Supporting Information:

A DNA nanodevice that loads and releases a cargo with hemoglobin-like allosteric control and cooperativity

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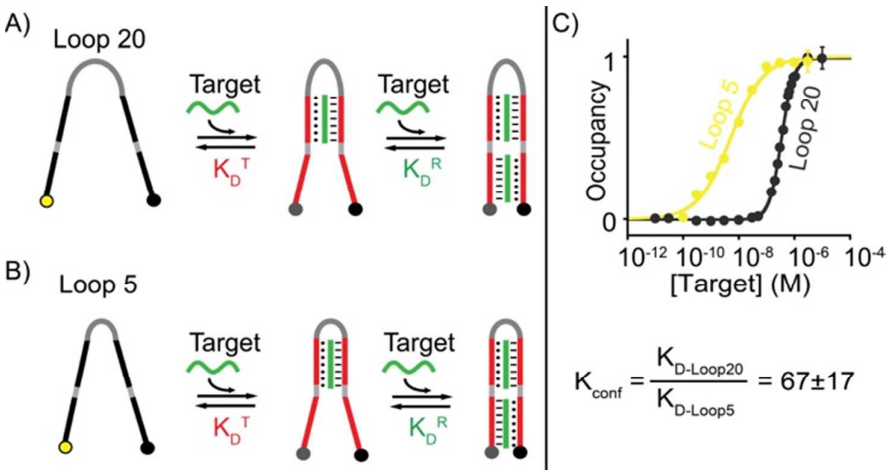

Figure S1. Measuring K_{conf}. We estimated K_{conf}, the conformational equilibrium constant between the two states, from the ratio of the affinity constants of the cooperative, two-site nanodevice (A) to that of a nanodevice where the 20-base loop is substituted with a 5-base loop (B). The short loop minimizes the entropic cost associated with the closure of the nanodevice upon the first binding event, enhancing the population of the high affinity state. Using the affinity constant of this system (black curve, 5.1±1.1 nM) we estimated a K_{conf} of 67±17. The binding curves shown here were performed in 10 mM Tris-HCl, 5 mM $MgCl_2$ at pH 7.5, 35°C at a concentration of nanodevice of 3 nM and adding increasing concentrations of ligand DNA. Here and in the following figures, in order to better compare the different nanodevices, the fluorescence signals have been normalized and the relative occupancy used to create binding curves.

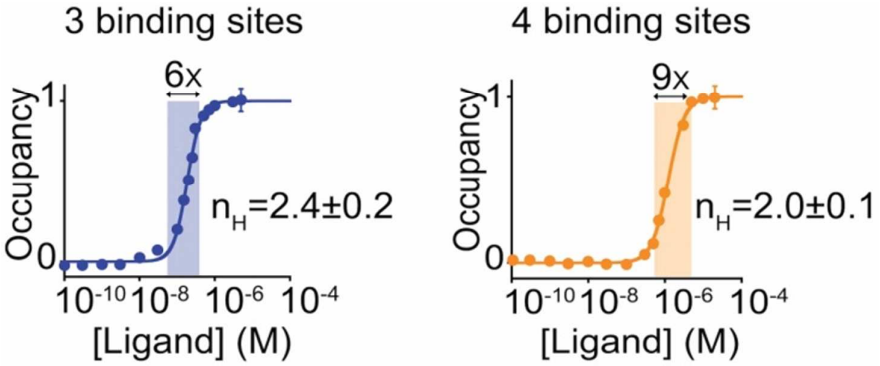

Figure S2. Cooperative nanodevices containing 3 or 4 binding sites. Our cooperative nanodevice architecture can be extended to include 3 or 4 binding sites. The maximum Hill coefficient we observe, however, is seen with the 3-site nanodevice rather than the 4-site device. We presume this occurs due to non-specific interactions among different binding sites.

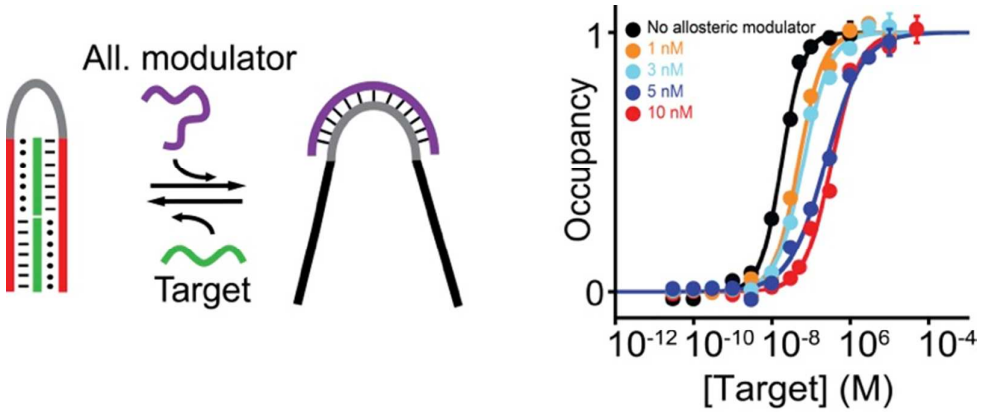

Figure S3. Allosteric control of the hemoglobin-like DNA nanodevice. The affinity of our DNA nanodevice can be finely regulated with an allosteric effector. The binding curves shown here were performed in 10 mM Tris-HCl, 5 mM $MgCl_2$ at pH 7.5, 35°C at a concentration of nanodevice of 3 nM and adding increasing concentrations of ligand DNA in the presence of the indicated concentrations of the allosteric modulator.

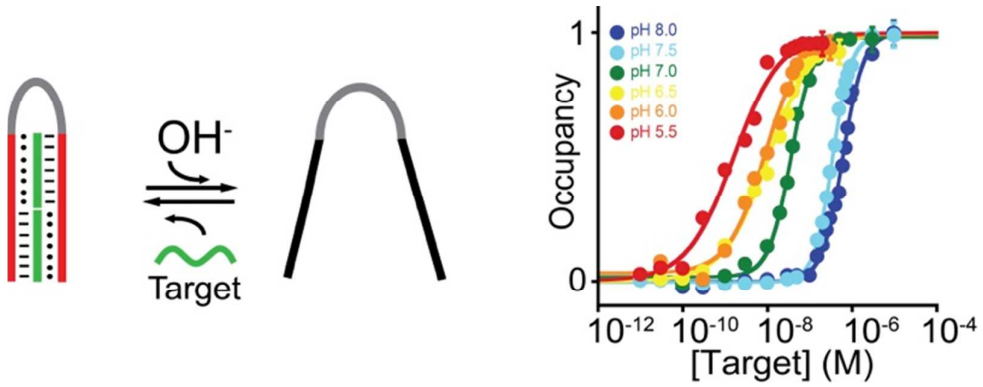

Figure S4. pH effect of the hemoglobin-like DNA nanodevice. The affinity of our DNA nanodevice is pH-dependent. The binding curves shown here were performed in 10 mM Tris-HCl, 5 mM $MgCl_2$ at 35°C at a concentration of nanodevice of 3 nM and adding increasing concentrations of ligand DNA at the indicated pH values.

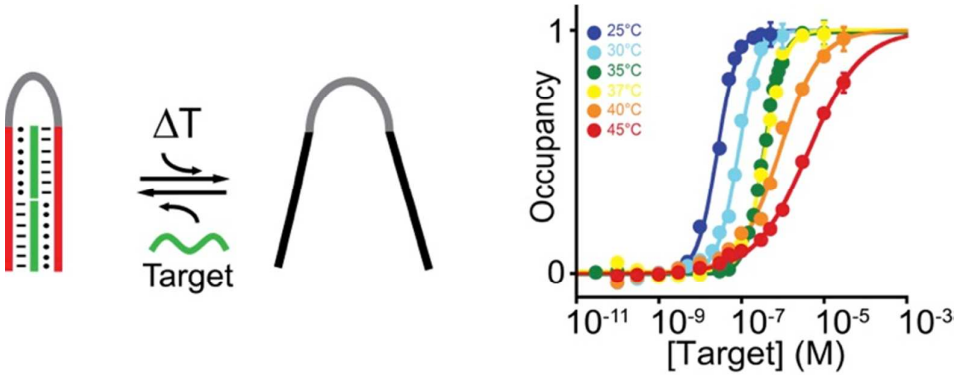

Figure S5. Temperature effect of the hemoglobin-like DNA nanodevice. The affinity of our DNA nanodevice is temperature dependent. The binding curves shown here were performed in 10 mM Tris-HCl, 5 mM $MgCl_2$ at pH 7.5 at a concentration of nanodevice of 3 nM and adding increasing concentrations of ligand DNA at the indicated different temperatures.