

Figure S1. <sup>1</sup>H NMR spectrum of HPG polymer

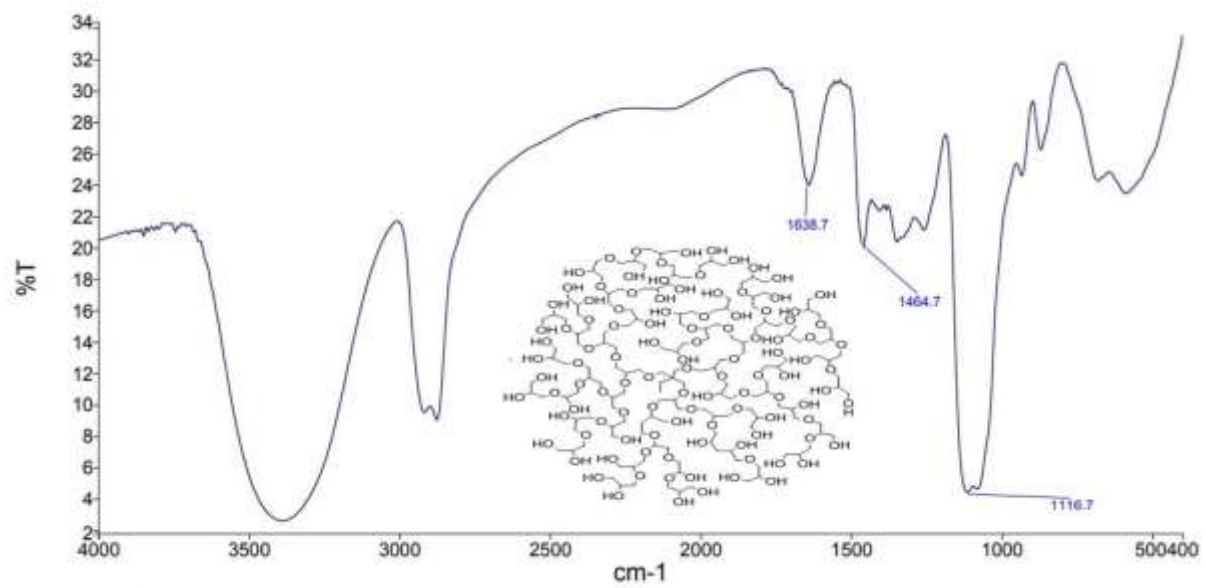


Figure S2. IR spectrum of HPG polymer

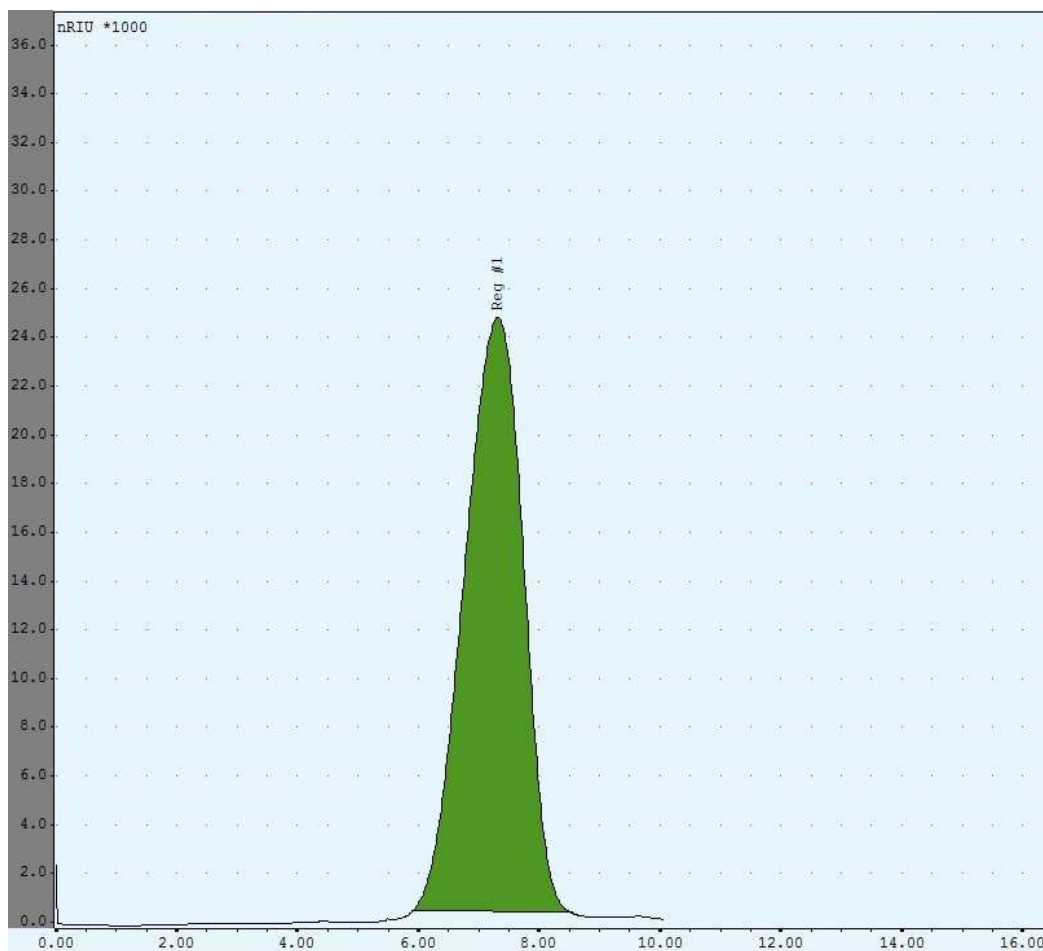


Figure S3. GPC chromatogram of HPG polymer

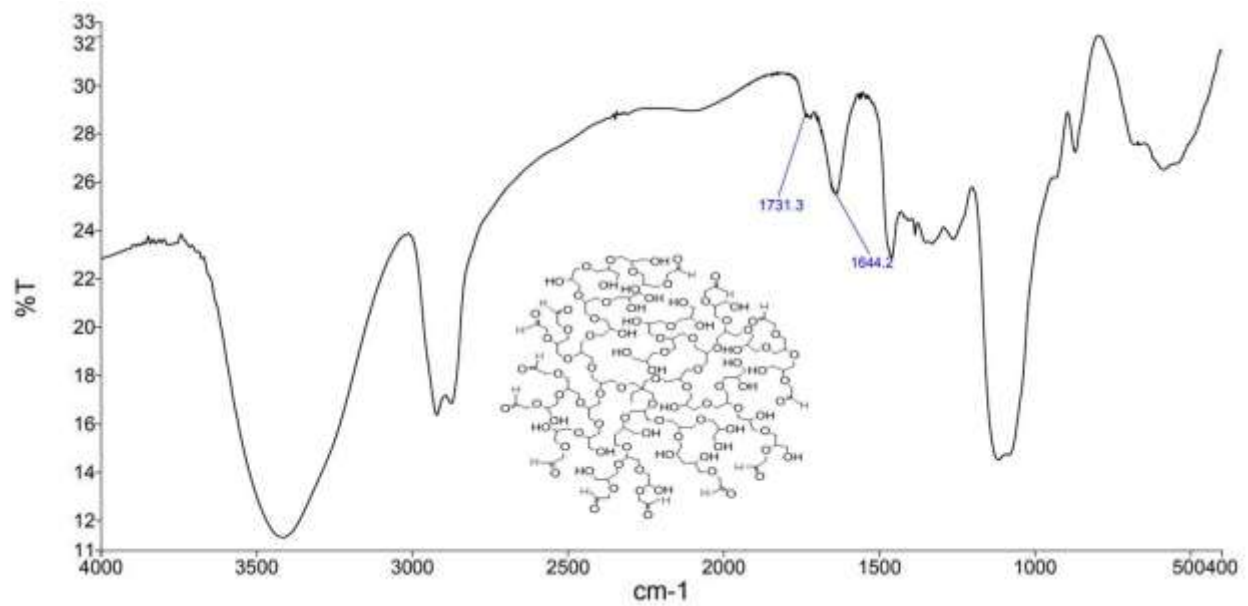


Figure S4. IR spectrum of oxidized polymer

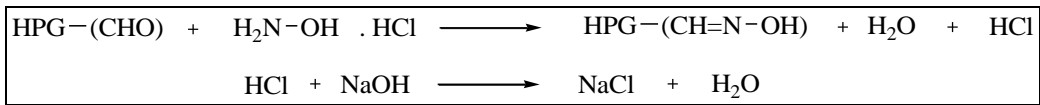


Figure S5. A schematic of the Hydroxylamine hydrochloride test. To carry out this test, a 0.25 M aqueous solution of hydroxylamine hydrochloride was prepared, and its pH was adjusted to 4.5 using concentrated sodium hydroxide. A precisely measured amount of the oxidized polymer (10 mg) was then added to 25 mL of the prepared solution. The mixture was allowed to react at room temperature for 2 hours (Figure 3-3). During the reaction, a pH decrease from 4.5 to 3.78 was observed, indicating the formation of oxime and the release of hydrochloric acid. The liberated acid was neutralized by titration with 2.5 mL of 0.01 M sodium hydroxide to restore the pH to its initial value. The titration process was monitored using a pH meter. The aldehyde content in the sample was then calculated to be 2.5  $\mu\text{mol}/\text{mg}$  using the following formula:

$$\frac{\text{The volume of NaOH consumed} \times [\text{NaOH}] \times 10^{-3}}{\text{Weight of the sample (g)}} = \frac{\text{Mole of aldehyde}}{\text{g of sample}}$$

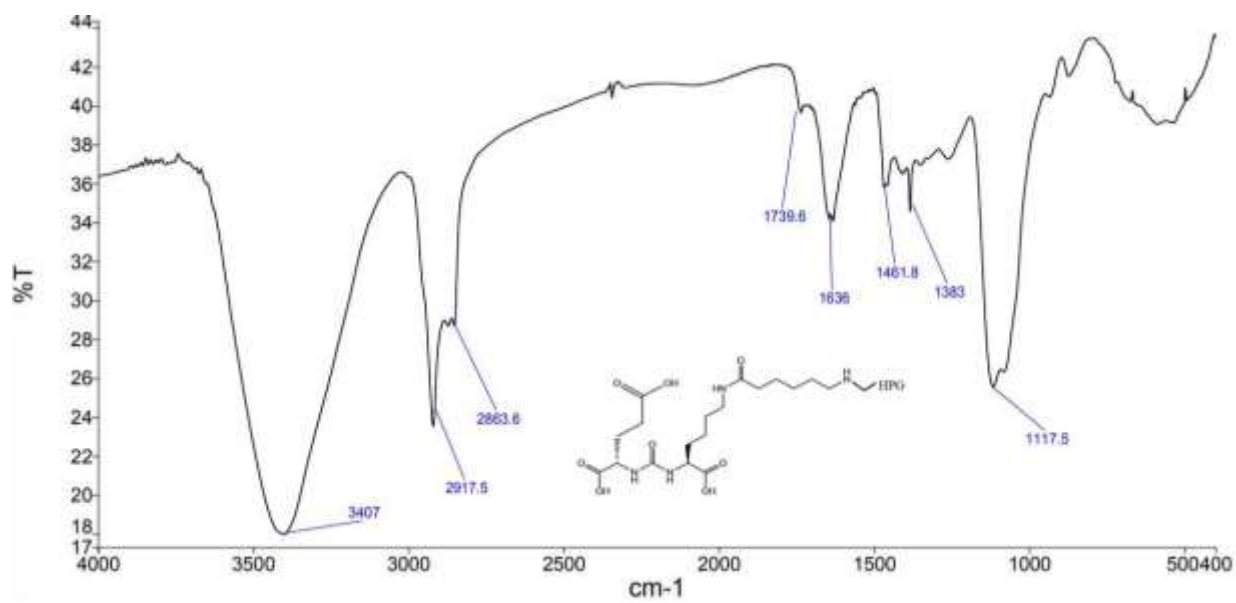


Figure S6. IR spectrum of HPG-PSMA compound

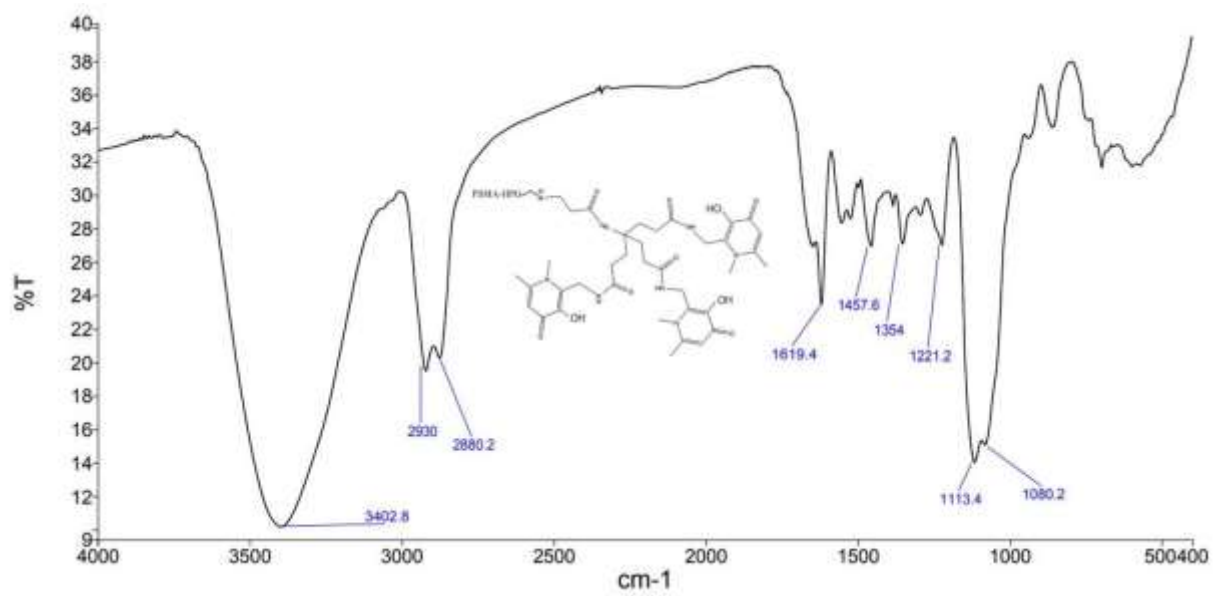


Figure S7. IR spectrum of the THP-HPG-PSMA compound

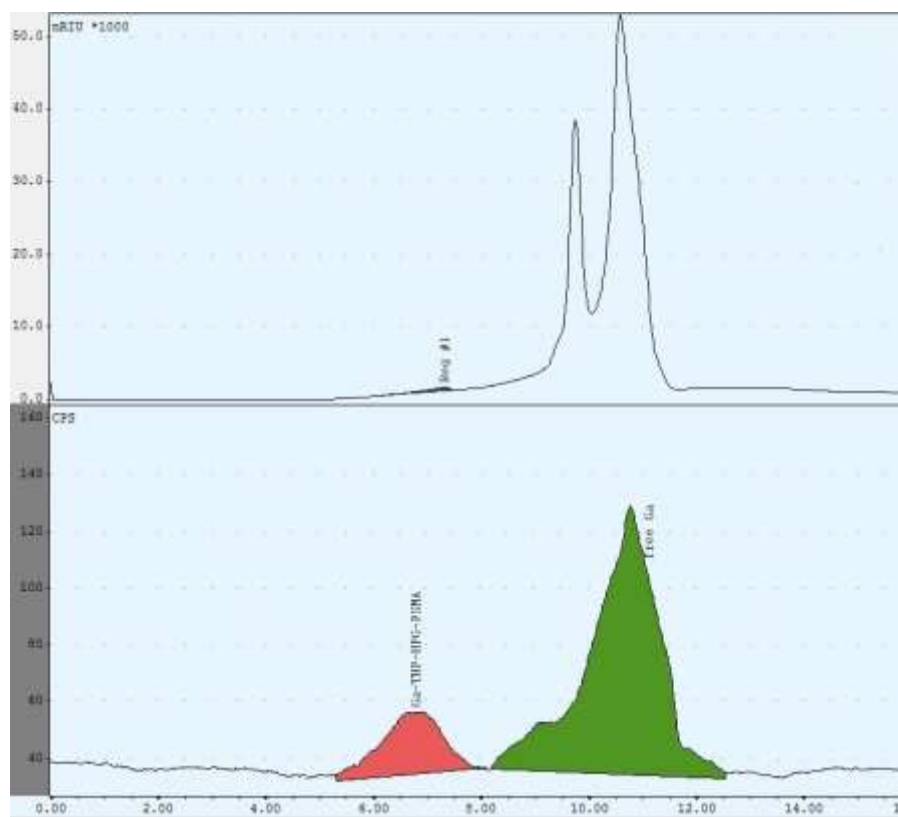


Figure S8. GPC chromatogram of [<sup>66</sup>Ga]Ga-THP-HPG-PSMA in presence of excess non-radioactive Ga<sup>3+</sup>

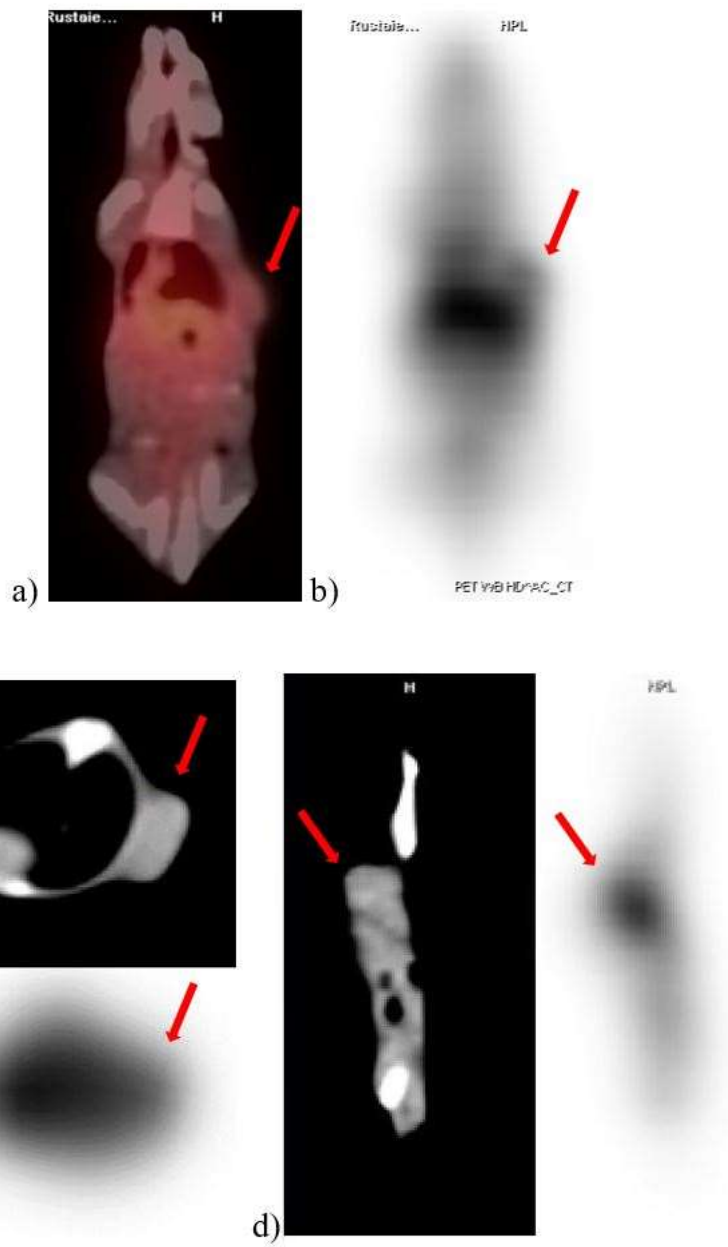


Figure S9. Imaging results 30 minutes after intravenous injection of 100–150  $\mu\text{Ci}$  of the  $^{68}\text{Ga}$ -labeled compound in a prostate tumor-bearing mouse with PSMA expression in the right scapular region. a) Coronal PET-CT view, b) Coronal PET view, c) Axial CT (top) and PET (bottom) views, and d) Sagittal CT (left) and PET (right) views. Tumor sites are indicated by arrows

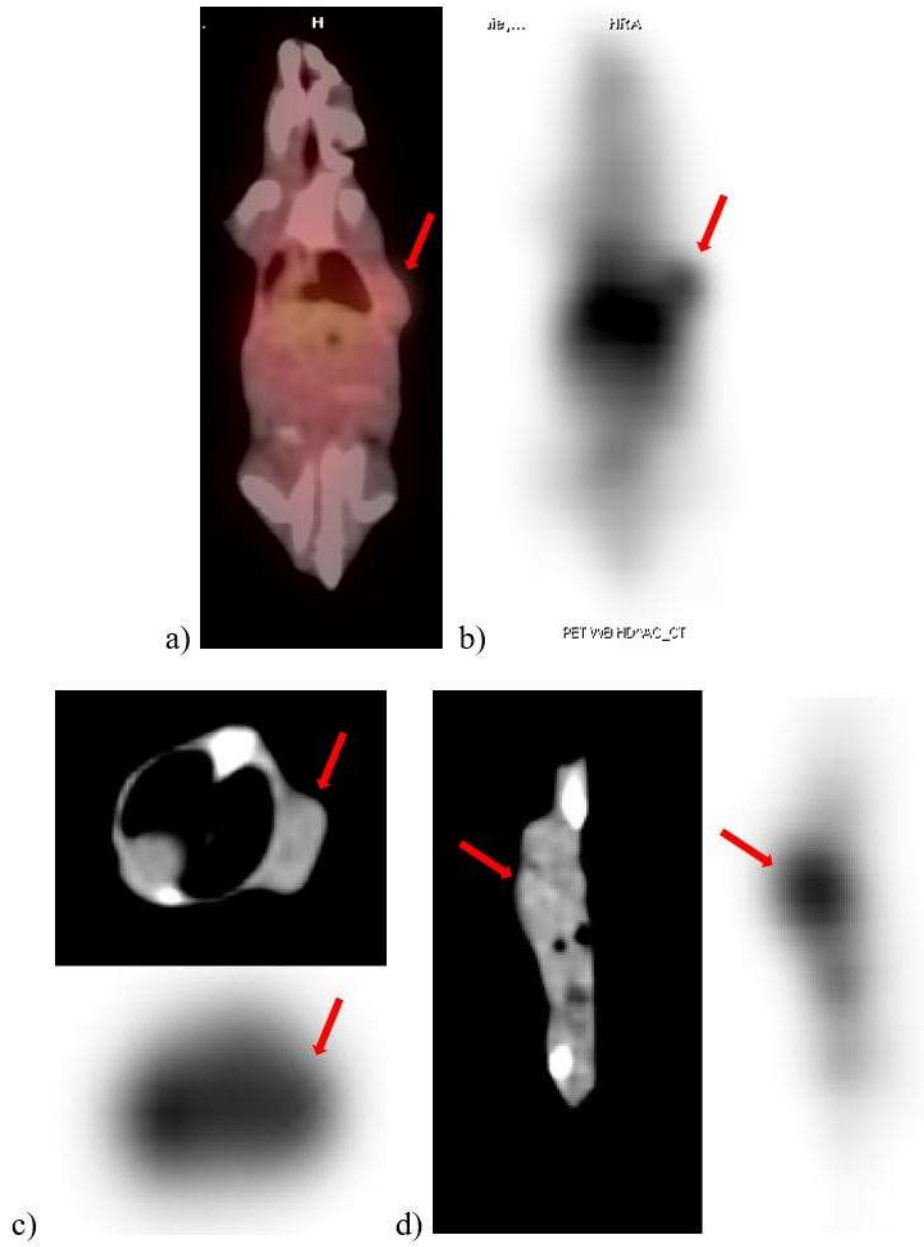


Figure S10. PET-CT imaging 60 minutes after intravenous injection of 100–150  $\mu\text{Ci}$  of the  $^{68}\text{Ga}$ -labeled compound in a prostate tumor-bearing mouse with PSMA expression in the right scapular region. a) Coronal PET-CT view, b) Coronal PET view, c) Axial CT (top) and PET (bottom) views, and d) Sagittal CT (left) and PET (right) views. The tumor site is marked with an arrow

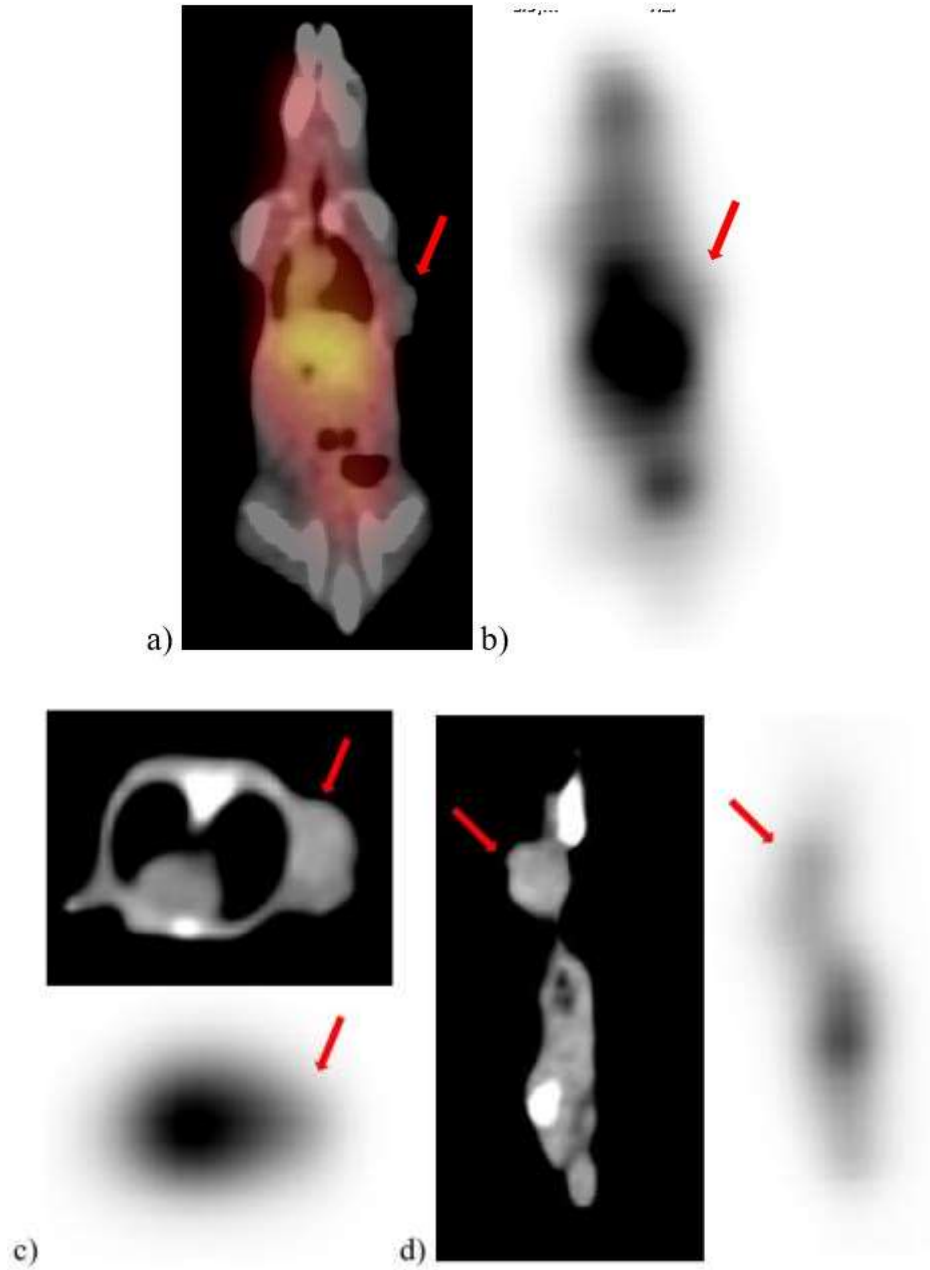


Figure S11. PET-CT images from the blocking study, captured 30 minutes after administration of 100  $\mu\text{Ci}$  of the  $^{68}\text{Ga}$ -labeled compound in a prostate tumor-bearing mouse with PSMA expression in the right scapular region. a) Coronal PET-CT view, b) Coronal PET view, c) Axial CT (top) and PET (bottom) views, and d) Sagittal CT (left) and PET (right) views. The tumor site is highlighted with an arrow