



Cite this: *Chem. Sci.*, 2025, **16**, 11413

All publication charges for this article have been paid for by the Royal Society of Chemistry

Received 20th March 2025
Accepted 16th May 2025

DOI: 10.1039/d5sc02168e
rsc.li/chemical-science

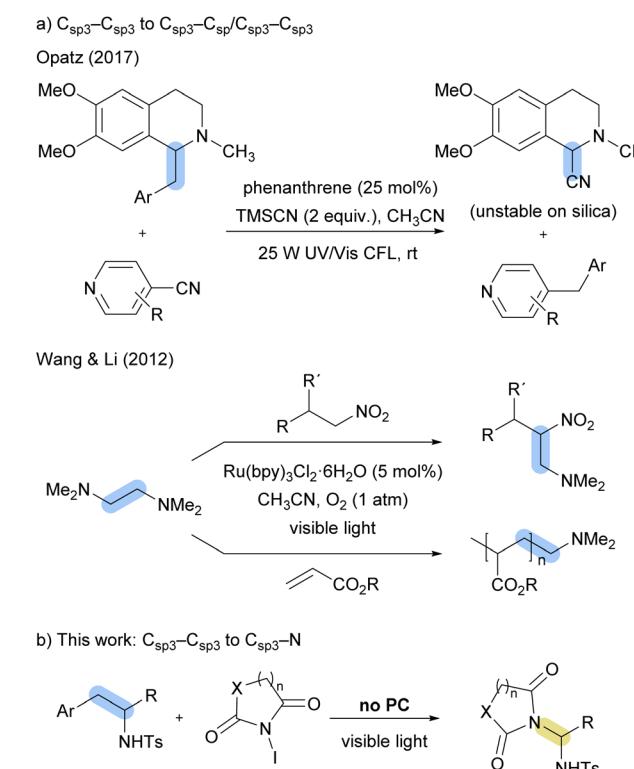
Introduction

Visible-light-promoted reactions have become in recent years a powerful tool for both the formation and cleavage of chemical bonds and generate molecular complexity.¹ The cleavage of C–C bonds is of particular interest,^{2,3} due to their ubiquity in organic molecules and the inherent challenge caused by their high dissociation energies. Similarly, the formation of C–N bonds under mild reaction conditions is highly relevant given the widespread presence of nitrogen-containing substructures in bioactive compounds and materials science.⁴

Nitrogen-centered radicals have proven to be valuable intermediates for the formation of new C–N bonds.⁵ In recent years, numerous visible-light-promoted methodologies have been developed to provide aminyl, amidyl, iminyl or aminium radicals as key intermediates.⁵ Sulfonamidyl radicals promoted by visible-light have been employed in the literature in reactions involving the addition to unsaturated systems,⁶ such as aromatic rings or alkenes, in Hofmann–Löffler reactions *via* 1,5-HAT mechanism,⁷ as well as cycloadditions.⁸

Notably, iminyl and aminium radicals have been exploited as transient species in tandem C–C bond cleavage/functionalisation processes mediated by visible light.^{2c} This reactivity can be divided into two groups, (i) the use of cyclic ketone-derived oximes, in which an iminyl radical intermediate cleaves a C_{sp}²–C_{sp}³ bond, leading to the formation of a nitrile group and the subsequent generation of a new C_{sp}³–C or C_{sp}³–X

bond,⁹ and (ii) the formation of radical aminium cations, which trigger C_{sp}³–C_{sp}³ bond cleavage followed by the formation of a different C_{sp}³–C_{sp}/_{sp}³ bond¹⁰ (Scheme 1a). Cycloadditions have also been reported from cyclopropyl or cyclobutylanilides.¹¹ Meanwhile C_{sp}³–N cleavage/C_{sp}³–C_{sp}³ bond formation has been described in the literature by photoredox procedures,¹² to the best of our knowledge no tandem C_{sp}³–C_{sp}³ bond cleavage/C_{sp}³–N formation has been reported from nitrogen-centered radicals.



Scheme 1 Tandem C_{sp}³–C_{sp}³ bond cleavage/functionalisation processes initiated by N-centered radicals mediated by visible-light.

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† Electronic supplementary information (ESI) available. CCDC 2431275. For ESI and crystallographic data in CIF or other electronic format see DOI: <https://doi.org/10.1039/d5sc02168e>



Table 1 Reaction optimization (selected data)^a

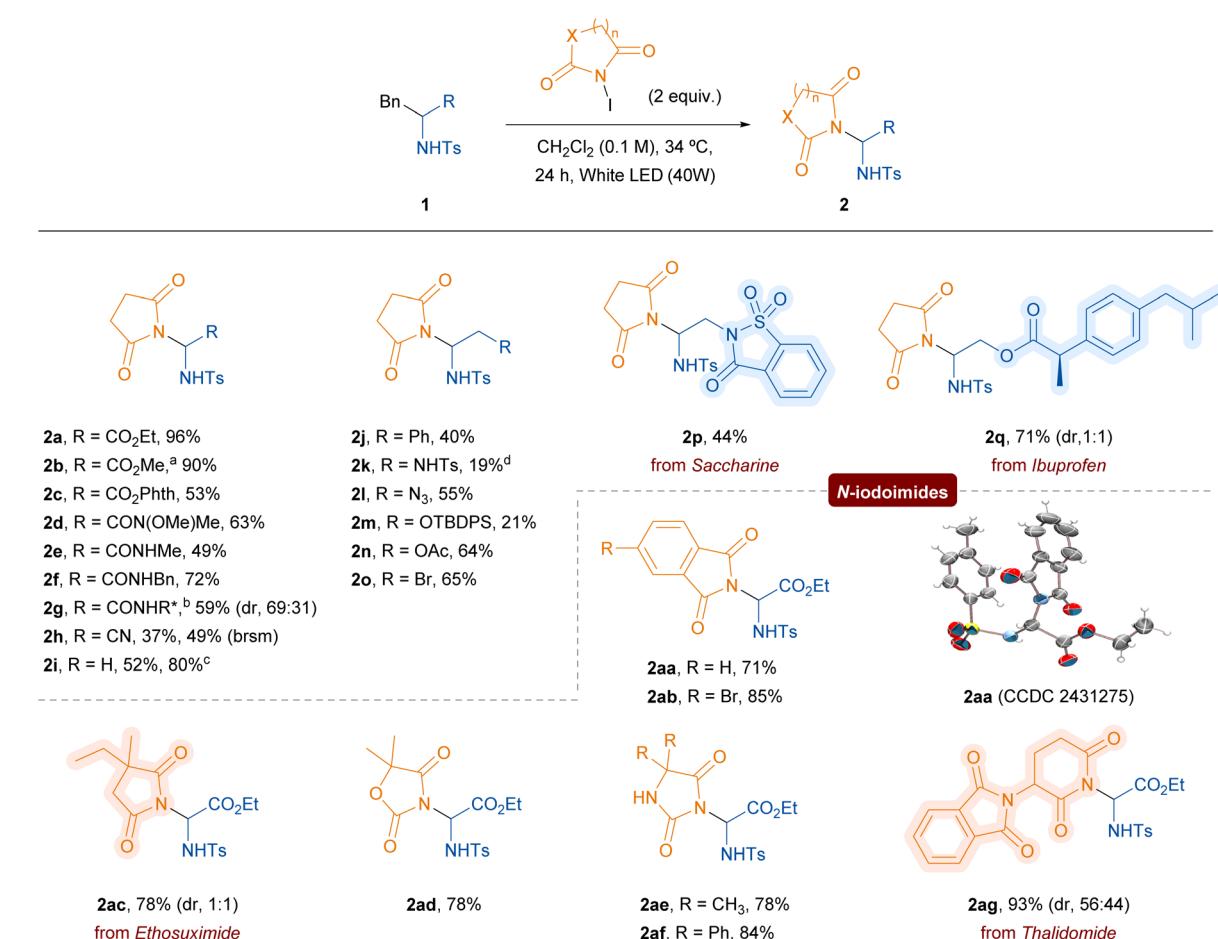
Entry	Deviation from above	Conv. [%]
1	None	100 (96) ^b
2	CH ₃ NO ₂ instead of CH ₂ Cl ₂	82
3	CH ₃ CN instead of CH ₂ Cl ₂	30
4	NIS (1.5 equiv.)	78
5	NBS (1.5 equiv.)	—
6	Blue instead of white LED	86
7	No light irradiation (40 °C)	—
8	NHMs instead of NHTs	100 (81) ^b
9	1 mmol scale	82 (70) ^b

^a Reaction conditions: **1a** (0.2 mmol), NIS (2 equiv.), CH₂Cl₂ (0.1 M), 34 °C, white Kessil® (40 W), EvoluChem PhotoRedOx Box™ photoreactor, 24 h. Conversion measured by ¹H NMR. ^b In parenthesis, isolated yield.

Herein we describe the reactivity of homobenzylic sulfonamides with *N*-iodoimides, leading to the formation of sulfonylamidyl radicals which results in the C_{sp}³–C_{sp}³ bond cleavage followed by amination with the imide group (Scheme 1b). Under visible light irradiation, no photocatalyst is required,¹³ and the reaction produces compounds featuring a *gem*-diamino motif,¹⁴ found in several bioactive molecules, and an *N*-substituted imides,¹⁵ substructure with applications in medicinal chemistry or agrochemicals.

Results and discussion

We began our investigation studying the reactivity of *N*-tosyl L-phenylalanine ethyl ester as model substrate in the presence of *N*-iodosuccinimide (NIS), upon irradiation with visible light. Initial experiments showed a clean transformation to the 1,1-diamine **2a**, by formal substitution of the benzyl group by the succinimide moiety. Optimisation parameters included solvent, concentration, NIS equivalents, as well as light source (see ESI for more details†). The best results were obtained with CH₂Cl₂ as solvent, comparing with a variety of polar and apolar solvents



Scheme 2 Substrate scope. Reaction conditions: **1** (0.2 mmol), *N*-iodoimide (2 equiv.), CH₂Cl₂ (0.1 M), 34 °C, white Kessil® (40 W), EvoluChem PhotoRedOx Box™ photoreactor, 24 h. Isolated yields. ^a(*S*)-Methyl-2-(4-methylphenylsulfonamido)-3-(4-(tosyloxy)phenyl)propanoate employed as substrate. ^bR* = (*S*)-1-(1-naphthyl)ethylamine. ^c*N*-(2,2-Diphenylethyl)-4-methylbenzenesulfonamide employed as substrate. ^d(*S*)-4-Methyl-*N*-(1-tosyl-1,2,3,4-tetrahydroquinolin-3-yl)benzensulfonamide was obtained as the major product (see ESI†).



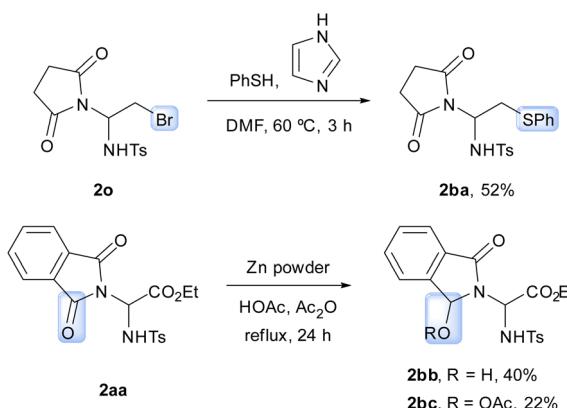
(Table 1, entries 2 and 3, see ESI†). Changes in the concentration had minor effect on the outcome of the reaction. An excess of 2 equivalents of NIS were necessary to achieve full conversion, while *N*-bromosuccinimide (NBS) gave no product (entries 4 and 5). White LED showed the best results comparing with different wavelengths (entry 6, see ESI†), and the irradiation was essential for the reaction to proceed (entry 7). In summary, the optimized reaction parameters involved 2 equiv. of NIS in CH_2Cl_2 (0.1 M) under 40 W white LED irradiation for 24 h to get the product in 96% isolated yield. Other sulfonamides were tested, while mesylate gave a good result (81%, entry 8), triflimide showed low conversion (14%, see ESI†). When scaling up the reaction to 1 mmol under the same conditions we isolated diamine **2a** in 70% yield (entry 9).

Next, the scope of the transformation was explored (Scheme 2). Several carbonyl derivatives successfully led to the corresponding 1,1-diamines. Methyl ester (**2b**) was obtained from the *O*-tosyl tyrosine derivative and photoactive *N*-hydroxyphthalimide ester (**2c**) was also compatible with the reaction conditions. Different amides provided the products in good yields (49–72%, **2d**–**2f**). A chiral amide, from (*S*)-1-(1-naphthyl)ethylamine, was tested to obtain a moderate diastereoselectivity in the process (**2g**). Ketones were unsuccessful under these reaction conditions, whereas nitrile derivative had a moderate reactivity to the corresponding product (**2h**). Remarkably, simple homobenzyl *p*-toluenesulfonamide led to the formation of the corresponding *gem*-diamine (**2i**) in moderate yield. The result could be improved by using diphenylmethyl chain as formal leaving group (80%). A range of functionalities were also included as substituents, such as phenyl, sulfonamide, azide, ether, ester or bromine (**2j**–**2o**). Moderate to good yields were obtained in all the cases (19–65%). The presence of an additional sulfonamide resulted in the competitive formation of the tetrahydroquinoline ring,¹⁶ decreasing the isolated yield of the 1,1-diamine **2k**. We have also incorporated bioactive structures in the molecule, such as saccharine or ibuprofen (**2p**, **2q**). Unfortunately, functional groups like alcohol, acetal, sulfide or sulfone led to decomposition or no reaction under the reaction conditions (see ESI†).

Subsequently, we explored the substrate scope with respect to *N*-iodoimides. The reaction was tested with different phthalimide and succinimide derivatives with good isolated yields (71–85%, **2aa**–**2ac**). The structure of compound **2aa** was unambiguously established by single crystal X-ray diffraction (XRD) analysis. Different heterocycles, such as oxazolidine-2,4-dione or hydantoin are also compatible with this transformation (**2ad**–**2af**). Some bioactive imides, such as ethosuximide or thalidomide were successfully incorporated into the products (**2ac**, **2ag**).

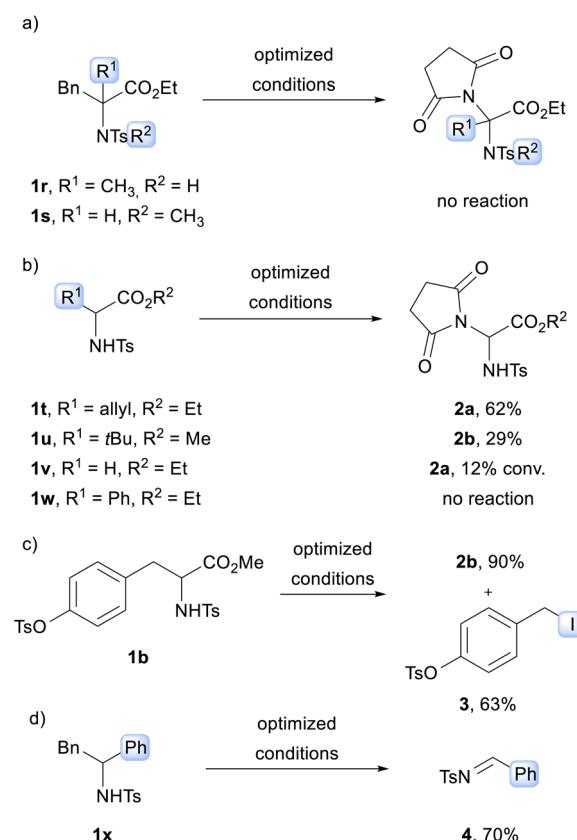
To illustrate the application of resulting *gem*-diamines, some transformations are presented in Scheme 3. The bromine derivative **2o** was subjected to nucleophilic substitution by thiophenol in good yield. Furthermore, a carbonyl group of the phthalimide on compounds **2aa** was partially reduced to obtain hydroxy and acetyl isoindolinone derivatives, substructure present in bioactive compounds.¹⁷

To establish a mechanistic proposal several experiments were performed. Initially, methylated derivatives **1r** and **1s** were



Scheme 3 Derivatization of 1,1-diamines.

subjected to the standard conditions (Scheme 4a). The reaction did not occur, indicating the transformation is initiated with a nitrogen-centred radical, and the alpha position cannot be a quaternary centre to allow the C–C bond cleavage. Different groups have also been placed at the alpha position of the sulfonamide to compare different formal leaving groups (Scheme 4b). Allyl or *tert*-butyl substituted glycine derivatives **1t** and **1u**, which can also stabilise radicals, led to moderate yields of the corresponding *gem*-diamino derivatives. Protected glycine derivative **1v** only provides a low conversion to the product, while phenyl glycine derivative **1w** does not react. These results suggest the C–C bond cleavage generates a carbon radical as



Scheme 4 Control experiments.



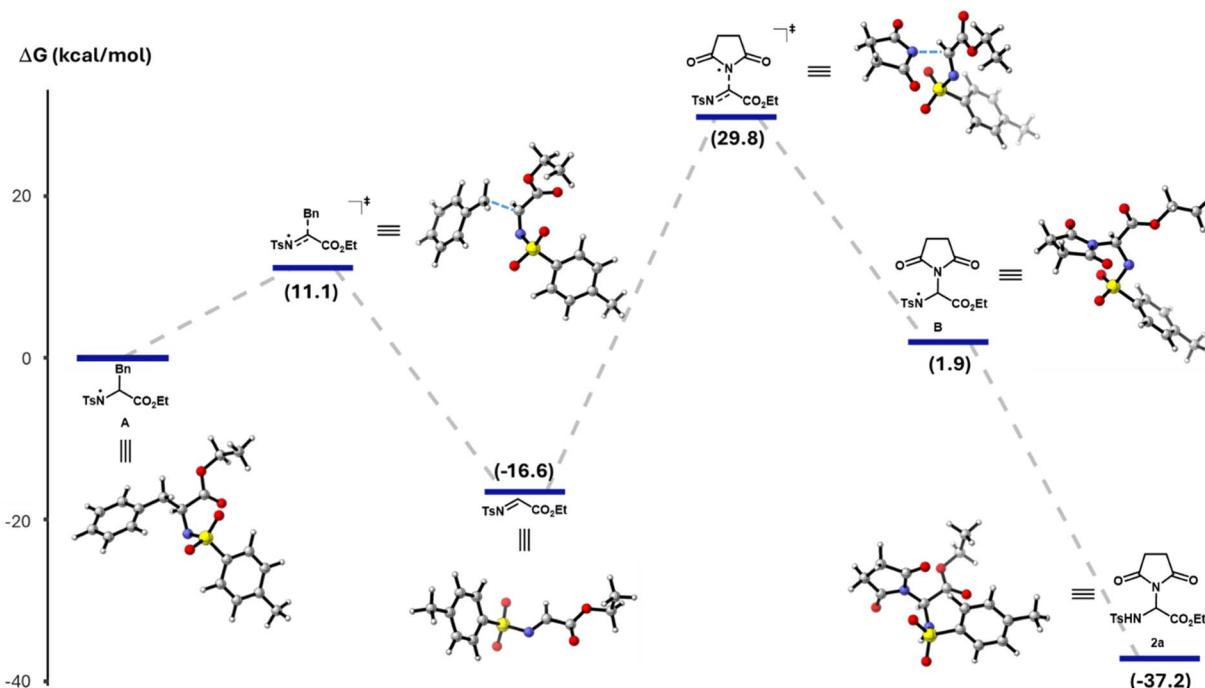


Fig. 1 Energy profile calculated with B3LYP/LANL2DZ for I and 6-311+g(d,p) basis set for all other atoms. Solvent effects were considered by applying the solvation model based on IEPCM, using dichloromethane as the solvent at 313.15 K. Activation free energies are given in kcal mol^{-1} .

byproduct and are consistent with the improved yields observed for product **2i** when using diphenyl methyl instead of benzyl as the formal leaving group. This was confirmed in the reaction with *O*-tosyl tyrosine (**1b**), as benzyl iodide derivative **3** was isolated (Scheme 4c). When sulfonamide **1x** was subjected to standard conditions, imine **4** was obtained as the sole product (Scheme 4d), rather than the expected *gem*-diamine. This suggests that the imine acts as a reaction intermediate. For this substrate, stabilisation by conjugation to the aromatic ring probably prevents diamine formation.¹⁸

Based on these set of results we proposed the mechanism outlined in Fig. 1, supported by DFT calculations. Initially, NIS is activated in the presence of light, generating the succinimide radical, which abstracts a hydrogen atom from phenyl alanine derivative **1a**, forming sulfonamidyl radical **A** through an exothermic process (see ESI†). These species have been identified by Muñiz *et al.* using EPR spectroscopy.¹⁹ Subsequently, tosyl imine is generated *via* a transition state with a ΔG^\ddagger of +11.1 kcal mol^{-1} , with the release of the benzyl radical, which would couple with iodine radical. An imine was the product obtained in the reaction with **1w**, with phenyl as a substituent, and a benzyl iodide derivative (**3**) was obtained by reacting **1b** (Scheme 4), giving experimental support to these steps. Next, the attack of the succinimide radical leads to sulfonamidyl radical **B** *via* a transition state with a ΔG^\ddagger of +29.8 kcal mol^{-1} . Finally, hydrogen abstraction results in the formation of **2a** through an exothermic process.²⁰

Conclusions

In summary, we have developed a tandem $\text{C}_{\text{sp}}^3\text{--C}_{\text{sp}}^3$ bond cleavage/ $\text{C}_{\text{sp}}^3\text{--N}$ bond formation for homobenzyllic

sulfonamides in the presence of *N*-iodoimides under visible-light irradiation and metal-free conditions. The reaction features a wide scope in both reagents and led to a new method to prepare functionalized compounds that incorporate both *gem*-diamine and *N*-substituted imide motifs. Additionally, further functionalisation has also been demonstrated. Mechanistic study suggests that sulfonamidyl radicals initially generated provide imine intermediates through the loss of the benzyl group. These imines subsequently react with imide radicals to yield the products. This study extends the reactivity described for sulfonamidyl radicals, limited to addition to unsaturated systems, Hofmann–Löffler reactions and cycloadditions.

Data availability

All experimental and characterization data, as well as NMR spectra are available in the ESI.† Crystallographic data for compound **2aa** has been deposited at the Cambridge Crystallographic Data Centre under accession number CCDC 2431275.

Author contributions

G. M.-O. carried out the experimental work and E. M. the computational study. J. C. designed the project and supervised the work. J. C. wrote the paper and all the authors discussed the manuscript.

Conflicts of interest

There are no conflicts to declare.



Acknowledgements

We gratefully acknowledge financial support of this work by Ministerio de Ciencia, Innovación y Universidades of Spain (Agencia Estatal de Investigación, grants: CNS2022-135208 and CNS2022-135304 financed by MCIN/AEI/10.13039/501100011033 and EU NextGenerationEU/PRTR), Instituto de Salud Carlos III and EU (RICORS2040, Kidney Disease, RD24/0004/0008) and Universidad de Alcalá (Plan Excelencia: EPU-INV-UAH/2023/002 and EPU-INV-UAH/2023/005). G.M.-O. thanks Comunidad de Madrid & MICIU for predoctoral contracts. Computational resources for the DFT calculations were provided by the HERMES Cluster at the University of Zaragoza. We also thank Marina Manzanares for initial experiments and Dr. R. Martín Romero for fruitful discussions.

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