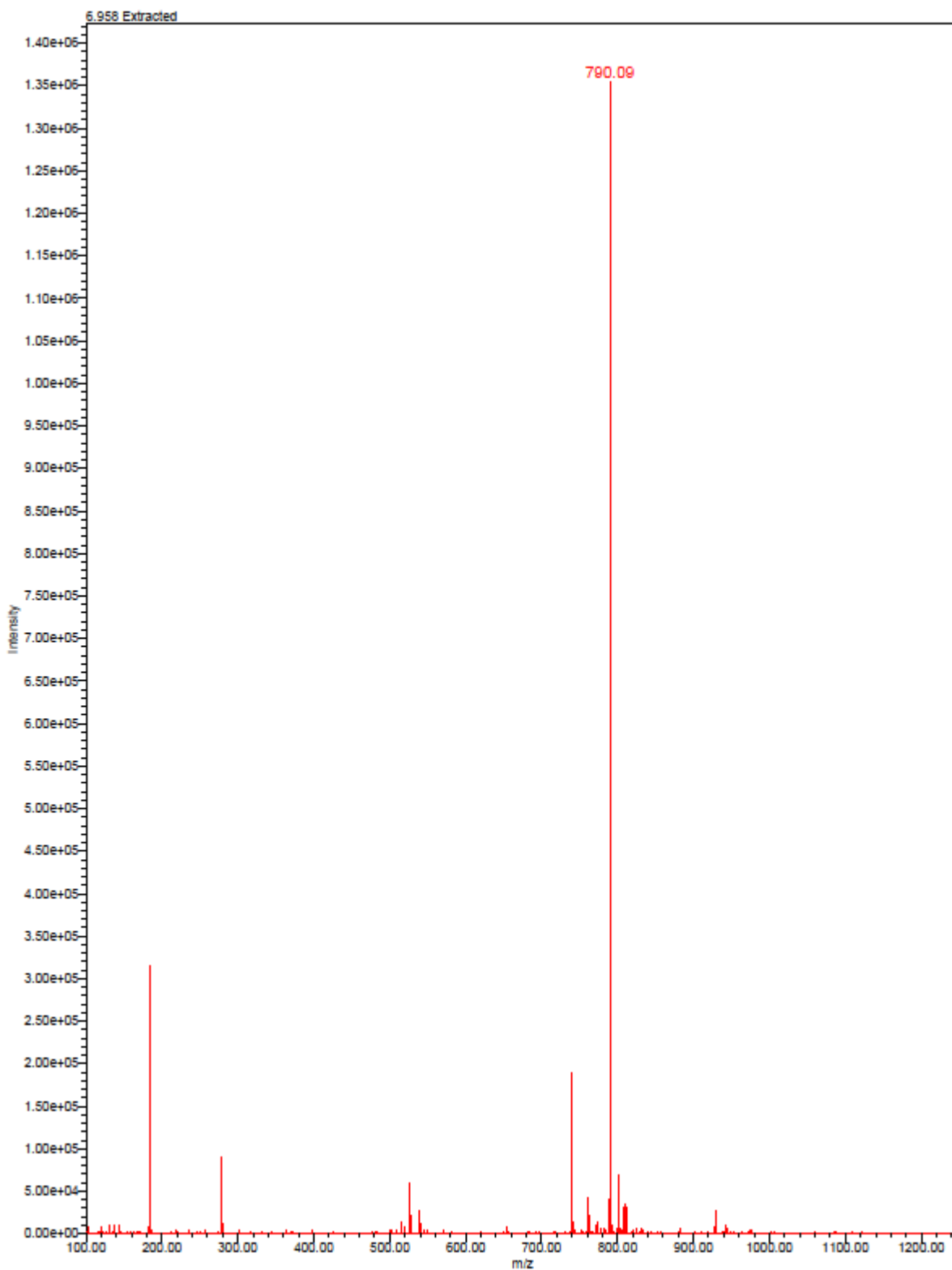


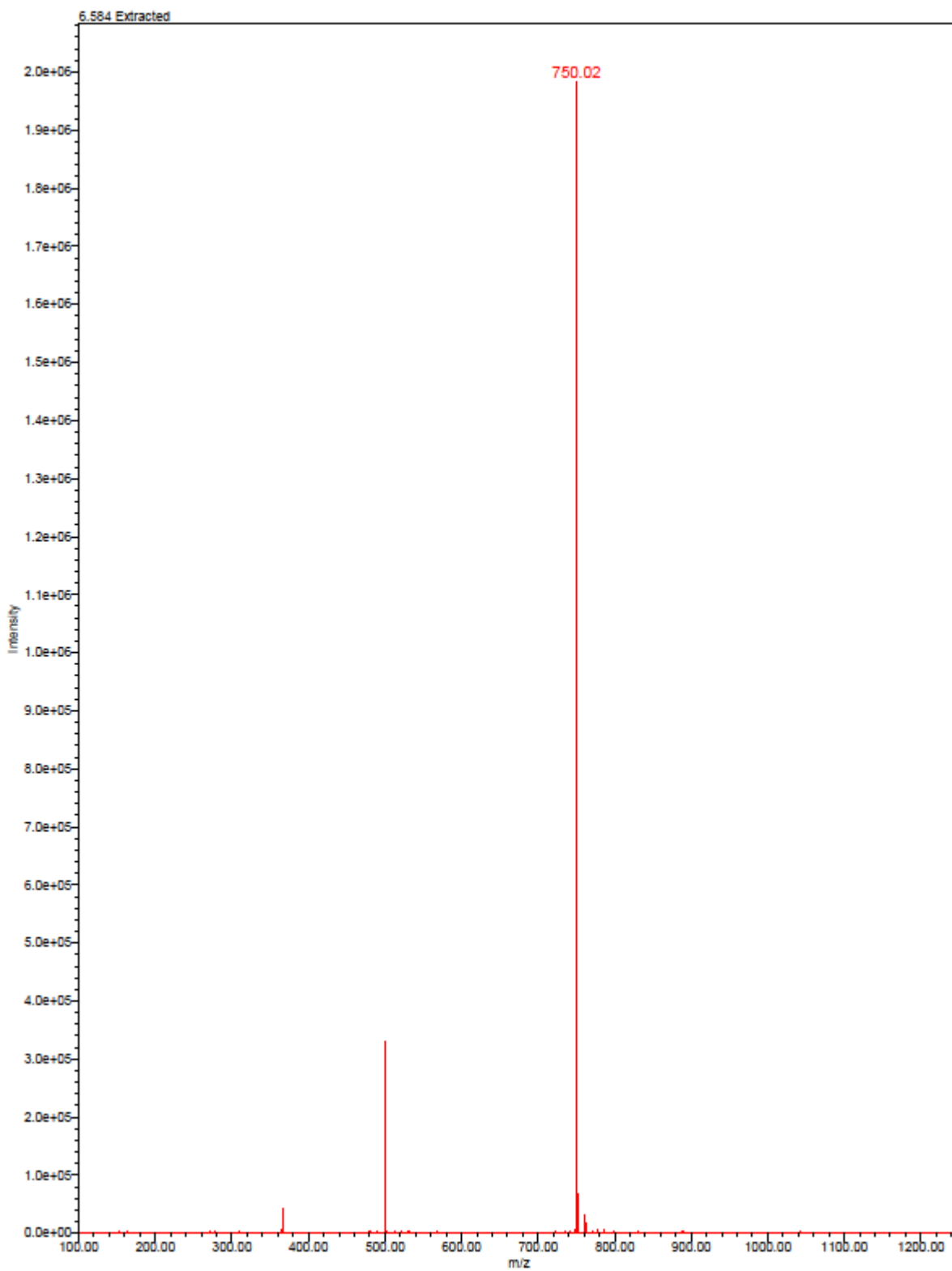
## SUPPLEMENTAL INFORMATION

**Table S1:** HPLC purification conditions and MS characterizations of LW02075 and LW02050.

Compound name	HPLC conditions	Retention time (min)	Yield (%)	Calculated mass (m/z)	Found (m/z)	Purity
LW02075	22% CH <sub>3</sub> CN and 0.1% TFA in H <sub>2</sub> O	28.6	3.4	[M+2H] <sup>2+</sup> 789.9	[M+2H] <sup>2+</sup> 790.1	>99%
LW02050	20% CH <sub>3</sub> CN and 0.1% TFA in H <sub>2</sub> O	15.8	9.5	[M+2H] <sup>2+</sup> 749.9	[M+2H] <sup>2+</sup> 750.0	>99%



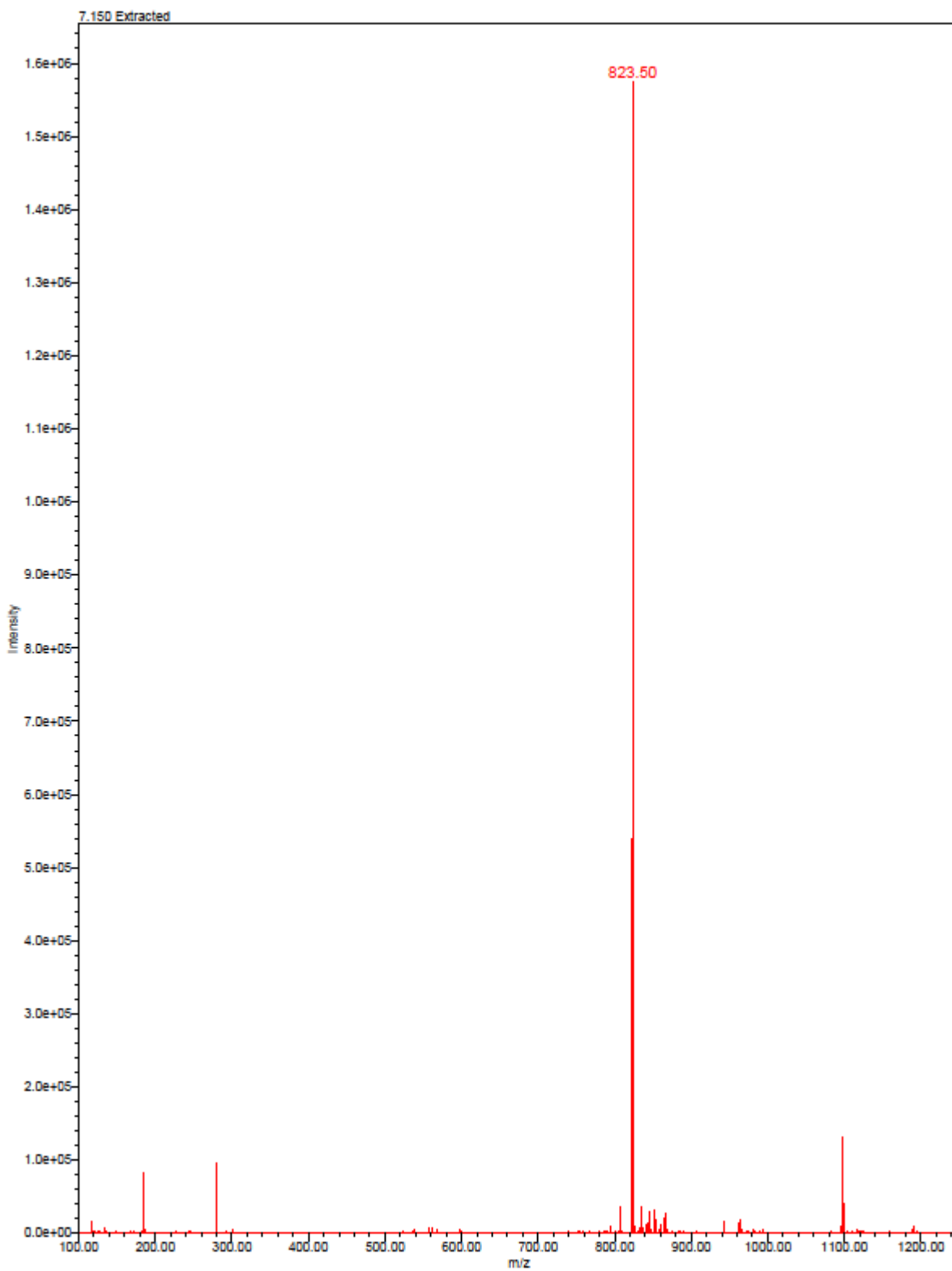
**Figure S1:** The MS spectrum of LW02075: calculated  $[M+2H]^{2+}$  (m/z) 789.9; found 790.1.



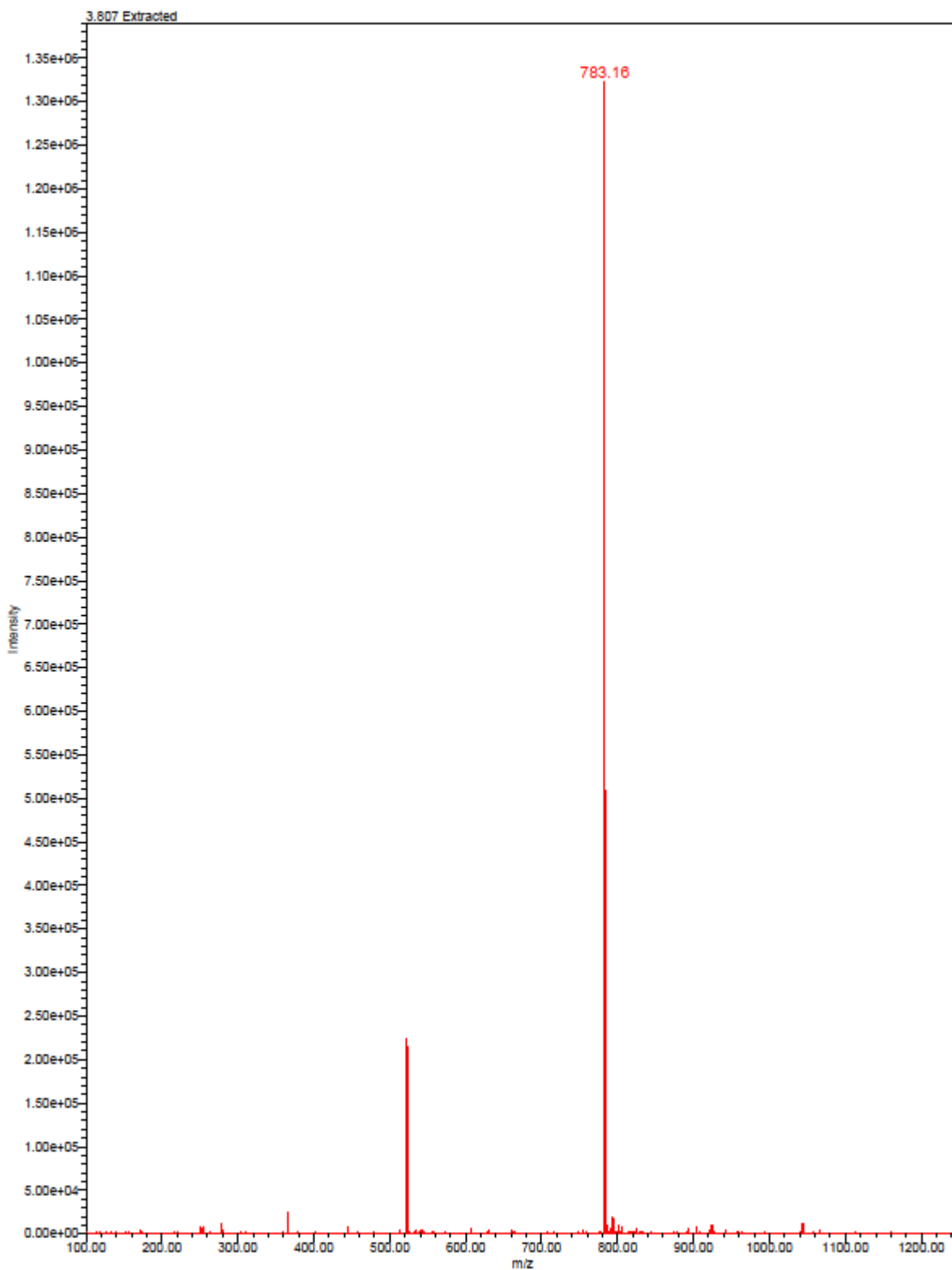
**Figure S2:** The MS spectrum of LW02050: calculated  $[M+2H]^{2+}$  (m/z) 749.9; found 750.0.

**Table S2:** HPLC purification conditions and MS characterizations of Ga-LW02075 and Ga-LW02050.

Compound name	HPLC conditions	Retention time (min)	Yield (%)	Calculated mass (m/z)	Found (m/z)	Purity
Ga-LW02075	25% CH <sub>3</sub> CN and 0.1% TFA in H <sub>2</sub> O	17.3	25	[M+2H] <sup>2+</sup> 822.9	[M+2H] <sup>2+</sup> 823.5	>99%
Ga-LW02050	20% CH <sub>3</sub> CN and 0.1% TFA in H <sub>2</sub> O	22.0	82	[M+2H] <sup>2+</sup> 782.9	[M+2H] <sup>2+</sup> 783.2	>99%



**Figure S3:** The MS spectrum of Ga-LW02075: calculated  $[M+2H]^{2+}$  (m/z) 822.9; found 823.5.



**Figure S4:** The MS spectrum of Ga-LW02050: calculated  $[M+2H]^{2+}$  (m/z) 782.9; found 783.2.

**Table S3:** HPLC conditions for the purification and quality control of [<sup>68</sup>Ga]Ga-LW02075, [<sup>68</sup>Ga]Ga-LW02050, and [<sup>68</sup>Ga]Ga-SB3. FA: formic acid.

Compound name	HPLC conditions		Retention time (min)
[ <sup>68</sup> Ga]Ga-LW02075	Semi-Prep	20% CH <sub>3</sub> CN and 0.1% FA in H <sub>2</sub> O; flow rate 4.5 mL/min	17.0
	QC	21% CH <sub>3</sub> CN and 0.1% FA in H <sub>2</sub> O; flow rate 2.0 mL/min	7.7
[ <sup>68</sup> Ga]Ga-LW02050	Semi-Prep	15% CH <sub>3</sub> CN and 0.1% FA in H <sub>2</sub> O; flow rate 4.5 mL/min	15.8
	QC	18% CH <sub>3</sub> CN and 0.1% FA in H <sub>2</sub> O; flow rate 2 mL/min	5.5
[ <sup>68</sup> Ga]Ga-SB3	Semi-Prep	29% CH <sub>3</sub> CN and 0.1% FA in H <sub>2</sub> O; flow rate 4.5 mL/min	22.5
	QC	32% CH <sub>3</sub> CN and 0.1% FA in H <sub>2</sub> O; flow rate 2 mL/min	11.0

**Table S4:** Biodistribution (mean  $\pm$  SD, n = 4) and uptake ratios of  $^{68}\text{Ga}$ -labeled GRPR-targeted tracers in PC-3 tumor-bearing mice. The mice in the blocked group were co-injected with 100  $\mu\text{g}$  of their nonradioactive Ga-complexed standard (Ga-LW02075 or Ga-LW02050). Statistical analyses were conducted to compare uptake values and uptake ratios between baseline and blocked groups. \*  $p < 0.05$ , \*\*  $p < 0.01$ , \*\*\*  $p < 0.001$ .

Tissue (%ID/g)	$^{68}\text{Ga}$ Ga-SB3	$^{68}\text{Ga}$ Ga-LW02075		$^{68}\text{Ga}$ Ga-LW02050	
	1 h	1 h	1 h blocked	1 h	1 h blocked
Blood	0.50 $\pm$ 0.09	0.76 $\pm$ 0.41	1.15 $\pm$ 0.26	0.57 $\pm$ 0.11	0.34 $\pm$ 0.06*
Fat	0.05 $\pm$ 0.01	0.08 $\pm$ 0.05	0.12 $\pm$ 0.02	0.05 $\pm$ 0.01	0.04 $\pm$ 0.01
Testes	0.13 $\pm$ 0.01	0.17 $\pm$ 0.10	0.30 $\pm$ 0.06	0.17 $\pm$ 0.05	0.13 $\pm$ 0.05
Small intestine	6.03 $\pm$ 1.07	6.72 $\pm$ 1.94	5.62 $\pm$ 1.86	0.43 $\pm$ 0.04	0.26 $\pm$ 0.09*
Large intestine	2.20 $\pm$ 0.89	1.71 $\pm$ 0.47	0.40 $\pm$ 0.17*	0.34 $\pm$ 0.15	0.19 $\pm$ 0.17
Spleen	0.33 $\pm$ 0.17	0.26 $\pm$ 0.13	0.35 $\pm$ 0.12	0.25 $\pm$ 0.04	0.12 $\pm$ 0.02**
<b>Pancreas</b>	<b>37.3 <math>\pm</math> 6.90</b>	<b>17.8 <math>\pm</math> 5.24</b>	<b>1.55 <math>\pm</math> 0.83***</b>	<b>0.53 <math>\pm</math> 0.11</b>	<b>0.10 <math>\pm</math> 0.03***</b>
Stomach	1.42 $\pm$ 0.57	1.23 $\pm$ 0.22	0.73 $\pm$ 0.29*	0.34 $\pm$ 0.14	0.04 $\pm$ 0.02**
Liver	2.10 $\pm$ 0.45	4.14 $\pm$ 1.50	5.92 $\pm$ 1.69	0.44 $\pm$ 0.05	0.29 $\pm$ 0.20
Adrenal glands	2.07 $\pm$ 0.66	4.42 $\pm$ 2.41	1.02 $\pm$ 0.59*	0.43 $\pm$ 0.15	0.05 $\pm$ 0.05**
Kidneys	2.26 $\pm$ 0.26	3.11 $\pm$ 1.18	4.98 $\pm$ 1.59	2.88 $\pm$ 0.68	1.95 $\pm$ 0.44
Heart	0.16 $\pm$ 0.02	0.24 $\pm$ 0.13	0.34 $\pm$ 0.04	0.19 $\pm$ 0.03	0.10 $\pm$ 0.02**
Lungs	1.18 $\pm$ 0.23	0.75 $\pm$ 0.41	0.94 $\pm$ 0.17	0.55 $\pm$ 0.06	0.33 $\pm$ 0.09**
<b>PC-3 tumor</b>	<b>6.98 <math>\pm</math> 1.36</b>	<b>3.97 <math>\pm</math> 1.71</b>	<b>1.83 <math>\pm</math> 0.22*</b>	<b>5.38 <math>\pm</math> 1.00</b>	<b>0.42 <math>\pm</math> 0.08***</b>
Bone	0.12 $\pm$ 0.01	0.20 $\pm$ 0.20	0.21 $\pm$ 0.07	0.14 $\pm$ 0.06	0.03 $\pm$ 0.03*
Muscle	0.18 $\pm$ 0.10	0.21 $\pm$ 0.18	0.27 $\pm$ 0.14	0.13 $\pm$ 0.02	0.06 $\pm$ 0.02**
Brain	0.04 $\pm$ 0.01	0.03 $\pm$ 0.01	0.03 $\pm$ 0.01	0.03 $\pm$ 0.01	0.01 $\pm$ 0.00**
Tumor/bone	61.0 $\pm$ 15.5	30.6 $\pm$ 18.9	9.30 $\pm$ 2.39	43.6 $\pm$ 22.6	27.1 $\pm$ 18.1
Tumor/muscle	48.2 $\pm$ 24.9	24.8 $\pm$ 9.99	7.85 $\pm$ 3.13*	44.4 $\pm$ 14.1	8.28 $\pm$ 2.70**
Tumor/blood	14.4 $\pm$ 3.88	5.51 $\pm$ 1.49	1.65 $\pm$ 0.40**	9.68 $\pm$ 2.84	1.23 $\pm$ 0.14**
Tumor/kidney	3.09 $\pm$ 0.51	1.28 $\pm$ 0.33	0.39 $\pm$ 0.09**	1.92 $\pm$ 0.41	0.22 $\pm$ 0.05***
Tumor/pancreas	0.19 $\pm$ 0.03	0.22 $\pm$ 0.05	1.46 $\pm$ 0.76*	10.7 $\pm$ 4.17	4.30 $\pm$ 0.83*