Supplemental Figure S1. The superimposed 600 MHz $^1$H-$^{15}$N HSQC spectra of $^{15}$N-E78Q-BcX (initially 0.69 mM) titrated with X4 at 25 °C (blue peaks changing to yellow with X4 concentrations of 0, 1.1, 3.0, 7.4, and 21.6 mM, respectively). Peaks corresponding to backbone $^1$H-$^{15}$N, Asn/Gln sidechain $^1$H$^\delta/\epsilon$-$^{15}$N$^\delta/\epsilon$ (connected by horizontal lines) and Trp $^1$H$^\epsilon$-$^{15}$N$^\epsilon$ groups (lower case w) that undergo shift or intensity perturbations upon X4 binding are labeled. Mainchain amides from the SBS (bold labels) show monophasic titrations in the fast exchange limit. Due to a larger $\Delta \omega$, some exchange broadening is observed for sidechain amides within the SBS, such as N54. Residues from the AS (italics, underlined) show monophasic titrations in the intermediate-fast exchange regime.
Supplemental Figure S2. Mainchain $^1$H$_N$ and $^{15}$N chemical shift perturbations $\Delta \delta$ (equation 1) of inactive E78Q-BcX upon titration with X2, X4, X6, and PNP-X12, as well as fractionated soluble birchwood xylan (DP ~ 180), and of WT-BcX with xylose (X1, to which it has no activity). The $\Delta \delta$ values are taken from titration points of 0.5 mM protein without and with 1940 mM X1, 128 mM PNP-X2, 7 mM X4, 7 mM X6, and 5.5 mg/mL xylan. Due to limited quantities, only 0.076 mM X12, leading to ~ 50% saturation, was utilized. Note that PNP-X12 and xylan bind the AS in the slow exchange regime, produces changes in amide signal intensity rather than chemical shift. The secondary structure elements of BcX, defined by Promotif $^7_1$, are identified by arrows ($\beta$-strands, sheet A black, sheet B white) and a coil ($\alpha$-helix).
Supplemental Figure S3.

Binding of insoluble birchwood xylan and avicel by E78Q-BcX (●) and AAA-E78Q-BcX (○) as measured with a sedimentation assay. Panel (a) shows the binding isotherms for xylan, whereas panels (b) and (c) are the linearized plots from which $K_{dR}$ values for xylan and avicel were fit (Table 1). $[P]_F$ are $[P]_B$ the concentrations of unbound and bound protein, respectively, determined after sedimentation.