



Strategies and Tactics in Organic Synthesis:

Chapter 2. Total Synthesis of the Unusual Peptide Celogentin C

Steven L. Castle, Bing Ma

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This account describes the total synthesis of the title compound, an antimitotic bicyclic peptide. A first-generation approach involving right-hand ring formation followed by left-hand ring annulation was unsuccessful but yielded several interesting observations. A revised strategy was devised in which left-hand ring synthesis would precede right-hand macrocycle construction. A suitably functionalized tryptophan derivative was prepared via phase transfer-catalyzed asymmetric alkylation and Larock heteroannulation. A Knoevenagel condensation–radical conjugate addition sequence fashioned the tryptophan–leucine cross-link, and macrolactamization furnished the left-hand ring. The concise, high-yielding nature of the route compensated for the low diastereoselectivity of the radical conjugate addition. Discovery of an NCS-promoted indole–imidazole oxidative coupling facilitated by proline benzyl ester enabled construction of the tryptophan–histidine cross-link, and right-hand macrolactamization delivered the target compound after deprotection. The chemical shifts of the imidazole hydrogens were strongly dependent on pH, temperature, and concentration. The title compound exhibited modest anticancer activity.

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