb	1.78	P	P	P	P
63	<i>Oroxylum indicum</i> (L.) Kurz	Bignoniaceae	seed	13.18	P	P	P	P

* G= gel; V= viscous; S=solutions; P= precipitate; "-" not determined; ^a n-hexane; ^b Alcohol-water=1: 0.7.
Tested concentration=10 % w/v

Table S2. Gelation Results for Compounds 1-6 in 48 Solvents and Morphology for Natural Product Gels

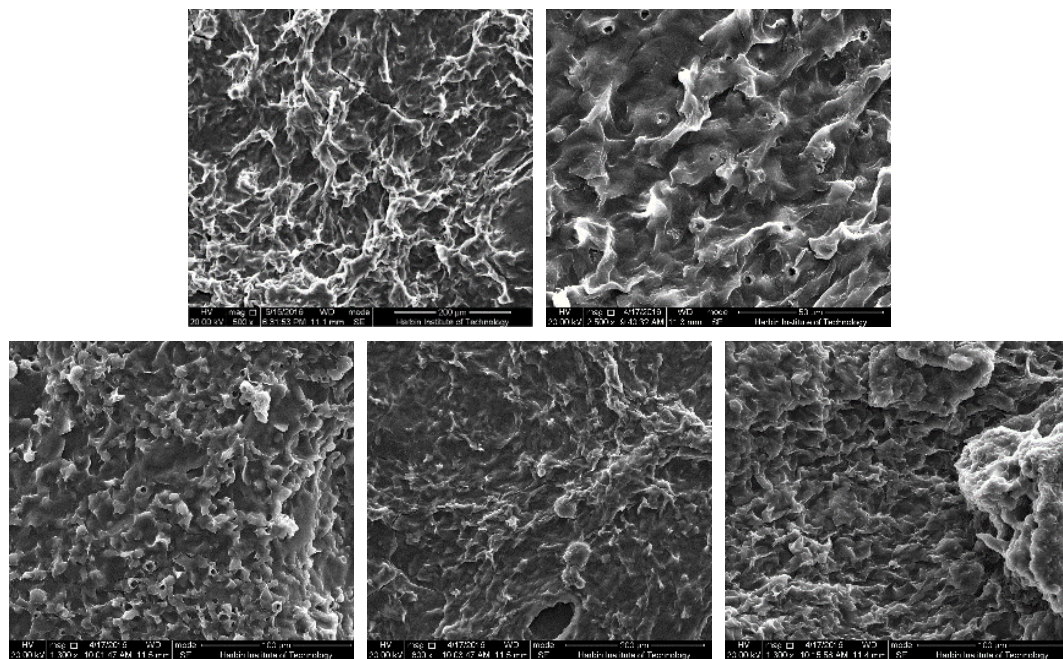
Solvents	State (CGC ³)/ Morphology					
	1	2	3	4	5	6
Methanol	S	S	G (1.50) rod-like	G(1.50) fibrous	I	S
Ethanol	S	S	S	V	G(1.50) lamellar	S
n-Propanol	S	S	S	R	R	S
n-Butanol	S	S	S	S	S	I
n-Pentanol	S	S	S	S	S	I
n-Hexanol	S	S	S	S	S	I
n-Heptanol	S	S	S	S	S	I
n-Octanol	S	S	S	S	S	I
n-Nonanol	S	S	S	S	S	I
n-Decanol	S	S	S	S	S	I
Ethylene glycol	P	P	P	I	I	I
Isopropanol	S	S	S	G(2.00) pellet ^b	G(2.00) lamellar	S
Benzyl alcohol	S	S	S	S	S	I
Benzene	I	G(0.21) fibrous	S	R	P	I
Toluene	I	V	S	S	P	I
o-Xylene	I	G(1.90) fibrous	S	S	P	I
m-Xylene	I	G(2.00) fibrous	S	S	P	I
p-Xylene	I	G(1.50) fibrous	S	S	P	I
Mesitylene	I	G(2.00) fibrous	S	S	P	I
Chlorobenzene	I	G(2.00) fibrous	S	S	P	I
Bromobenzene	I	G(2.00) fibrous	S	S	P	I
o-Dichlorbenzene	I	G(2.00) fibrous	S	S	P	I

Nitrobenzene	I	V	S	G(0.67) pleated	G(1.50) rod-like	I
Dichloromethane	I	G(0.80) fibrous	S	S	S	I
Chloroform	I	G(1.50) fibrous	S	S	S	I
Carbon tetrachloride	I	G(1.10) fibrous	S	S	P	I
1,1,2,2-Tetrachloroethane	I	G(2.00) fibrous	S	S	S	I
Cyclohexane	I	I	S	G(1.00) rod-like	G(1.00) pleated	I
n-Hexane	I	I	G(1.60) fibrous	G(1.00) fibrous	P	I
n-Heptane	I	I	G(2.00) fibrous	G(1.25) rod-like	P	I
Triethylamine	S	V	S	S	S	I
Butylamine	I	S	S	S	S	I
Aniline	S	S	S	S	R	I
Benzylamine	S	S	S	S	S	I
DMSO	S	S	G(2.00) lamellar	V	G(0.70) lamellar	S
DMF	S	S	S	S	R	S
Pyridine	S	S	S	S	S	S
THF	S	S	S	S	S	I
Acetonitrile	G(0.90) fibrous	G(2.00) fibrous	G(0.40) fibrous	G(1.40) fibrous	R	G(1.00) fibrous
Acetic acid	S	S	R	V	I	S
Acetone	S	I	S	G(2.00) rod-like	R	G(0.80) fibrous
Ethyl acetate	I	P	S	G(2.00) rod-like	S	G(1.00) pleated
Petroleum ether	I	I	G(1.00) fibrous	V	P	I
Diethyl ether	V	I	S	G(1.50) rod-like	I	I
Methanol-water ^c	G(0.80) fibrous	I	I	I	I	S
Ethanol-water ^c	G(1.00) fibrous	I	I	I	I	S
n-Propanol-water ^c	G(1.30) fibrous	I	I	I	I	S

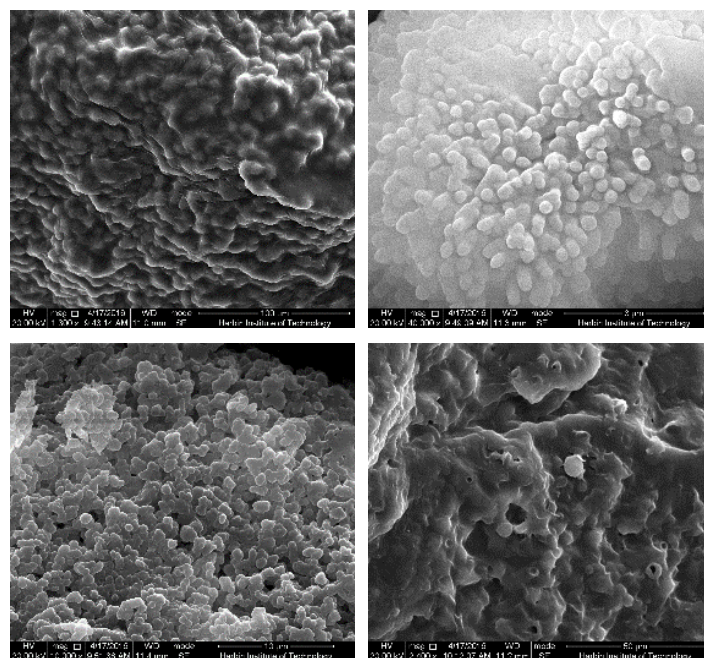
Water	I	I	I	I	I	S
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* G = gel; V = viscous; S = solutions; I = insoluble; P = precipitate; ^a Critical gelation concentration; ^b Linked pellet; ^c Alcohol:water = 1:0.7; Tested concentration=2% w/v;

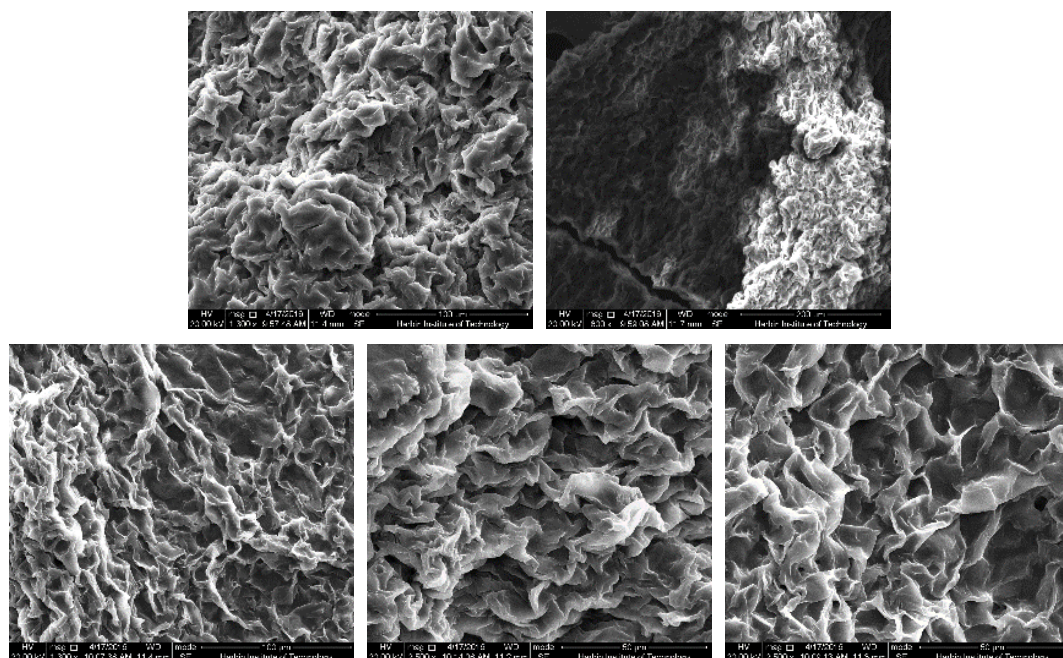
3. Supporting Figures



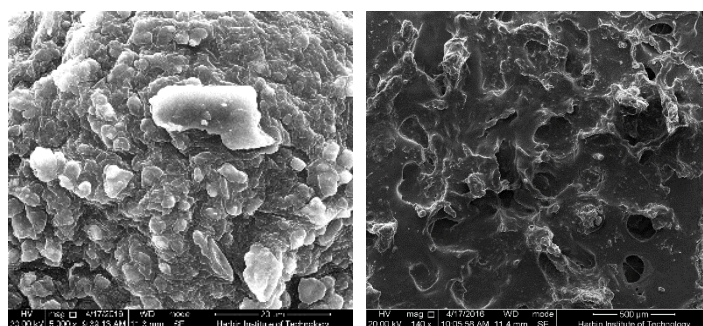
flake (number in **Table 1**: 3, 9, 41, 42, 52)



spherical (18, 20, 33, 47)

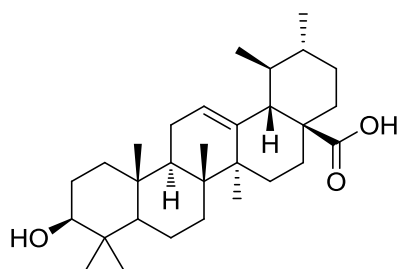


folds (35, 37, 46, 48, 54)



irregular (5, 45)

Figure S1. SEM Images of 16 Extracts Gels



7, Ursolic acid

Figure S2. Structure of the compound **7**

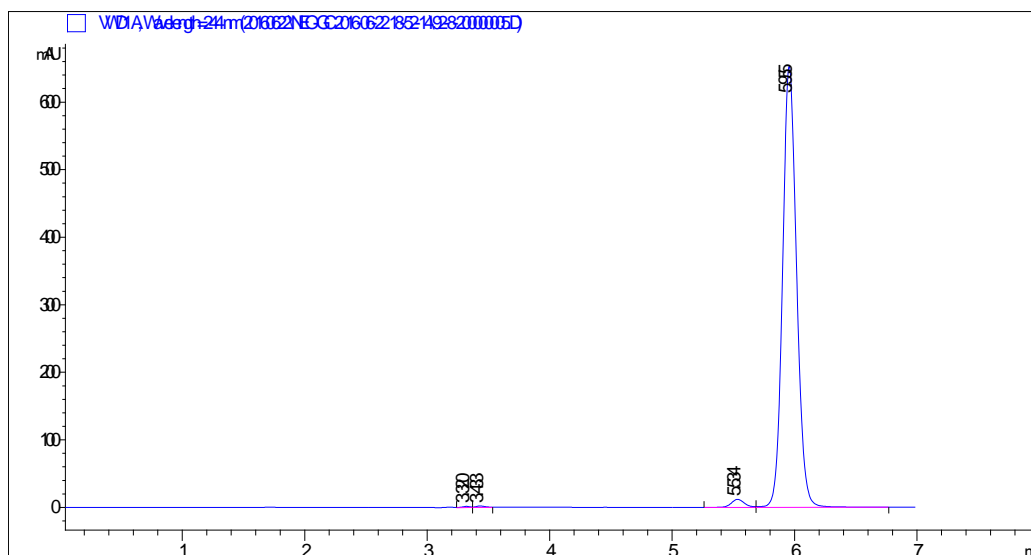


Figure S3-1. Determination of the purity of the compound **1** (Mobile phase: Methanol-0.1% acetic acid aqueous solution=90:10; Detection wavelength: 244 nm; Flow rate: 1 mL/min; Column temperature: 30°C; t_R (97.94% purity) = 5.955 min.)

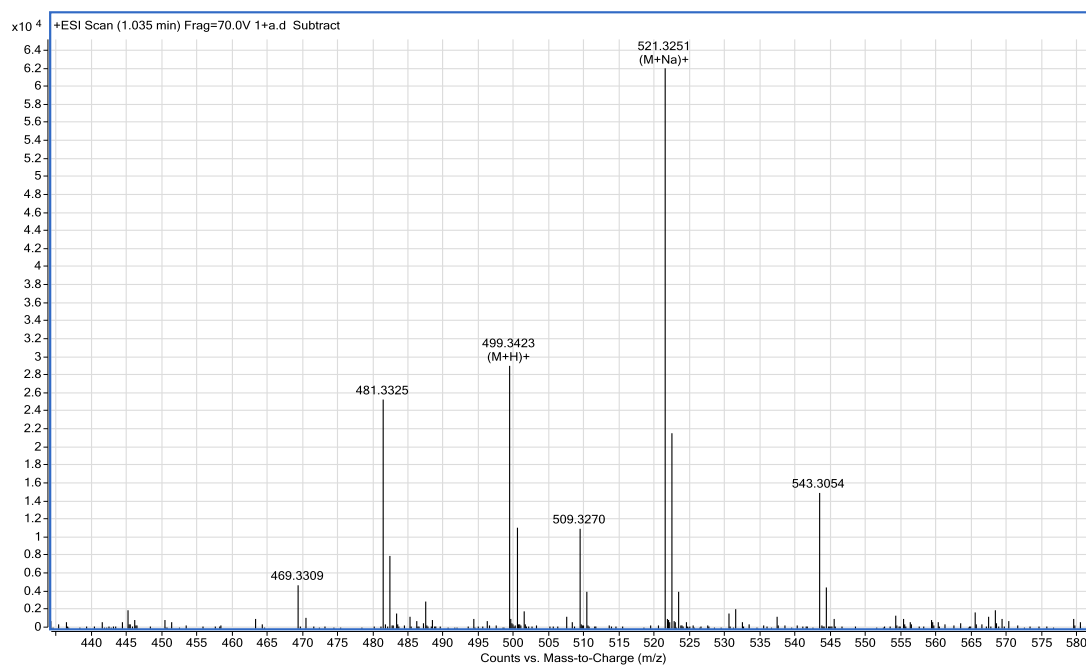


Figure S3-2. HRESIMS of compound **1**.

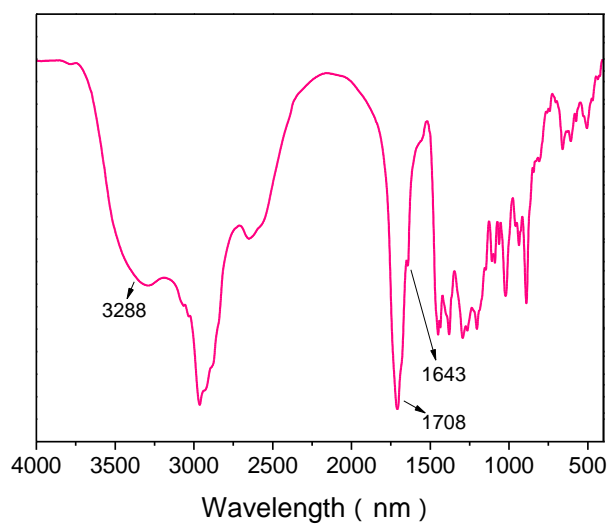


Figure S3-3. IR of compound **1**.

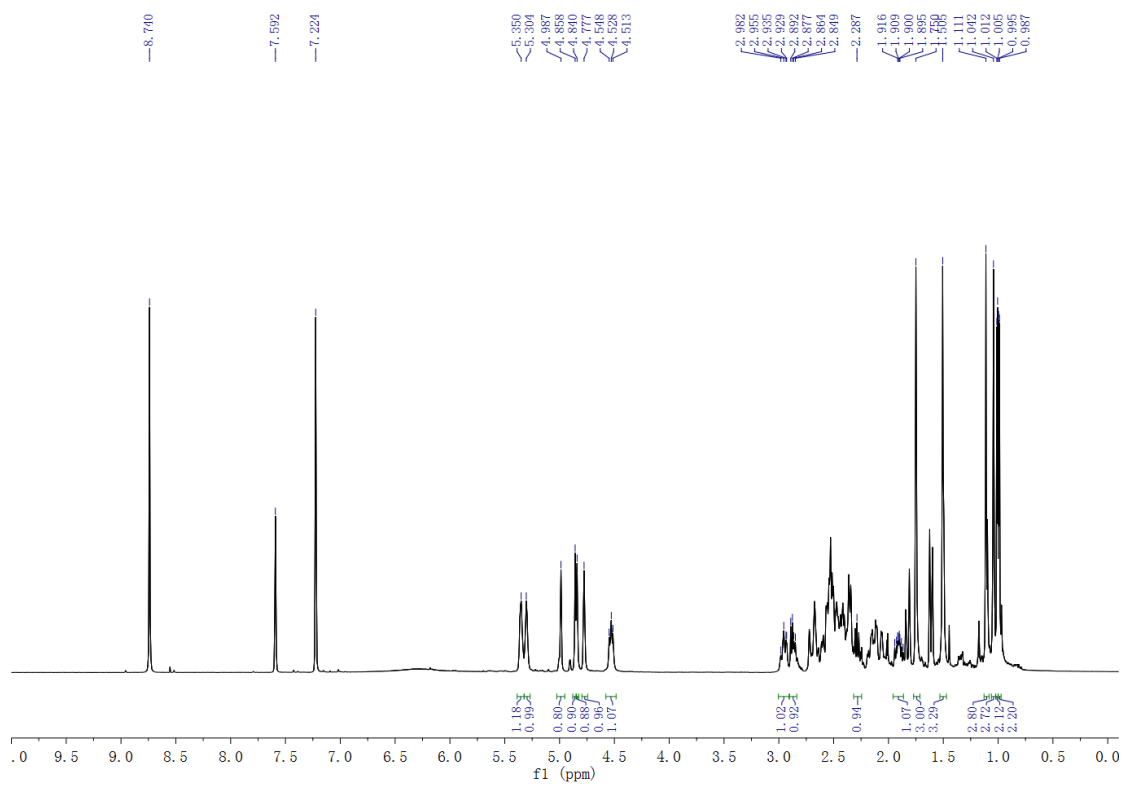


Figure S3-4. ¹H-NMR of compound **1** in C₅D₅N .

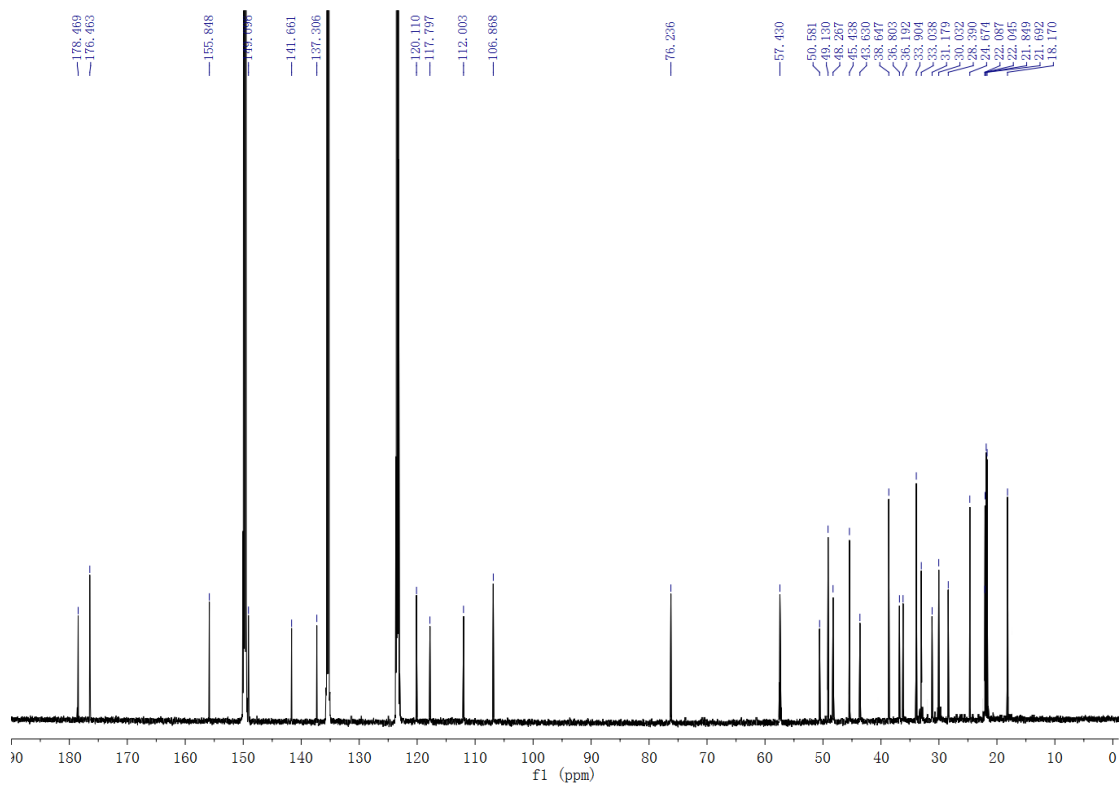


Figure S3-5. ^{13}C -NMR of compound **1** in $\text{C}_5\text{D}_5\text{N}$.

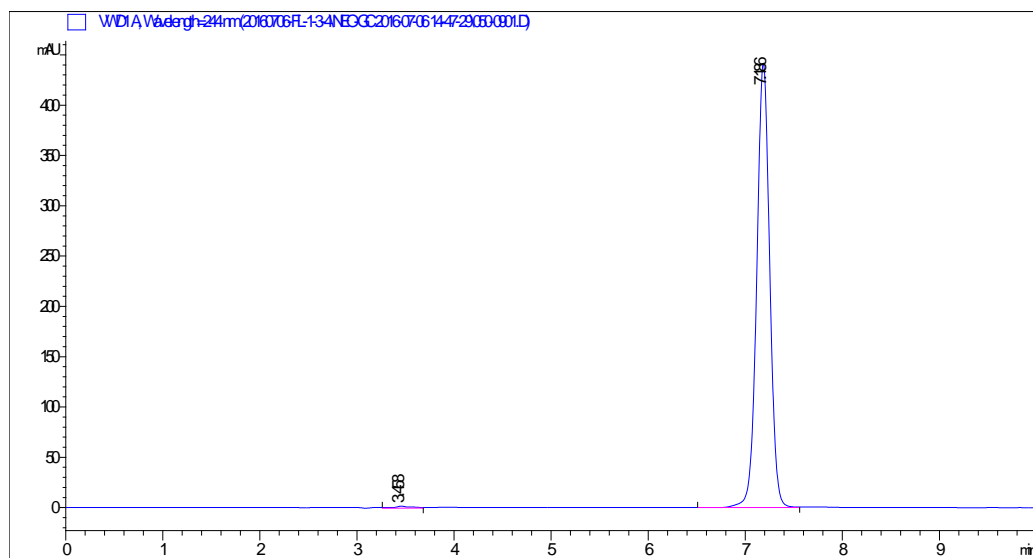


Figure S3-6. Determination of the purity of the compound **2** (Mobile phase: Methanol-0.1% acetic acid aqueous solution=90:10; Detection wavelength: 244 nm; Flow rate: 1 mL/min; Column temperature: 30 °C; t_R (99.40% purity) = 7.186 min.)

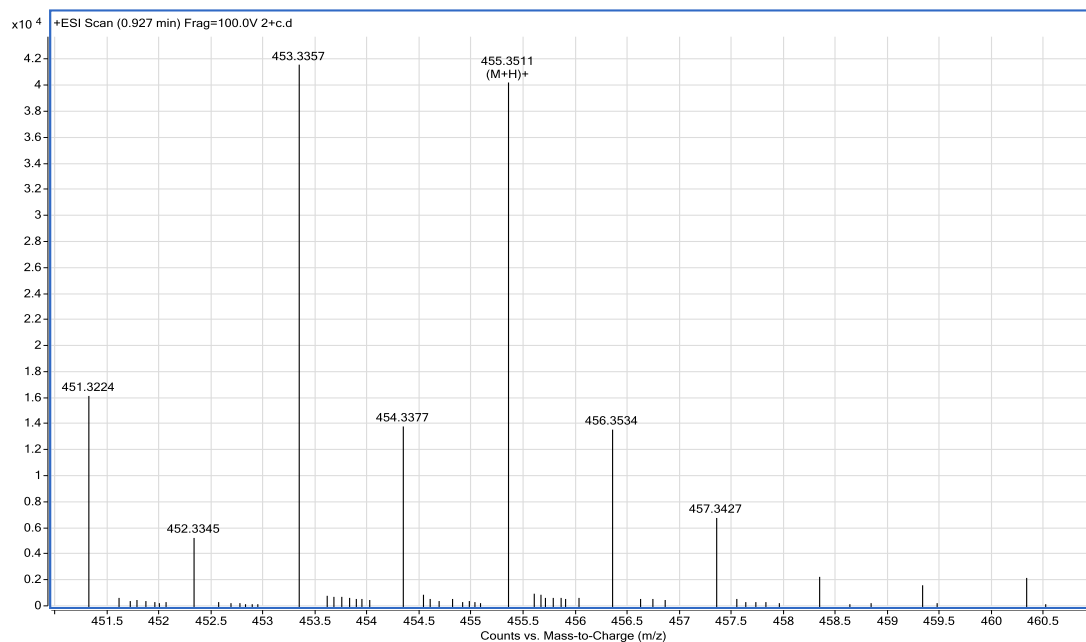


Figure S3-7. HRESIMS of compound **2**.

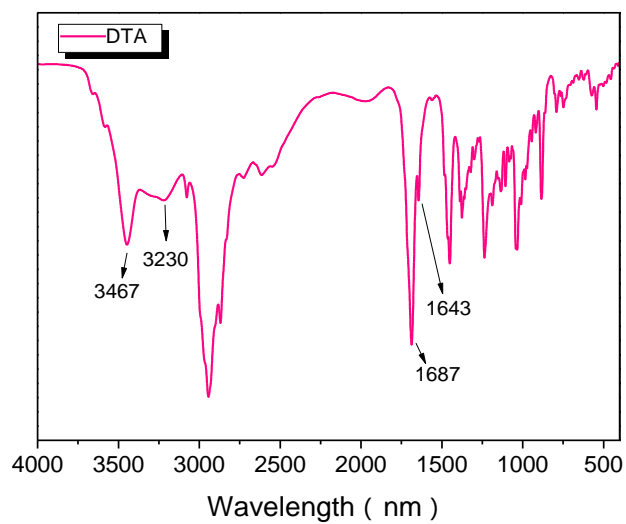


Figure S3-8. IR of compound **2**.

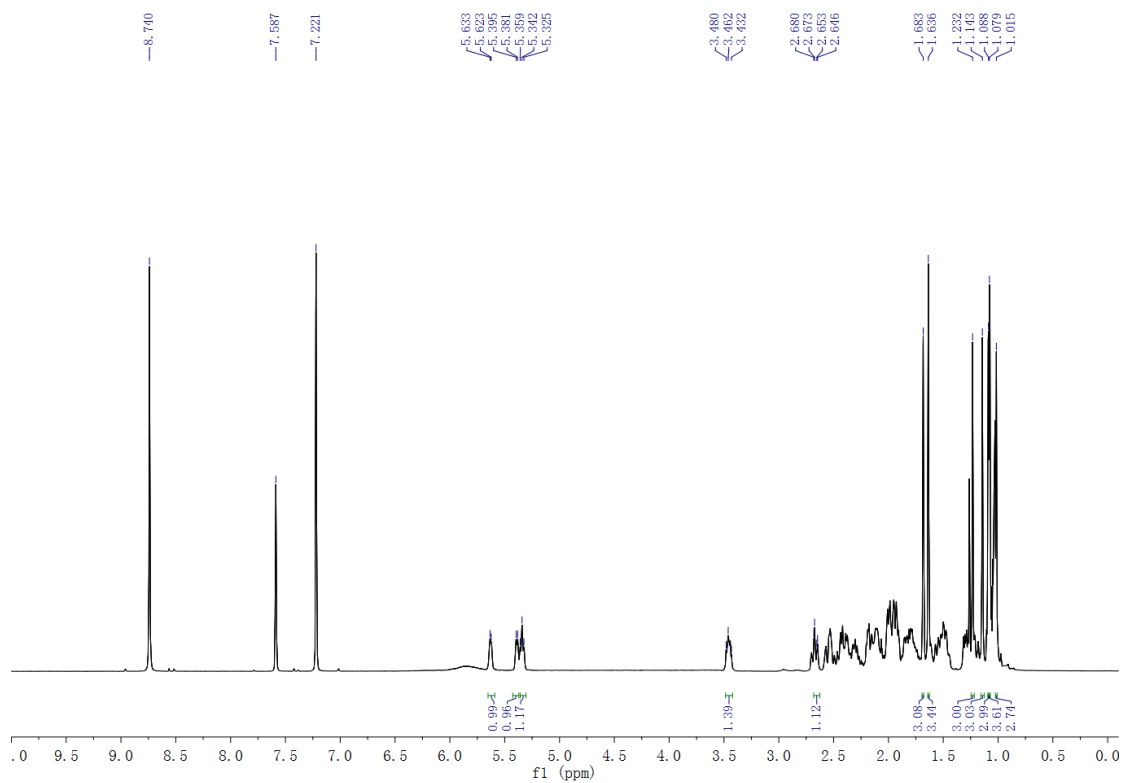


Figure S3-9. ¹H-NMR of compound **2** in C₅D₅N .

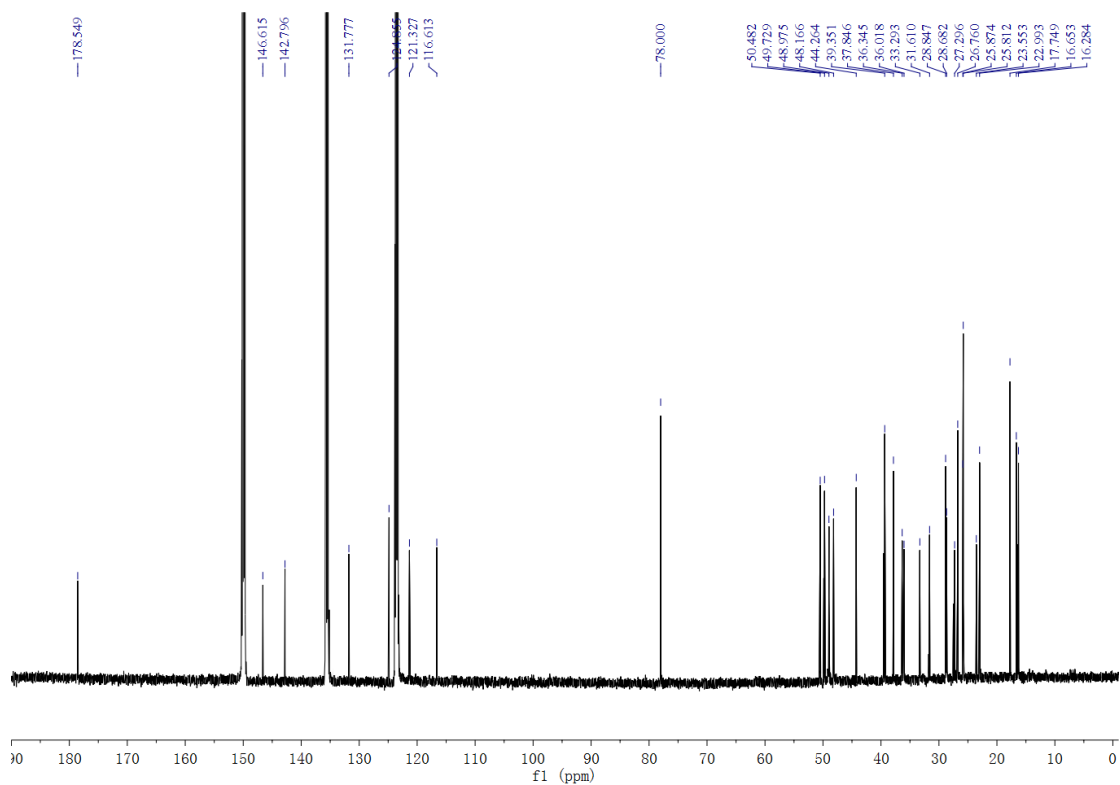


Figure S3-10. ¹³C-NMR of compound **2** in C₅D₅N .

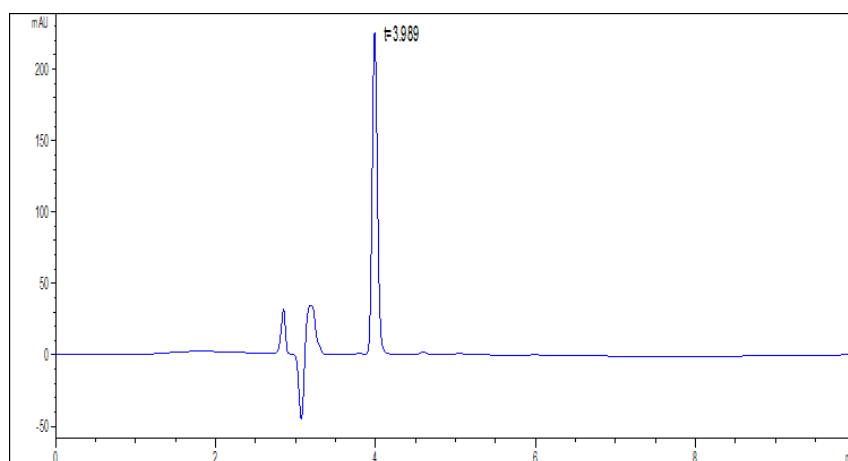


Figure S3-11. Determination of the purity of the compound **3** (Mobile phase: Methanol-0.1% acetic acid aqueous solution=90:10; Detection wavelength: 210nm; Flow rate: 1 mL/min; Column temperature: 30°C; t_R (95.3% purity) = 3.989 min.)

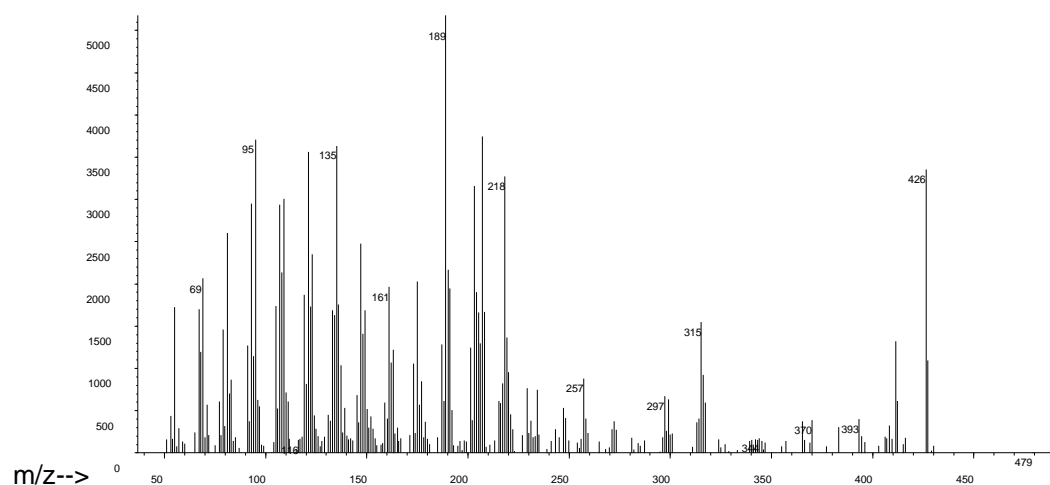


Figure S3-12. MS of compound **3**.

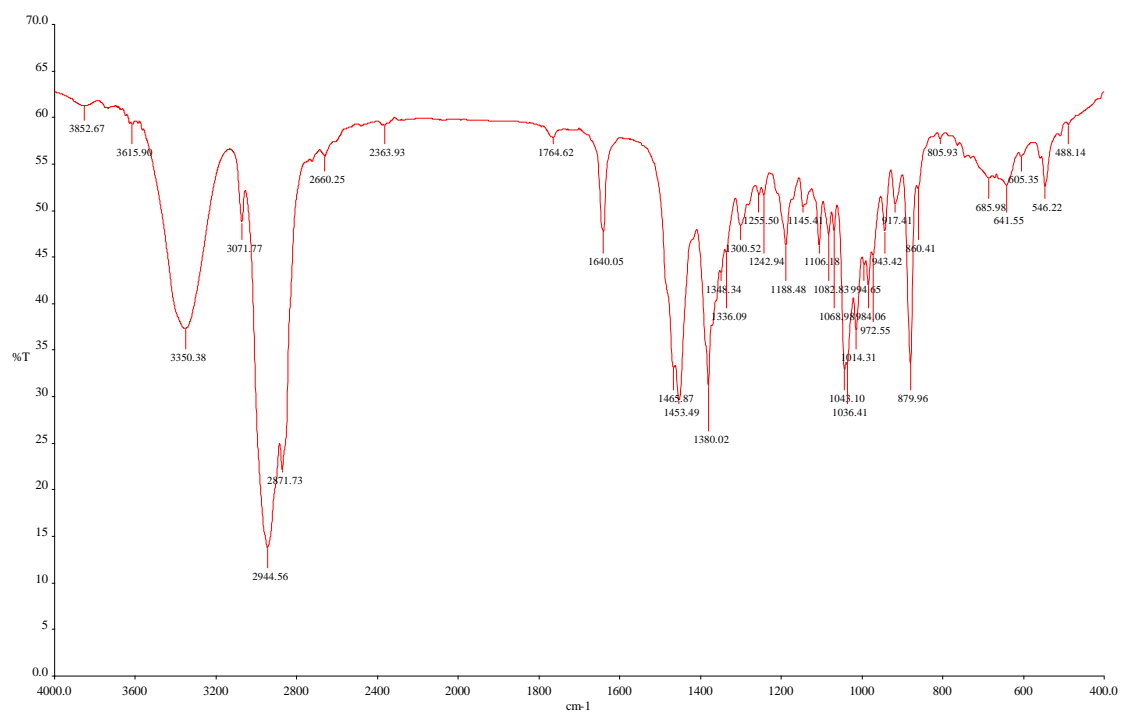


Figure S3-13. IR of compound **3**

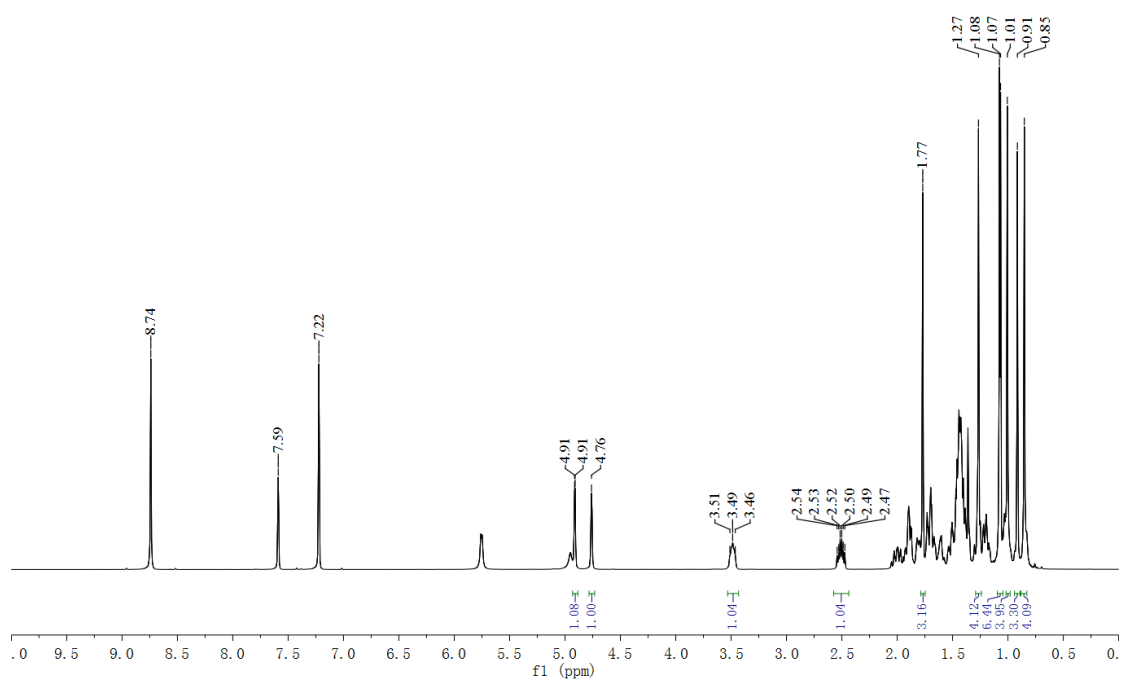


Figure S3-14. ¹H-NMR of compound **3** in C₅D₅N

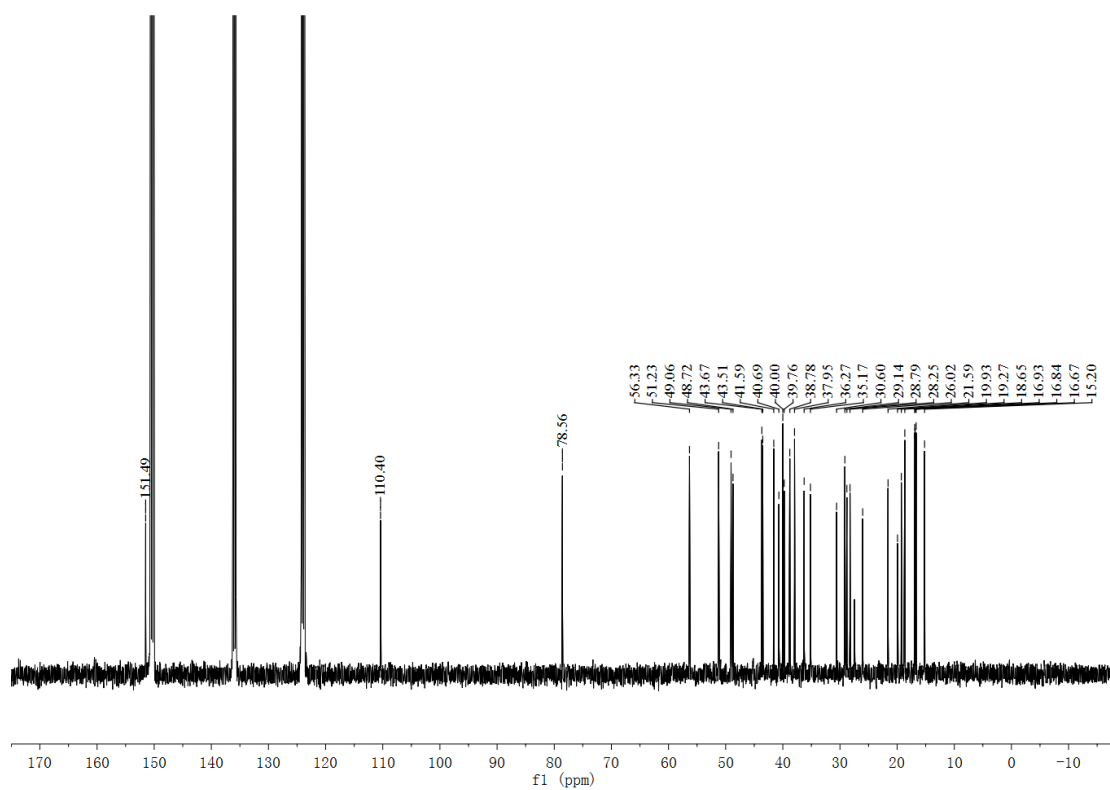


Figure S3-15. ^{13}C -NMR of compound **3** in $\text{C}_5\text{D}_5\text{N}$

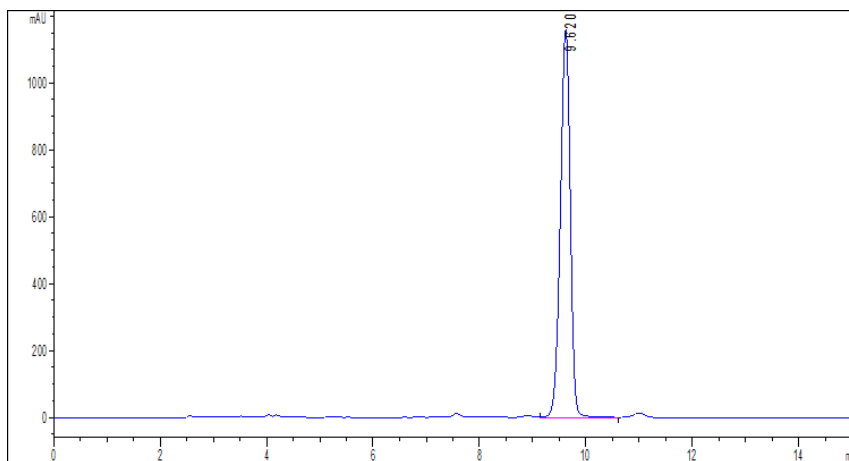


Figure S3-16. Determination of the purity of the compound **5** (Mobile phase: Methanol-0.1% acetic acid aqueous solution=99:1; Detection wavelength: 283nm; Flow rate: 1.3mL/min; Column temperature: 30°C; t_R (95.3% purity) = 9.620 min.)

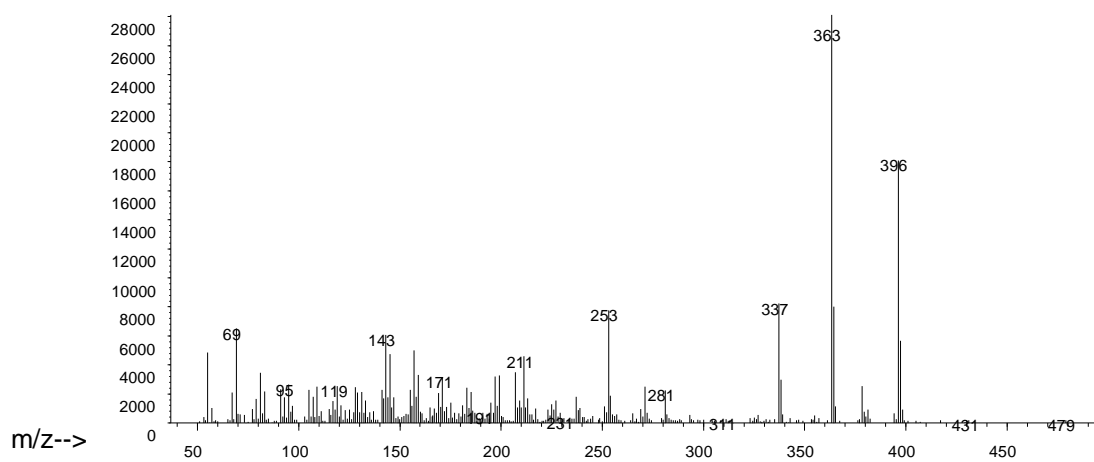


Figure S3-17. MS of compound **5**

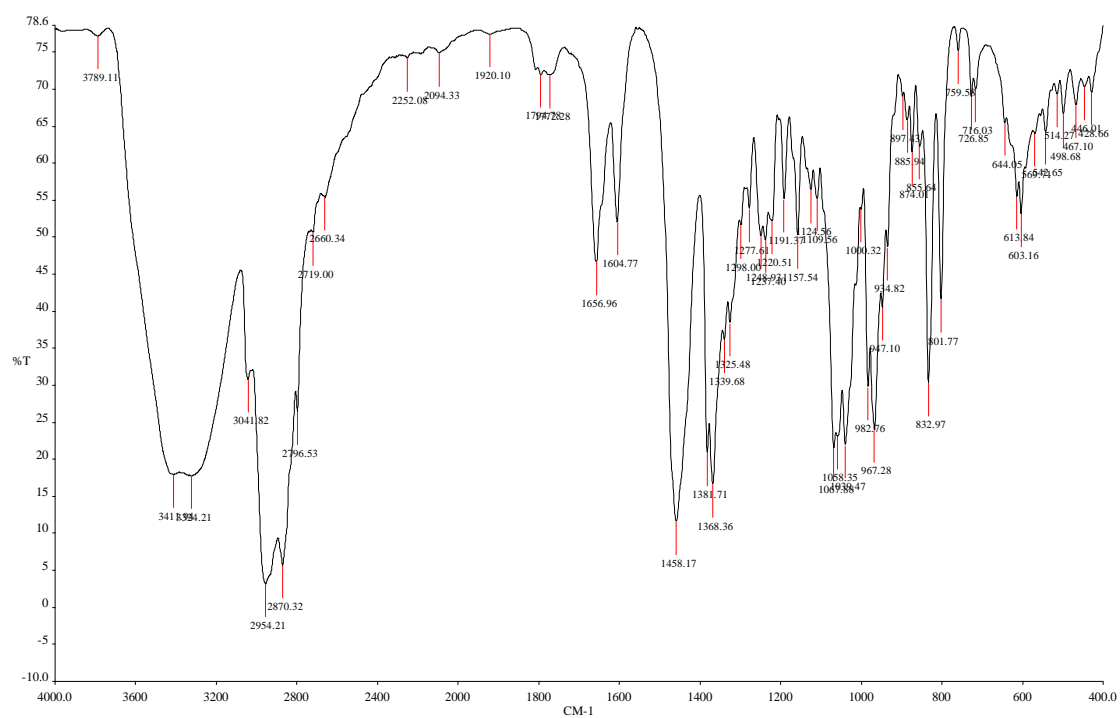


Figure S3-18. IR of compound **5**

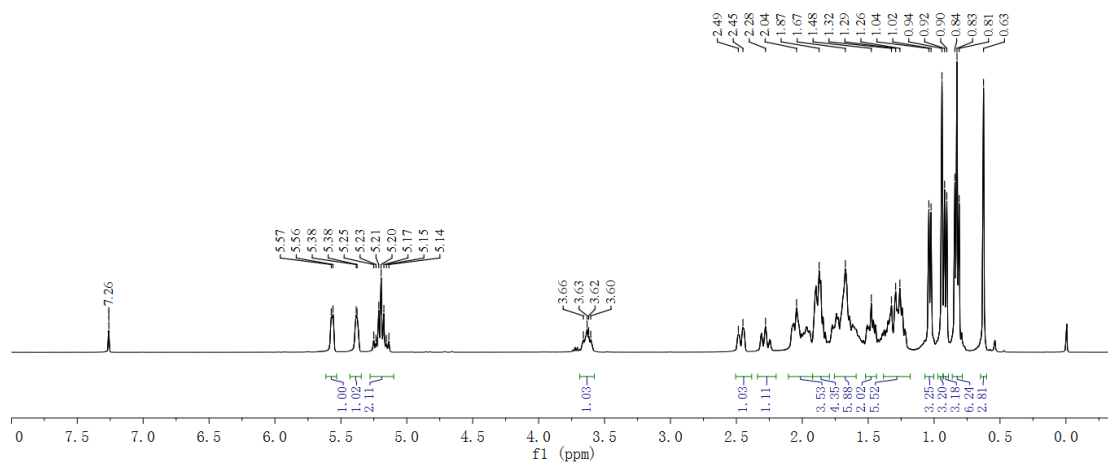


Figure S3-19. ¹H-NMR of compound **5** in CDCl₃

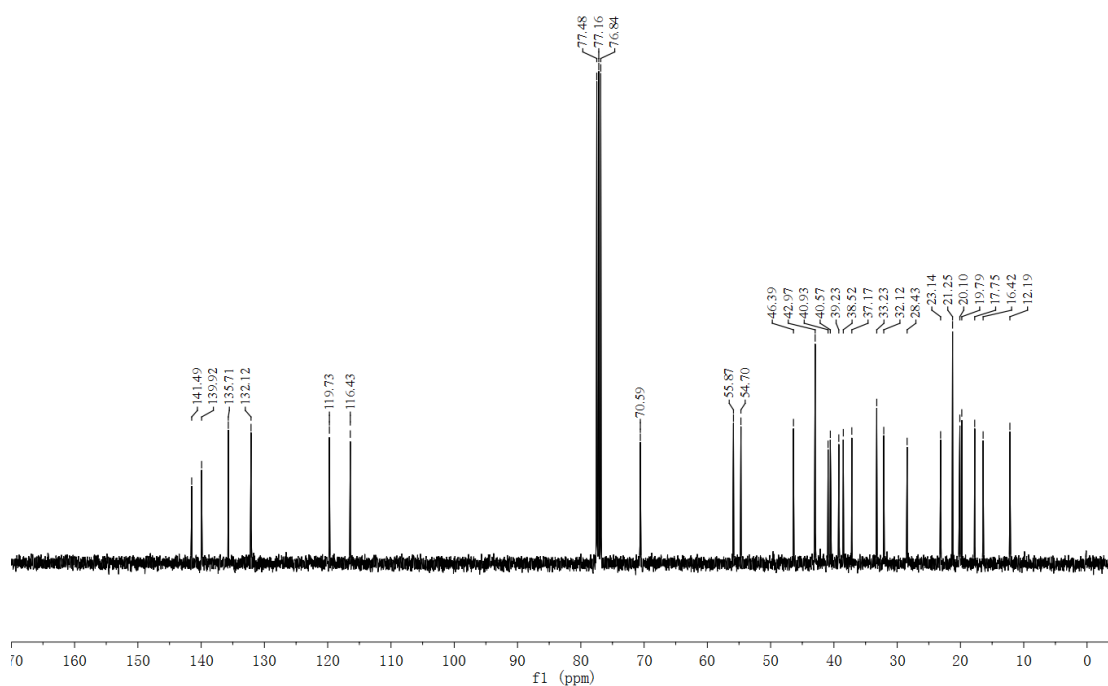


Figure S3-20. ¹³C-NMR of compound **5** in CDCl₃

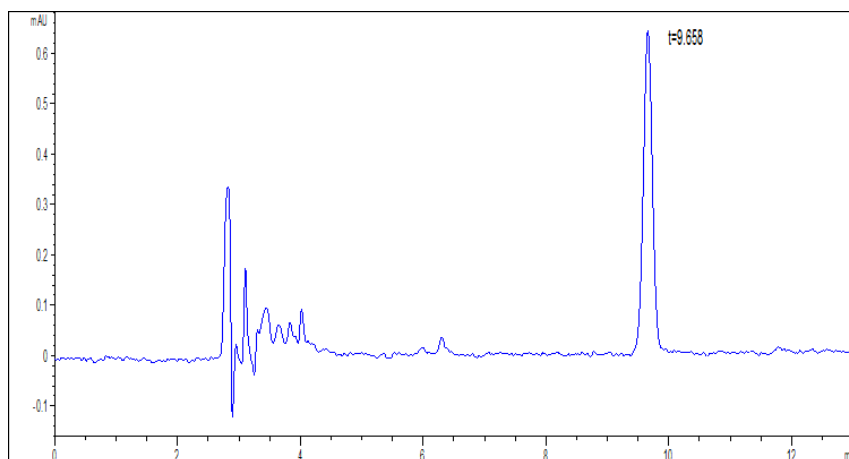


Figure S3-21. Determination of the purity of the compound **7** (Mobile phase: Methanol-0.1% acetic acid aqueous solution=85:15; Detection wavelength: 216 nm; Flow rate: 1.0 mL/min; Column temperature: 30 °C; t_R (92.3% purity) = 9.658 min.)

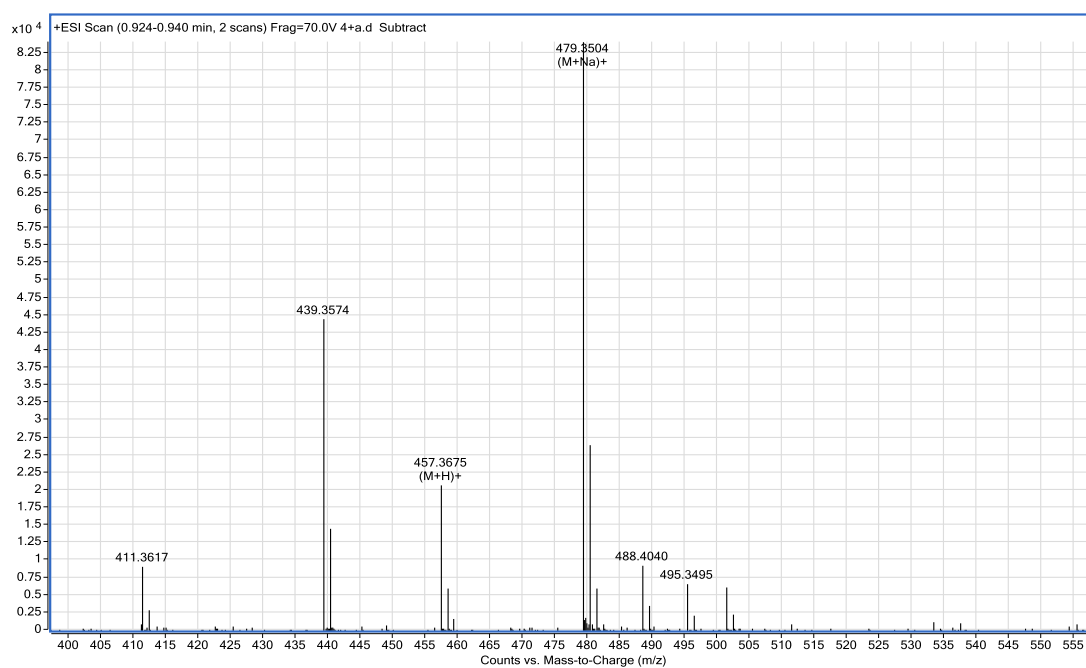


Figure S3-22. MS of compound **7**

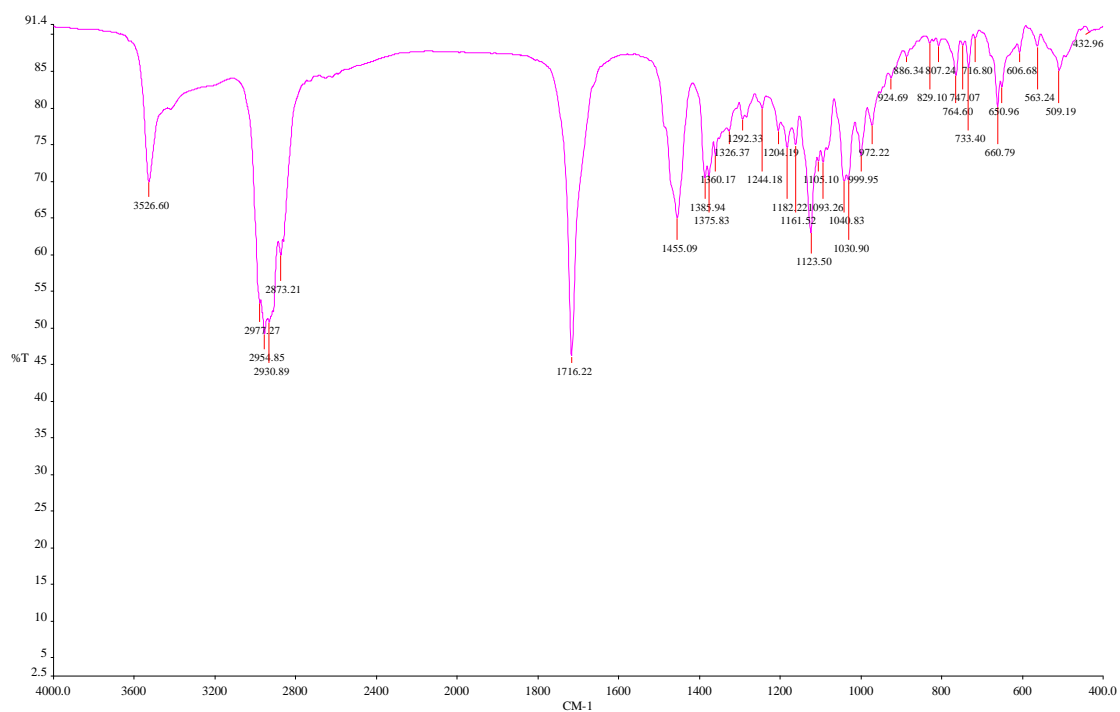


Figure S3-23. IR of compound **7**

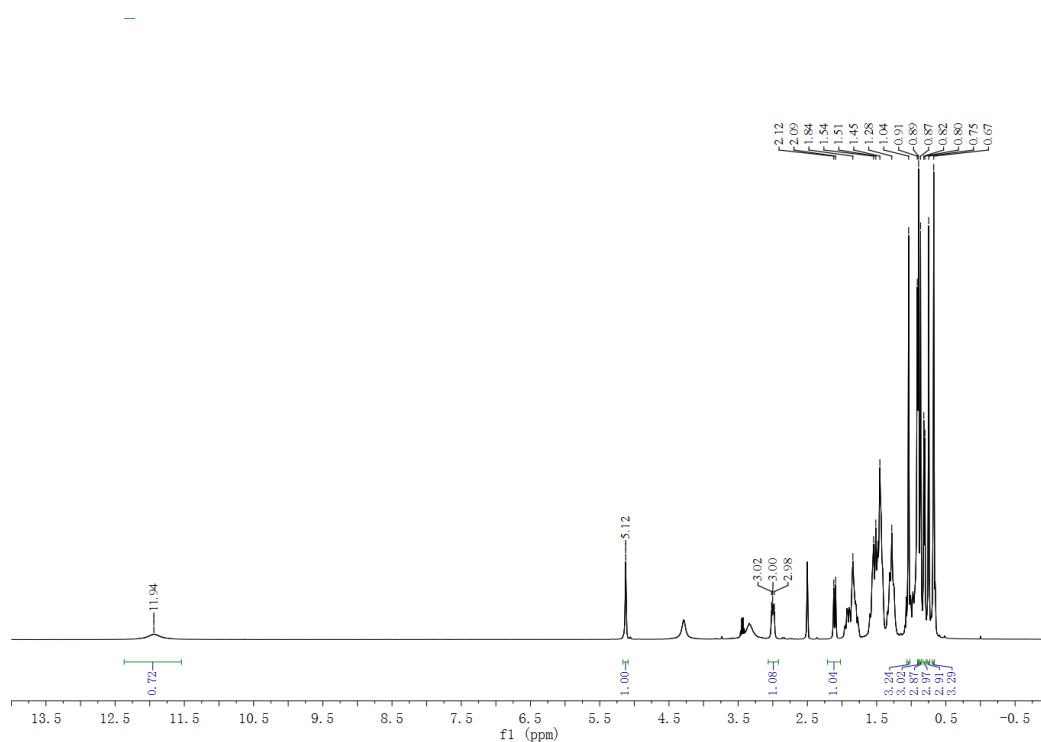


Figure S3-24. ^1H -NMR of compound **7** in $\text{DMSO}-d_6$

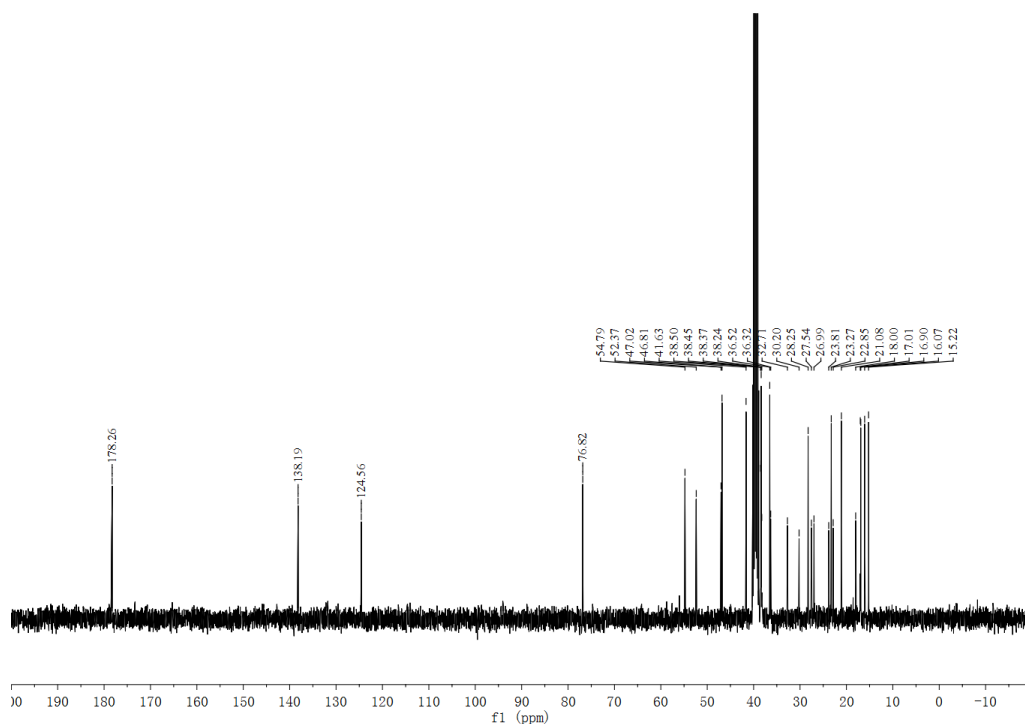


Figure S3-25. ^{13}C -NMR of compound **7** in DMSO- d_6

Figure S3. Spectra of Compound **1-7**

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