The interaction between β-catenin and B-cell CLL/lymphoma 9 (BCL9), critical for the transcriptional activity of β-catenin, is mediated by a helical segment from BCL9 and a large binding groove in β-catenin. Design of potent, metabolically stable BCL9 peptides represents an attractive approach to inhibit the activity of β-catenin. In this study, we report the use of the Huisgen 1,3-dipolar cycloaddition reaction to generate triazole-stapled BCL9 α-helical peptides. The high efficiency and mild conditions of this “click” reaction combined with the ease of synthesis of the necessary unnatural amino acids allows for facile synthesis of triazole-stapled peptides. We have performed extensive optimization of this approach and identified the optimal combinations of azido and alkynyl linkers necessary for stapling BCL9 helices. The unsymmetrical nature of the triazole staple also allowed the synthesis of double-stapled BCL9 peptides, which show a marked increase in helical character and an improvement in binding affinity and metabolic stability relative to wild-type and linear BCL9 peptides. This study lays the foundation for further optimization of these triazole-stapled BCL9 peptides as potent, metabolically stable, and cell-permeable inhibitors to target the β-catenin and BCL9 interaction.