

# Water-Soluble Curcumin Derivatives Including Aza-Crown Ether Macrocycles as Enhancers of Their Cytotoxic Activity

Antonino Arenaza-Corona,<sup>[a]</sup> Paola Sánchez-Portillo,<sup>[b]</sup> Lucero González-Sebastián,<sup>[b]</sup> Arturo Sánchez-Mora,<sup>[a]</sup> Brian Monroy-Torres,<sup>[a]</sup> Teresa Ramírez-Apan,<sup>[a]</sup> Nicolás Puentes-Díaz,<sup>[c]</sup> Jorge Ali-Torres,<sup>[c]</sup> Víctor Barba,<sup>[d]</sup> Viviana Reyes-Marquez,<sup>[e]</sup> and David Morales-Morales<sup>[a]</sup>

The synthesis of three novel curcumin derivative compounds, featuring aza-crown ether macrocycles of various sizes (aza-12-crown-4, aza-15-crown-5, and aza-18-crown-6), is described. The incorporation of these aza-crown macrocycles significantly enhances their water solubility, positioning them as ground-breaking instances of curcumin derivatives that are fully soluble in aqueous environments. These curcumin ligands (L1, L2, and L3) were then reacted with zinc acetate to afford the coordination metal complexes (L1-Zn, L2-Zn, and L3-Zn). Comprehensive characterization of all compounds was achieved using various analytical techniques, including 1D and 2D NMR

spectroscopy, ATR-FTIR spectroscopy, mass spectrometry (ESI<sup>+</sup>), elemental analysis and UV-Vis spectroscopy. The *in vitro* cytotoxic activity of both, ligands and complexes were evaluated on three human cancer cell lines (U-251, MCF-7, and SK-LU-1). Compared to conventional curcumin, these compounds demonstrated improved antiproliferative potential. Additionally, a wound healing assay was conducted to assess their antimigration properties. The obtained results suggest that these modifications to the curcumin structure represent a promising approach for developing therapeutic agents with enhanced cytotoxic properties.

## 1. Introduction

In the ongoing pursuit of effective therapies for proliferative diseases, there has been a remarkable surge in interest surrounding natural bioactive compounds. Notably, curcumin, extracted from the rhizomes of *Curcuma longa*, has emerged as a promising candidate due to its multifaceted potential in therapeutic intervention. This polyphenol exhibits a wide array of activities, including anti-inflammatory,<sup>[1]</sup> antioxidant,<sup>[2,3]</sup> antitumoral,<sup>[4–7]</sup> antimicrobial, antiviral, chemotherapeutic<sup>[8]</sup> effects, and serves as a wound healing agent.<sup>[9,10]</sup> However, despite these beneficial attributes, its application in medicine is

constrained by challenges such as low stability, limited bioavailability and rapid metabolism.<sup>[11]</sup>

As a result, the design of curcumin-based compounds by incorporating various groups into its framework has become a key area of interest in current research. For instance, efforts to enhance its transport and bioavailability have led to the incorporation of cyclodextrins, resulting in curcumin derivative compounds soluble in water and with increased dispersibility. Additionally, other strategies<sup>[12]</sup> include attaching curcumin to dendrimer supports and organic polymers<sup>[13]</sup> as well as developing co-drugs such as piperine,<sup>[14–16]</sup> all aimed at improving its bioavailability.

Given the fact that the  $\beta$ -diketone system within the curcumin structure serves as a chelate for metal complex formation, this trait has been harnessed to produce a range of curcumin-metal compounds incorporating diverse divalent and trivalent metals. This approach enhances the stability and bioavailability of curcumin derivatives, thereby amplifying their biological activity.<sup>[17–19]</sup> In this context, the zinc metal center, has been shown to enhance the activity of these curcuminoids.<sup>[20]</sup> Another successful strategy employed involves structural modifications of the parent molecule, including substitutions on the OH group of the phenol with either aliphatic or acetyl chains.<sup>[21,22]</sup>

Thus, the structural modification of curcumin has rendered valuable derivatives as therapeutic agents, achieved by incorporating specific molecules that enhance the stability, transportation, and bioavailability of its derivatives. In this context, aza-crown ethers that are supramolecular macrocycles possessing an amphiphilic nature<sup>[23]</sup> and noted for their antitumor activity,<sup>[24]</sup> have been selected to be incorporated in the

[a] A. Arenaza-Corona, A. Sánchez-Mora, B. Monroy-Torres, T. Ramírez-Apan, D. Morales-Morales  
Instituto de Química, Universidad Nacional Autónoma de México, Circuito Exterior S/N, Ciudad Universitaria, Mexico City C.P. 04510, Mexico  
E-mail: damon@quim.unam.mx  
antonarenaza@quim.unam.mx

[b] L. González-Sebastián  
Universidad Autónoma Metropolitana-Itzapatlapan, Av. San Rafael Atlixo No. 186, Ciudad de México C.P. 09340, México

[c] N. Puentes-Díaz, J. Ali-Torres  
Departamento de Química, Universidad Nacional de Colombia- Sede Bogotá, Bogotá 111321, Colombia

[d] P. Sánchez-Portillo, V. Barba  
Centro de Investigaciones Químicas-ICQA, Universidad Autónoma del Estado de Morelos, Av. Universidad 1001, Col. Chamilpa, Cuernavaca, Morelos C. P. 62209, México

[e] V. Reyes-Marquez  
Departamento de Ciencias Químico-Biológicas, Universidad de Sonora, Luis Encinas y Rosales s/n, Hermosillo, Sonora C.P. 83000, México

Supporting information for this article is available on the WWW under <https://doi.org/10.1002/cbdv.202402083>

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