"During the many years he presided over the town government, he improved its finances, executed the laws with firmness, and was distinguished for gentleness and urbanity of manners, integrity and purity of character." - Josiah Quincy Boston's second mayor

At the early age of twenty-seven, Bulfinch was unanimously elected to fill a vacated seat on the Board of Selectmen for the Town of Boston (1789). In 1795, he resigned due to business pressure but returned again in 1799 as Chairman of the Board and served without any compensation, until his departure from Boston in 1817 to become Architect of the Capitol. Bulfinch improved the town's lighting, drains, and streets, and his involvement with the architecture of the city was expanded by his civil service. As Chairman he was elected to oversee and to design a number of iconic community spaces including the renovation of Faneuil Hall, the Boston Common and oversee much of the work involved with the filling of various tidal wastelands to accommodate the growing town.

In 1805, he was called upon to remodel and enlarge Faneuil Hall to fit the town's needs. The original design was based on the artist's Smibert's drawings. In an elegant and thoughtful design, Bulfinch doubled the height and width of the building, adding a third floor, while still preserving the atmosphere and effect of the old design. He increased the building by four bays, and added pilasters on the first two floors of the doric order and pilasters of the ionic order to the third floor. Another public project which Bulfinch was selected to oversee was the transformation of the Boston Common (1810), the country's first public park established in 1634. Used as a cow pasture throughout the 18th century, the park was in need of repairs due to the war as well as a violent storm from the previous year. A large portion of the work involved with the filling of wetlands, including the Mill Pond, fell to Bulfinch. Streets and parcels of land had to be mapped and many new buildings designed. The Mill Pond, a tidal wetland, became the Bulfinch Triangle, an area in Boston's West End bounded by North Washington, Merrimac and Causeway Streets. By 1867, the area was dominated by industrial factories, because of its proximity to the railways. Another early project was the construction of Broad Street and India Street, where a wharf (1804) was built to accommodate international trade. Bulfinch designed the large stone building on top of the wharf, which was completed in 1807. Today the Harbor Towers, built in the brutalist style stand in the same spot.

Other projects Bulfinch took on while Selectman included Colonnade Row, a group of nineteen houses on land previously part of the Common. The houses were characterized by a doric colonnade topped with a balcony. Bulfinch also helped develop building standards, and designed and oversaw the building of the third Boston Latin School, Boylston Market and the West Boston Bridge. Known as the Salt and Pepper Bridge today. Despite the abundance of work and civic responsibilities, Bulfinch found himself in a state of insolvency a number of times while selectman. Not paid for many of his architectural projects or for his post as selectman, Bulfinch's income came from his position as police superintendent. In 1811, Bulfinch was jailed in a prison building of his own design because of debt.

Bulfinch's duty as Chairman of the Board of Selectmen led to his eventual appointment as Architect of the Capitol in 1817, ending his storied career of service to Boston. In July of 1817, President James Monroe visited Boston to tour what was then becoming one of America's leading urban centers. In his role as Chairman, one of Bulfinch's duties was to accompany the President during his visit.

"My duty as chairman led me to be almost constantly in company with the President during his visit of about a week..." - Charles Bulfinch, Life and Letters

Sometime later, upon hearing of Benjamin Henry Latrobe's resignation, Bulfinch applied to John Quincy Adams for the position and was soon appointed as the third Architect of the United States Capitol in Washington D.C.