Copper coating on Polyimide (PI) surface is of great interest due to both the flexibility and the high melting point of PI, and the low cost and high conductivity of copper. PI surface treatment for better adhesion has been critical in copper coating processes. This is particularly important and challenging for large area processing such as roll-to-roll processes. This work presents the use of an atmospheric pressure plasma jet (APPJ) to treat PI for electroless copper plating. The APPJ used in this work is a dielectric-barrier-discharge (DBD)-type plasma jet, which is a home-made system that can be operated under atmospheric pressure. This APPJ is comprised of two outer ring electrodes (one powered and one grounded) wrapped around the outer surface of the quartz tube, which guides the gas flow and serves as the dielectric layer. This APPJ is powered by a commercially available high voltage power source. Stable helium plasma jet can be sustained using a 10 kVpp high voltage at 25 kHz and the helium flow below 2 slm. After the plasma treatment on PI, silver nitrite ink, serving as the activated agent, is coated on the PI surface then heated at 55 °C. Electroless copper plating is then performed by soaking the PI in the copper plating bath and finally quantified by cross-cut test. It is found that the adhesion of the copper film can be greatly improved with plasma treatment for less than 5 mins. The jet design and the operation conditions will be optimized to improve the adhesion of the coated film with the goal toward the increase in the process compatibility for high speed and large area coating processes.