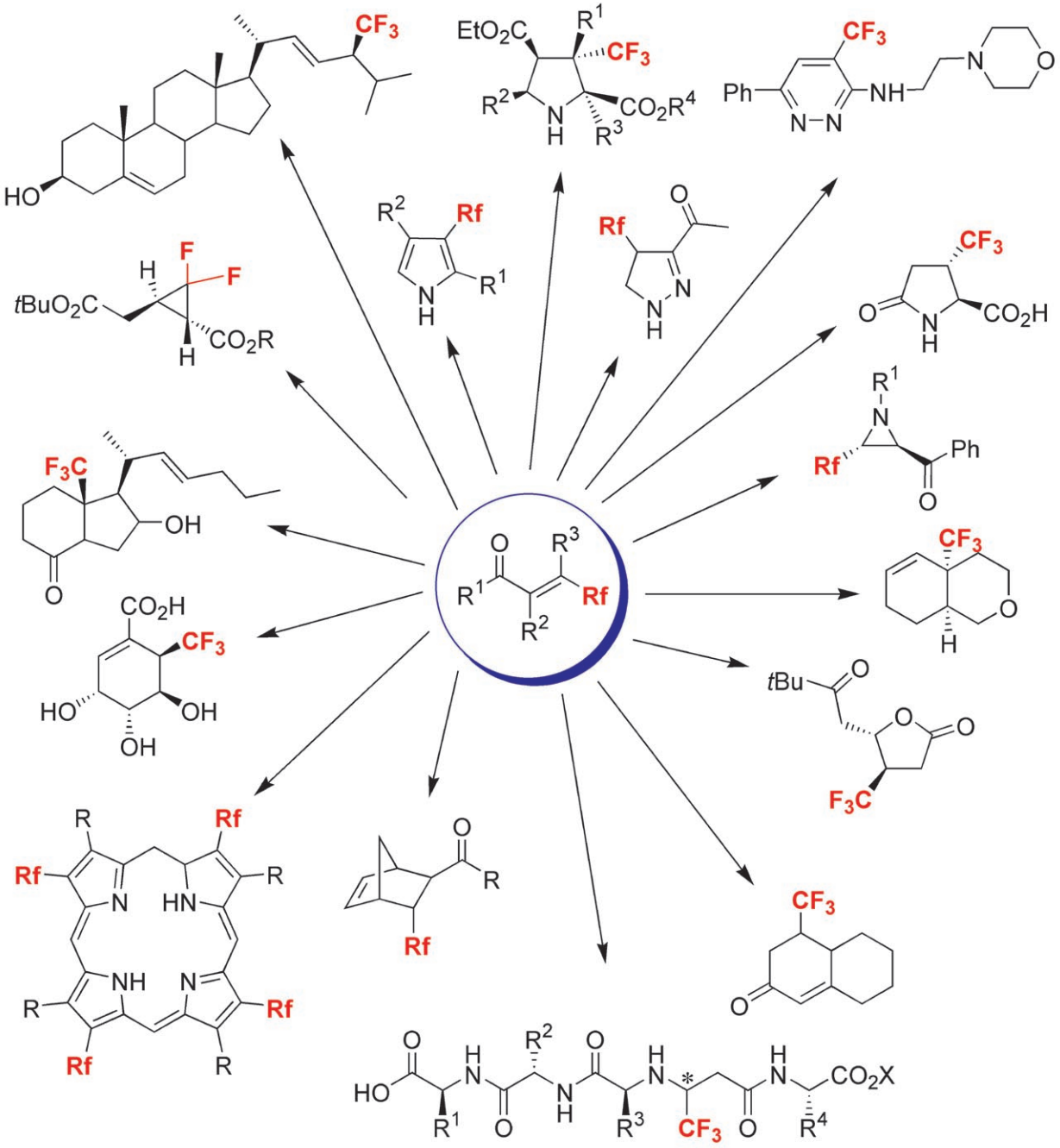

Synthetic applications of β-fluoroalkylated α,β-unsaturated carbonyl compounds

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Synthetic Applications of β-Fluoroalkylated α,β-Unsaturated Carbonyl Compounds

Thierry Billard*[a]

Abstract: β-Fluoroalkylated α,β-unsaturated carbonyl compounds constitute efficient building blocks for the synthesis of complex fluorinated compounds. As the fluorinated moiety generally increases their reactivity, it also brings important modifications which can change the chemical behavior and selectivity. Their use has been already largely demonstrated. Nevertheless, the synthetic potential has not yet been fully explored and, consequently should play an important role in the design of new sophisticated fluorinated molecules. Nevertheless, it shall be important to develop new synthetic methods to enlarge their availability and their diversity.

Keywords: carbonyl compounds · cycloaddition · fluorine · Michael addition

Introduction

Fluorine is an element which occupies a special place in the periodical classification. Because of its highest electronegativity and its specific properties, it is often considered on the fringe of the classical organic chemistry and the synthesis and properties of fluoro compounds are traditionally omitted in main stream organic chemistry.

However, the singular nature of the fluorine atom, combined with the unique physical and chemical properties that the fluorine substituent imparts its compounds, is responsible for the importance of the field of fluorine chemistry.[1] Indeed, the specific physico-chemical properties of fluorinated organic compounds are of huge interest in a wide range of applications.[1–2]

Since the Henri Moissan's first isolation of elemental fluorine in 1886, the interest in fluorine chemistry has grown steadily to a field of great significance which today plays a distinctive role in many and highly diverse technological developments (for example fluoropolymers, pharmaceutical and agrochemical products, material science).[3–4] The recent development of the Efavirenz (anti-HIV drug)[5] and Vinflunine (antitumour drug),[6] two fluorinated drugs promising in their prophylaxis, constitutes an excellent illustration of the important role played by the fluorinated compounds for society.

Nevertheless, despite this growing interest for fluorinated compounds, the synthesis of more sophisticated fluorinated molecules, in particular for biological applications, is still scarce because of the lack of various and useful fluorinated building blocks.

Towards Synthetic Applications of β-Fluoroalkylated α,β-Unsaturated Carbonyl Compounds

In non-fluorinated organic chemistry the α,β-unsaturated carbonyl compounds constitute interesting building blocks for further functionalisation by various reactions. Indeed, such substrates are well-known reagents for cycloaddition reactions and conjugated additions.[7] Some calculations on β-trifluoromethylated analogues of such substrates have shown that the LUMO energy level is lower and, consequently, led to suggest that these fluorinated compounds should be more reactive (Figure 1).[8]

Consequently, such β-trifluoromethylated α,β-unsaturated carbonyl compounds should constitute very efficient building blocks as starting material in the synthesis of fluorinated molecules.

The commercially availability of some esters of (*E*)-trifluorocrotonic acid has boosted their use in reactivity studies and synthetic applications. Thus we can propose that others

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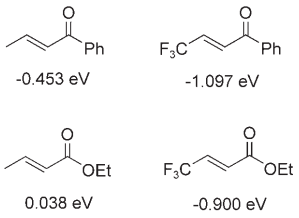
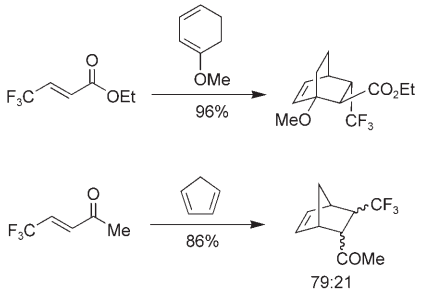


Figure 1. LUMO energy level.

β-fluoroalkylated α,β-unsaturated carbonyl compounds should be also reactive substrates for further reactions.

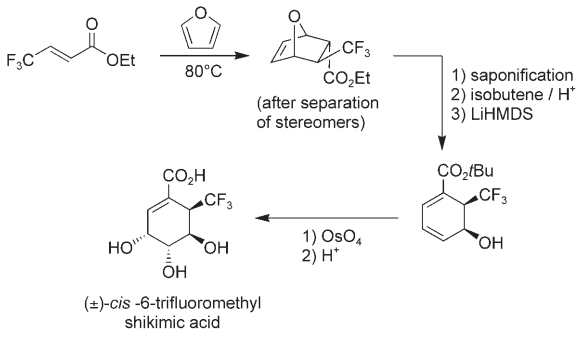
Cyloadditions Reactions with β-Fluoroalkylated α,β-Unsaturated Carbonyl Compounds

As early as 1963, fluorinated *trans*-olefinic acids have been used as dienophile in the Diels–Alder reaction with cyclopentadiene.[9] Further reactions have been developed with the ethyl (*E*)-trifluorocrotonate,[10] but a few Diels–Alder reactions have been conducted with trifluoromethylated enones.[11] (Scheme 1)



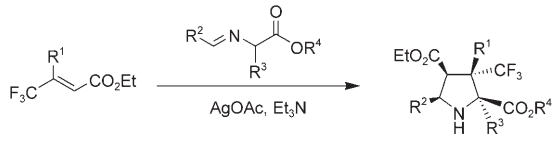
Scheme 1. Diels–Alder cycloadditions.

This type of Diels–Alder cyclization has found interesting applications in the synthesis of trifluoromethylated analogues of a natural product[10b] of medicinal importance, that is shikimic acid (Scheme 2),[26] which is involved in important biosynthetic pathways.



Scheme 2. Synthesis of (±)-*cis*-6-trifluoromethyl shikimic acid.

Trifluoromethylated pyrrolidines, which could be very efficient trifluoromethylated building blocks for the syntheses of fluorinated analogues of various alkaloids, have been obtained through 1,3-dipolar cycloadditions[11–12] with β-fluoroalkylated α,β-unsaturated carbonyl substrates (Scheme 3).[12c,d]



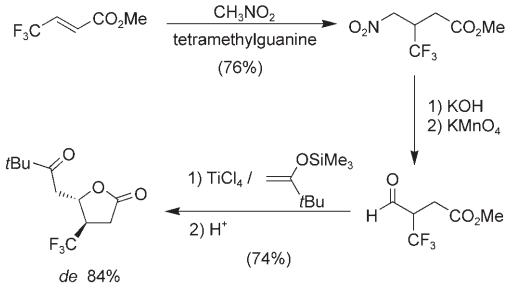
Scheme 3. Synthesis of 3-trifluoromethylated pyrrolidines.

Nevertheless, despite the high interest for such reactions and the expected enhancement of reactivity, the presence of a fluoroalkyl group at the β position of these enones brings about some modifications of selectivity or reactivity. For example, the Diels–Alder cyclization of (*E*)-4,4,4-trifluoro-1-phenylbut-2-en-1-one with cyclopentadiene leads to the cycloadduct with the CF_3 in *endo* position as major diastereomer.[27] Because of the steric hindrance of CF_3 moiety,[28] some cycloadditions are difficult to realize and require more drastic conditions (high pressure for instance).[12c]

Conjugated Additions with β-Fluoroalkylated α,β-Unsaturated Carbonyl Compounds

Conjugated additions are generally facilitated by the presence of fluoroalkyl group. Again, derivatives of (*E*)-trifluorocrotonic acid have mostly been used in these reactions.[8a,13–17] These conjugated additions certainly constitute the most frequently applied reaction with the β-fluoroalkylated α,β-unsaturated carbonyl compounds.

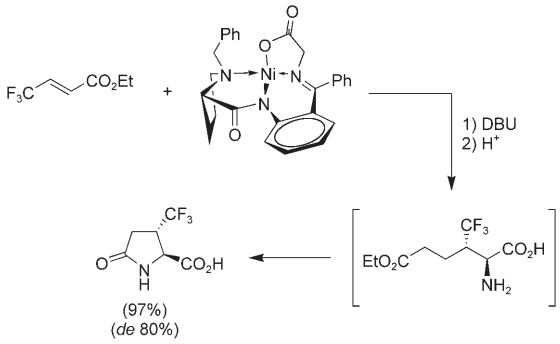
For example, trifluoromethylated γ-lactones have been easily obtained with nitromethane as the nucleophile (Scheme 4).[14] Such a method gives an interesting strategy



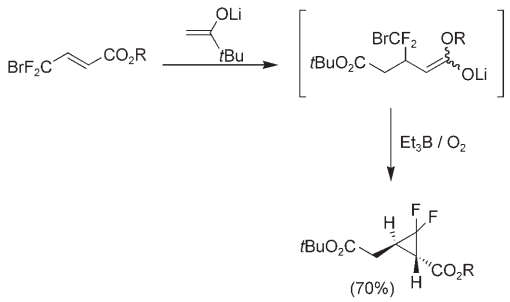
Scheme 4. Synthesis of trifluoromethylated γ-lactone.

for the synthesis of many trifluoromethylated analogues of biologically active compounds and natural products,[29] as, for instance, the oak lactone.[30]

The Michael additions also allowed the synthesis of various fluorinated analogues of amino acids.[15,19] Their incorporation into peptides gives rise to structural modifications (secondary structure), which also result in proteolytic stability (Schemes 5–6).[18]



Scheme 5. Synthesis of 3-trifluoromethylpyroglutamic acid.

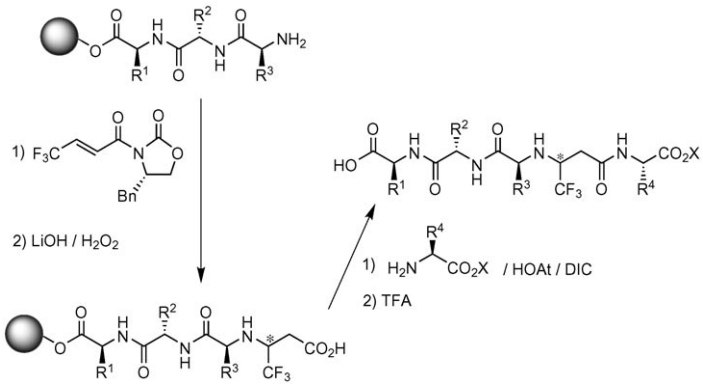


Scheme 6. Synthesis of *trans*-3,4-(difluoromethano)glutamic acid.

Zanda et al. have developed an interesting synthesis of partially modified retro and retro-inverso ψ-[NHCH(CF_3)]Gly peptides based on an aza-Michael reaction of α-amino esters with chiral β-trifluoromethylated α,β-unsaturated amides.[16] This methodology allows parallel solid-phase synthesis of small libraries of trifluoromethylated peptidomimetics to study the influence of the trifluoromethyl group in the biologic behavior of peptides (Scheme 7).[16b]

Conjugated addition of cyanide anion onto β-trifluoromethylated enones has been also applied in the synthesis of aminopyridazines as acetylcholinesterase inhibitors.[20]

As in the case of cycloadditions, the presence of the fluorinated moieties can also be associated with certain drawbacks. Indeed, the high electron-withdrawing fluoroalkyl group, relayed by conjugated π system of enone, leads to a decrease of charge density in the oxygen atom. Consequently, the activation of these fluorinated enones by Lewis acids is less efficient and can suffer of competition with other ligands. For example, Copper(II) triflate catalyses selected 1,4-additions onto fluoroalkylated enones whereas the same catalyst is inactive when chelated to a BOX ligand.[27]

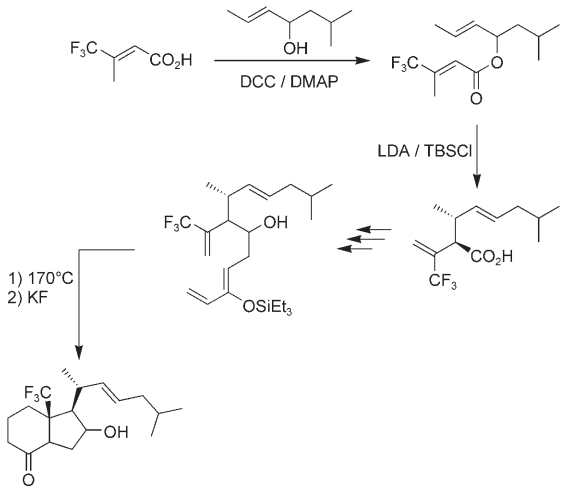


Scheme 7. Solid-phase synthesis of ψ[NHCH(CF_3)]Gly peptides.

Miscellaneous Reactions with β-Fluoroalkylated α,β-Unsaturated Carbonyl Compounds

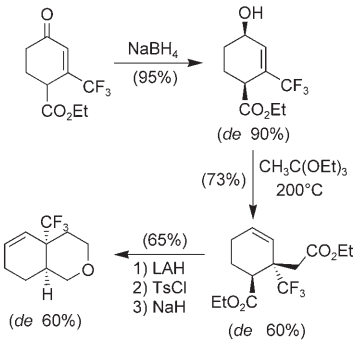
Other synthetic applications of β-fluoroalkylated α,β-unsaturated carbonyl compounds can be also envisaged by modification of carbonyl functionality.[23]

Thus, the synthesis of trifluoromethylated hydrindenes, as potential precursors for fluorinated vitamin D, has been described starting from 3-trifluoromethyl but-2-enoic acid. The key step is an Ireland–Claisen rearrangement of an allylic ester of this acid (Scheme 8).[21]



Scheme 8. Synthesis of trifluoromethylated hydrindenes.

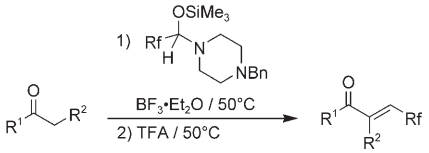
By reduction of the carbonyl moiety of a β-trifluoromethylated enones to obtain allylic alcohol, trifluoromethyl tetrahydroisochromanes could be easily achieved through a Johnson–Claisen rearrangement (Scheme 9).[22] Such methodology constitutes an interesting way to access very easily to various bioactive compounds as, for example, fluorinated analogues of the potent κ opioid receptor agonist salvinorin A.[31]



Scheme 9. Synthesis of trifluoromethylated tetrahydroisochromanes.

Syntheses of β-Fluoroalkylated α,β-Unsaturated Carbonyl Compounds

Despite the well demonstrated importance of the α,β-unsaturated carbonyl compounds, the methods to synthesize them are relatively restricted. The oldest and more direct method is the Wittig reaction with fluorinated aldehydes. Nevertheless, the fluorinated analogues of acetaldehyde, as trifluoroacetaldehyde, are not commercially available, very reactive gases which are not easy to handle. Recently, an efficient synthesis of β-fluoroalkylated enones has been described starting directly from corresponding ketones and, thus, enlarges the panel of available β-fluoroalkylated α,β-unsaturated carbonyl compounds, especially bearing fluoroalkyl moiety other than CF_3 (Scheme 10).[24]



Scheme 10. Efficient, one-pot, synthesis of β-fluoroalkylated enones.

Some other methods, involving more steps, have been also elaborated starting from ethyl trifluoroacetoacetate[32] or from 3,3,3-trifluoroprop-1-yne.[33] Nevertheless, all these methods don't allow to achieve all the expected compounds with diverse substituents anywhere. This lack of diversity in their syntheses can constitute a drawback in the development of their future uses.

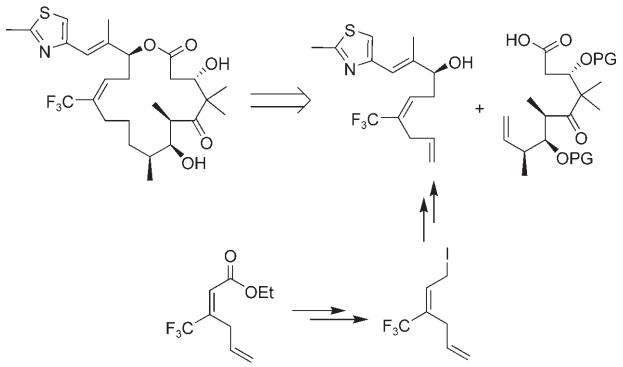
Conclusion and Perspectives

As their non-fluorinated analogues, β-fluoroalkylated α,β-unsaturated carbonyl compounds present interesting and valuable synthetic potential. As shown above in a few pertinent examples, some of them have already been studied and applied in the syntheses of several complex structures and fluorinated analogues of natural products.

The growing tremendous demand of more and more sophisticated fluorinated molecules should certainly imply an increased search of new fluorinated building blocks.

The previous works have shown that β-fluoroalkylated α,β-unsaturated carbonyl compounds, and in particular the derivatives of trifluorocrotonic acid, could constitute valuable building blocks and should play an important role in the future.

This has been recently demonstrated by the synthesis of trifluoromethylated epothilone analogues in which the fluorinated part is obtained from a β-trifluoromethylated α,β-unsaturated carbonyl compound (Scheme 11).[25]



Scheme 11. Synthesis of trifluoromethylated epothilone analogue.

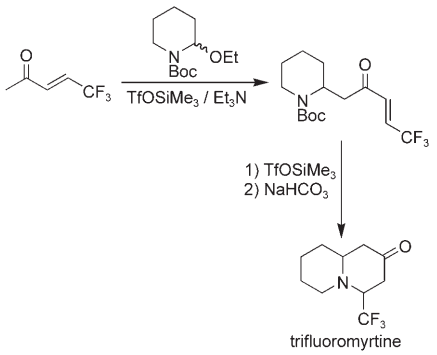
Such work opens the way of the applications of β-fluoroalkylated α,β-unsaturated carbonyl compounds into multistep syntheses of fluorinated analogues of natural compounds.

The already existing reactions, and the incessant development of new ones with non-fluorinated enones, which can be found in literature, in particular in the asymmetric field, expands the field of reactivity studies of β-fluoroalkylated α,β-unsaturated carbonyl compounds.

For instance, hetero-Diels–Alder, by using β-fluoroalkylated enones as heterodienes, could constitute a rapid access to trifluoromethylated carbohydrate mimetics.

Such perspectives make these β-fluoroalkylated α,β-unsaturated carbonyl compounds both classical and essential fluorinated building blocks in the incorporation of fluorinated analogues into natural compounds.

As example, trifluoromyrtine could be easily synthesized starting from (*E*)-5,5,5-trifluoropent-3-en-2-one, by applying a method previously described (Scheme 12).[34]



Scheme 12. Potential synthesis of trifluoromyrtine.

The most important challenge will be to elaborate new synthetic ways to obtain such β-fluoroalkylated α,β-unsaturated carbonyl compounds with various substituents.

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