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Current trends in semiconductor device manufacturing impose extremely stringent requirements on nanoscale processing techniques, both in terms of accurately controlling material properties and in terms of precisely controlling nanometer dimensions. Plasma-based processing remains key in next-generation device manufacturing with plasma-enhanced atomic layer deposition (PEALD) being a method that has obtained a very prominent position in obtaining ultrathin films with atomic scale precision. Although the effects of ion-surface interactions have been investigated for conventional plasma-enhanced chemical and physical vapor deposition in great detail, very little is known about the role of ions during PEALD. In this work, the role of ion-surface interaction during PEALD will be addressed by analyzing the flux and energy of ions arriving at the surface and by linking these results with the material properties obtained. Moreover, it will be demonstrated how the properties of materials (in particular oxides and nitrides of Ti, Hf and Si) can be tailored by controlling the kinetic energy of the ions impinging on the films with RF substrate biasing. This will be shown for planar substrates (up to 200 mm in size) as well as for 3D surface topologies yielding intriguing effects of inducing differing material properties at different surfaces of the nanostructures (see also Faraz et al., ACS Appl. Mater. Interfaces 10, 13158 (2018)).