Bulfinch made plans for a spacious library room in the center of the west front of the building. The library was functional, making space for work and still expanding on Latrobe’s initial designs for a reading room. It was occupied c.1824, however a year later a fire caused a great deal of damage. The Capitol Building was completed in 1826 and Bulfinch spent the last years of his post finishing the landscaping, designing the piers and gates and executing the already conceived plans for four lodges at the perimeter of the grounds.

While in Washington, Bulfinch missed attending Unitarian services and with the help of John Quincy Adams and John C. Calhoun, he raised money for and designed a Unitarian Church. The church was located on the corner of 6th and D streets and was dedicated on June 9, 1822. It was demolished in 1900 although a small book of plans by Bulfinch remained.

The building was constructed of brick and then painted white, with a tower above the entrance surrounded by a portico and a pediment of the doric order. There is some evidence that the plan for this church was used for another church in Peterborough, New Hampshire which was built in 1825. Bulfinch attended the Unitarian Church in Washington with his family for eight years and later his son, Rev. Stephen Greenleaf Bulfinch became its minister for a time.

Bulfinch was commissioned by President Andrew Jackson to build a Federal Penitentiary across the Potomac on Greenleaf's Point in 1826. The plan was based on the Auburn Principle, and the style suggested Bulfinch’s late interest in Gothic Revival architecture. Despite Bulfinch’s title as Architect of the Capitol payment for the commission of the penitentiary was still uncertain and had required Bulfinch to ask his State Congressman to intervene.

Bulfinch’s tenure as Architect of the Capitol ended abruptly on June 25th, 1829 under the Presidency of Andrew Jackson. Without any notice, Bulfinch received a letter from the Commissioner of Public Buildings instructing him to prepare to vacate the position at the end of the month. He stayed on in Washington until the summer of 1830, finishing other projects, before retiring to Maine.