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Organometallic chemistry

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Highlights from the organometallic literature appearing in 2011 are surveyed. Selected reports are compiled, uses of organometallic species as drugs, organometallic compounds in synthesis and particular but not exclusive attention is paid to the areas of organometallics for solar and electronics applications.

Introduction

In an era of ever increasing popularity of organocatalysis and environmentally benign reaction, metals and hence organometallic complexes are often much maligned. However organometallic complexes remain of significant importance to technological applications. This chapter surveys reports from 2011 and selects a range applications of organometallic compounds from use as catalysts to drugs through to photovoltaics. Whilst review material mostly lies outside the remit of this report readers are directed to particularly noteworthy reviews appearing in 2011 on the green outlook for organometallic chemistry by Crabtree,¹ and a perspective on bioorganometallic chemistry by Hillard and Jaouen.²

Supramolecular

To mark the International Year of Chemistry 2011 (the year surveyed in this chapter) numerous activities took place, among the many conferences, meetings and reports especially produced was an online compendium of *Chemical Communications* articles dedicated to Supramolecular Chemistry.^{3,4} Such activities demonstrate the vitality of current supramolecular chemistry and provide a good overview of the current trends in cutting edge supramolecular chemistry. This chapter focuses on organometallic chemistry and as such a few examples of organometallic chemistry being used in supramolecular self-assembly are noted.

Supramolecular organometallic rectangles were synthesised and applied to nitroaromatic sensing by Bar *et al.*⁵ Among the compounds studied, **1** (Fig. 1) displayed a significant loss of fluorescence proportional to the presence of added picric acid (trinitrophenol or TNP). Thus **1** functions as a supramolecular chemosensor for the explosive material TNP.

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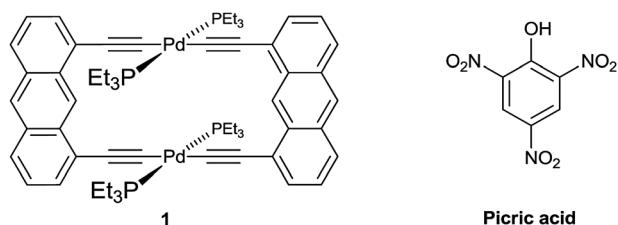


Fig. 1 Sensor 1 and picric acid.

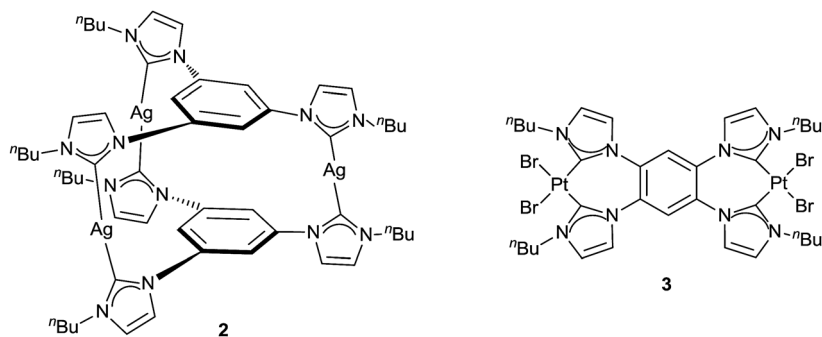


Fig. 2 Carbene complexes 2 and 3.

Rit *et al.* showed that self-assembled, polynuclear cylindrical complexes of carbenes could be generated from polyimidazolium salts **2**.⁶ The authors sought to demonstrate that *N*-heterocyclic carbenes (NHCs) could be used in the construction of Shionoya-type disks,⁷ which have found application as exquisite molecular rotors. In this work transmetalation routes to bisplatinum complexes of the type **3** were also discussed (Fig. 2).

Pitto-Barry *et al.* described the encapsulation of pyrene containing molecules in organometallic cages.⁸ The organometallic cages **7** provided water-soluble encapsulation for the lipophilic pyrene motifs of pyrenebutiric esters (of dendrimers) **5**, which were evaluated for activity against ovarian cancer cells (Fig. 3).

A similar self-assembled system reported by Vajpayee *et al.* was found to be cytotoxic (with no synthetically incorporated guest). Assembly manifested a hexanuclear ruthenium complex akin to a guestless **7** where the trispyridyl part was replaced by **8** (Fig. 4).⁹ This complex in particular was found to have an IC₅₀ better than Cisplatin against certain cancer cell lines (SK-Hel-1 and HCT-15); the authors demonstrated that activity of the supramolecular cage was higher than that of the cage building blocks alone.

A detailed study by Barry *et al.* of related tetranuclear ruthenium complexes,¹⁰ constructed from a range of analogous bis-ruthenium building blocks and nitrogen containing linkers **9** to **12** (Fig. 4) revealed enhanced activity against Cisplatin resistant cell lines was possible.

Anticancer compounds

Platinum complexes¹¹ are well established in anticancer therapies and the modes of action, in some cases, well understood.¹² The use of metals in therapeutic

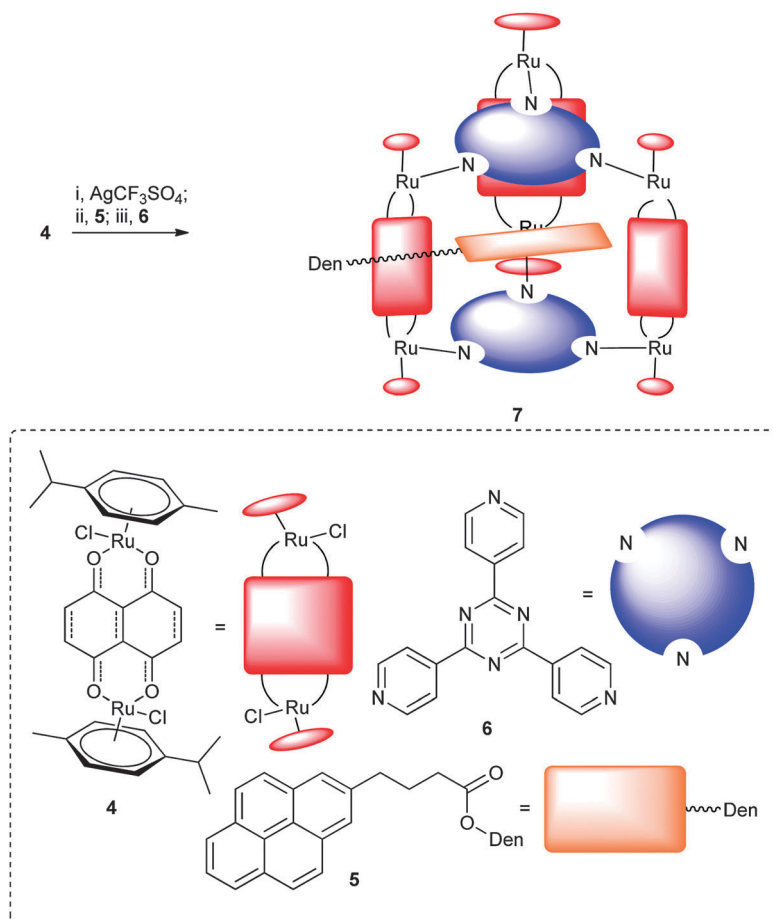


Fig. 3 Construction of host-guest ensemble 7.

compounds is not widespread but the cytotoxicity of different metal complexes, including organometallic complexes is attracting increasing attention,^{2,13} the aforementioned supramolecular constructs being cases in point.^{8–10} Reviews of the area appeared in 2011 comparing and contrasting numerous approaches, readers are directed to those for more details of the historical context.^{13,14}

During 2011 Mendes *et al.* reported on a range of metal containing complexes, including an organometallic ruthenium complex **13** (Fig. 5).¹⁵ The author looked at PARP-1 inhibition alongside the ability of their complexes to competitively bind the zinc-finger motif. The authors supported the hypothesis that the ability to displace zinc from the PARP-1 zinc finger decreases PARP-1 activity. Among the compounds tested **13** showed synergistic effects when combined with Cisplatin on certain cell lines, *i.e.* when administered at a non-cytotoxic dose with Cisplatin a synergistic cytotoxicity increase was observed. The authors suggested reduced repair of DNA bearing platinum due to partial PARP-1 inhibition by ruthenium could be the origin of this, noting that other mechanisms could not be ruled out.

In a report by Nowak-Sliwiska *et al.* variation of ligand structure delivered compounds **14** and **15** (Fig. 5),¹⁶ which displayed antiangiogenic activity.

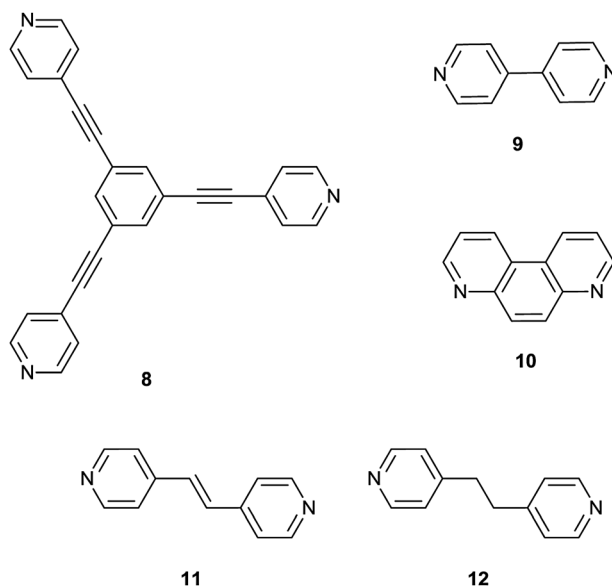


Fig. 4 Nitrogen containing linkers 8–12.

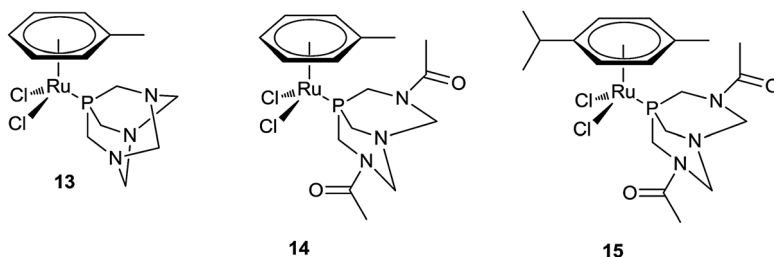


Fig. 5 Ruthenium arene complexes 13–15.

Hanif *et al.* examined saccharide derived phosphinite variants of **13**, and probed the effect of varying chloride for oxalate and malonate ligands. Among the compounds tested the acetonide protected series **16** to **18** (Fig. 6) were compared. The authors found anticancer activity to be linked to the ease of displacing the ancillary ligand with water, as such the more stable oxalato and malonato complexes, **17** and **18** respectively, showed less activity than the dichloride **16**.¹⁷

The renaissance of triazole formation by copper catalysed, so-called, *click chemistry* was discussed in the previous volume of this series.¹⁸ Triazoles have the potential to act as ligands for metals, and one such example where a *click* constructed 1,2,3-triazole derivative forms part of an organometallic complex, with anticancer activity, is given in Fig. 7 (**19**).¹⁹ Adding to the suite of organometallic ruthenium complexes reported in 2011 as potential anticancer agents **19** benefits from the *click* construction offering potential for rapid diversification. Whilst the organometallic complex **19** was not the most active (in these tests) the authors showed analogous coordination compounds with good activity and were able to correlate activity to rates of aquation (exchange of Cl for water in the case of **19**).

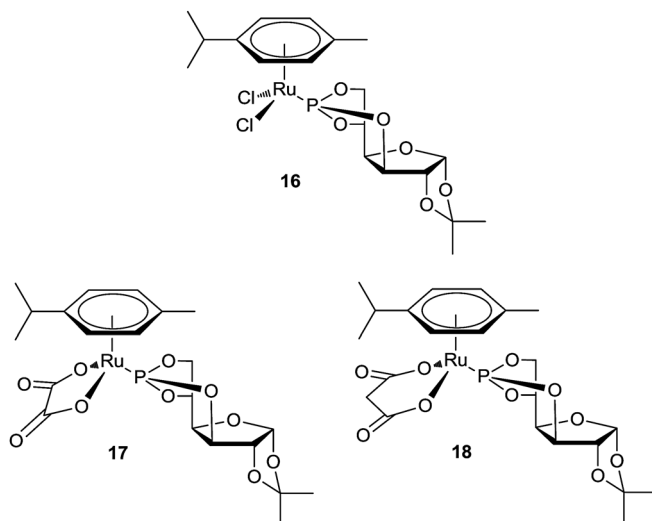


Fig. 6 Ruthenium arene complexes 16–18.

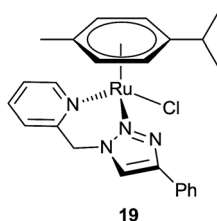


Fig. 7 Ruthenium triazole arene complex 19

Sadler and co-workers have continued their ground breaking studies into organometallic anticancer complexes and have reported ruthenium **20**,²⁰ osmium **21**,²¹ **22**,²² and iridium **23** (Fig. 8)²³ complexes in the last year operating by complementary mechanisms.

Organometallic reagents and catalyst

Asymmetric catalysis is given excellent coverage elsewhere in this volume,²⁴ and numerous reviews have appeared in 2011 covering more specialist areas which might also be of interested to readers of this chapter.²⁵

A mainstay of organometallic chemistry are organolithium reagents, the potential dangers associated with handling organolithiums without proper training remain at the forefront of our minds.²⁶ However, in trained hands organolithium reagents can be used safely and to great effect in organic synthesis; to this end a report by Hevia and Mulvey bring to readers' attention the important role of salt effects.²⁷ Whilst their report brings together information from the preceding years it's worth noting in this compendium of 2011 literature since some of the enhanced activity and reactivity effects obtained by addition of lithium chloride offer profound advantages and reaction variance (see references contained therein for historical details), indeed lithium chloride can play a cooperative role in some cases. Weng *et al.* demonstrated another cooperative salt effect in a different reaction,²⁸ where a silver salt was

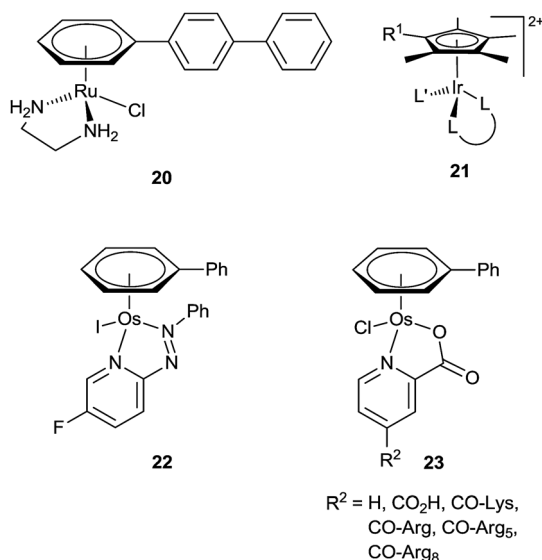
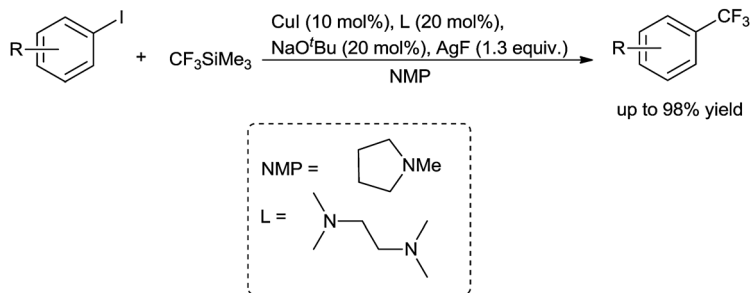


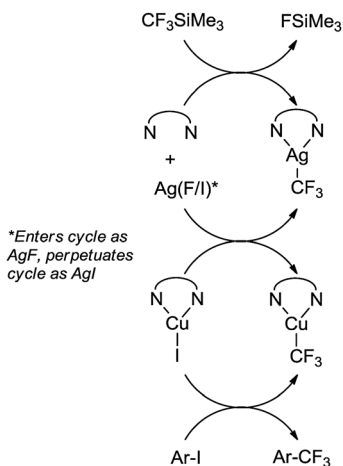
Fig. 8 Sadler and co-workers' ruthenium, iridium and osmium complexes 20–23.

important in affecting copper catalysed trifluoromethylation reactions of aryl iodides, a range of reaction conditions were probed and substrate scope investigated for the reaction above (Scheme 1).



The authors carried out numerous control experiments in order to unveil a plausible mechanism for the reaction. They proposed that the trimethyl(trifluoromethyl)silane trifluoromethylates ligated silver species exchanges a CF_3 for iodine on a ligated copper, which in turn delivers the CF_3 to the aryl iodide and regenerates the copper iodide intermediate (Scheme 2).

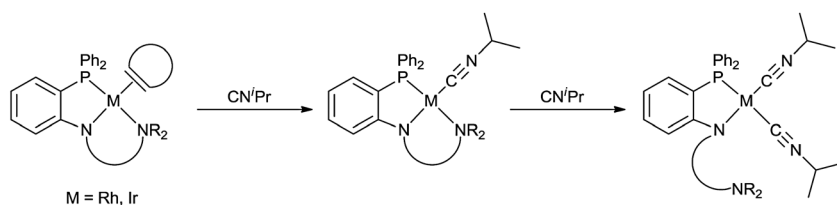
In catalysis the ability to modulate activity as well as stabilise reactive catalytic species is an important consideration. Tuneability of ligands, essentially tweaking electronic properties, is an important approach in catalysis. This is normally achieved by having a ligand or family of ligands that are easily accessible and readily interchangeable for forming catalytically active metal complexes for a given reaction. Hemilabile ligands, ligands which under the correct conditions, or in the desired scenario, dissociate and re-associate a catalytic active metal allow for unmasking and masking catalytically active sites on metals whilst attenuating



Scheme 2 Catalytic cycle of trifluoromethylation.

unwanted side reactions. Linder *et al.* reported the facile synthesis of a series of hemilabile anionic PNN' ligands.²⁹ An anionic N⁻ forms the centre of the pincer which is flanked by a phosphine and a hemilabile tertiary amine.

Rhodium and iridium complexes were studied by a variety of techniques including DFT, IR and NMR spectroscopy in order to delineate the role and mode of action of the hemilabile ligand, including actual and calculated reactions with isocyanates (Scheme 3). The authors discussed how their ligand set (Fig. 9) was adaptive to the demands of the system in question. The authors concluded that the extent of hemilability (the ease of with the third arm can dissociate) is governed by the nature of the amine nitrogen (alkyl *versus* aryl substituent) and ring size.



Ligands 24-26

Scheme 3 Isonitrile substitution onto rhodium and iridium complexes of ligands 24–26.

Hydroamination remains a versatile reaction for the introduction of nitrogen to unsaturated molecules. Metal containing catalysts offer the best activation regimes for this class of reaction, as reviewed by Patil and Singh³⁰ and Zi³¹ in 2011. Arrowsmith *et al.* reported on group 2 catalysed intramolecular hydroamination, the interplay between kinetic and thermodynamic parameters was discussed and the importance of ligand environments was key.³² That report concluded, asymmetric versions will offer challenges in substrate compatibility owing to the lability of these elements.

Chiral borato ligand **27** in combination with calcium gave a catalyst capable of catalysing a 5-*exo* trig hydroamination-cyclisation (Scheme 4),³³ with good yields. Although ee was not so high (up to 18% ee) the ligand architecture and challenging

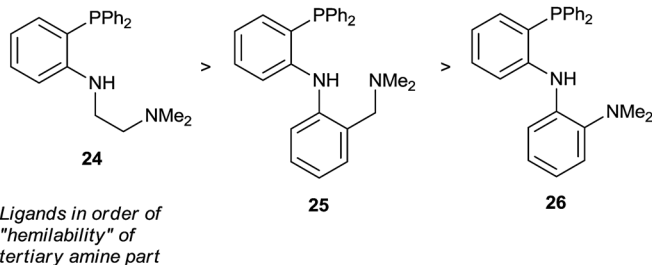
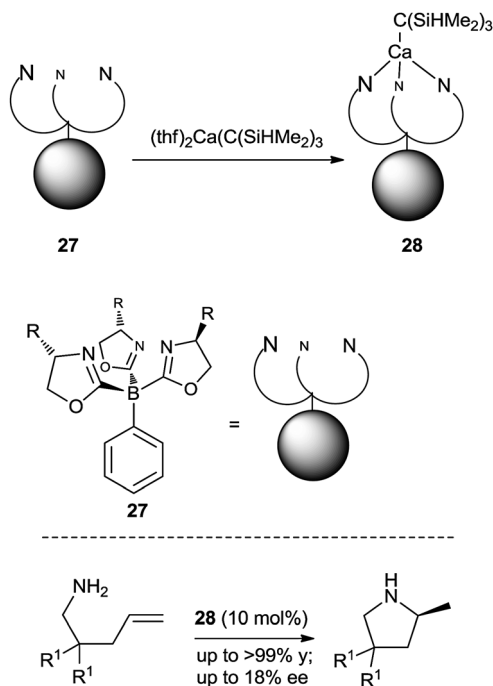


Fig. 9 Hemilability of ligands 24–26.

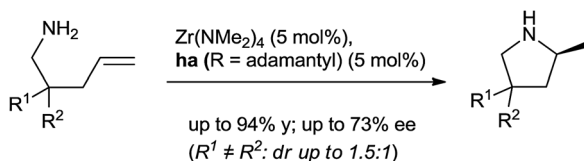
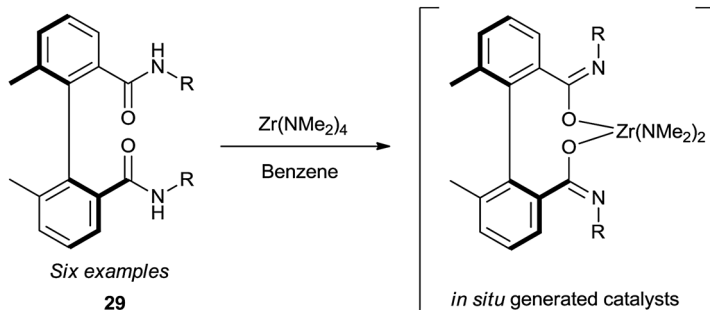
nature of this asymmetric transformation make this example worthy of note. Amidate ligands **29** displaying atropisomerism were used in zinc catalysed hydroamination of a similar reaction (Scheme 5),³⁴ which gave higher ee.



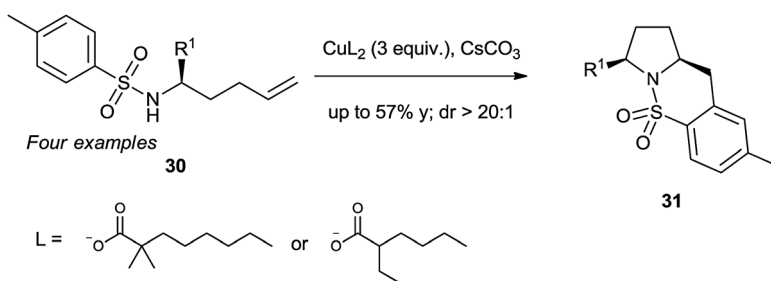
Scheme 4 Hydroamination catalysed by calcium complex **28**.

An exciting carboamination reaction of compounds **30** (Scheme 6) has been reported where bicyclic products **31** were obtained from a copper mediated reaction. It is worth highlighting diastereomer ratios were highest when R¹ was equal to H.³⁵

A noteworthy organometallic catalyst appearing in the surveyed literature include a triazole derived oxidation catalyst **11**, (Fig. 10).³⁶ Triazoles (formed from so-called "click reactions") continue to be of significant interest in the organometallic arena, ruthenium pincer complexes (**33**, Fig. 10),³⁷ and triazole carbene complexes carbene (**34**, Fig. 11),³⁸ all warrant attention.



Scheme 5 Hydroamination catalysed by a zirconium complex derived from ligand **29**.



Scheme 6 Copper mediated carboamination.

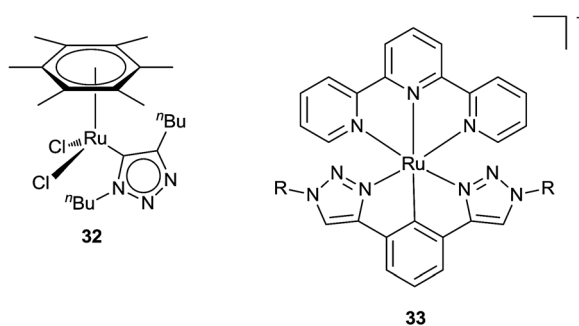


Fig. 10 Ruthenium complexes **32** and **33**.

In addition to the triazole derived metathesis catalyst **34**, another metathesis catalyst, (**35**, Fig. 11), was separately reported with an amide-type stabilising carbene ligand. The effect of the amide-type carbene was compared and contrasted to NHC and phosphine ligands in a report by Moerdyk and Bielawski.³⁹

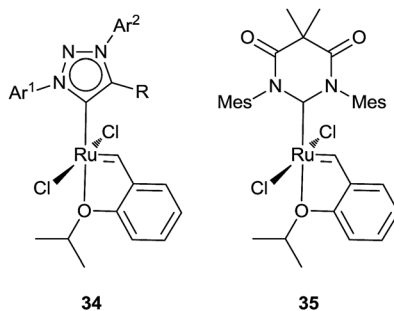


Fig. 11 Ruthenium complexes **34** and **35**.

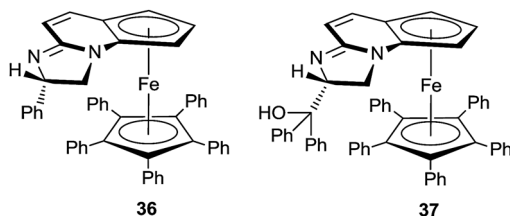
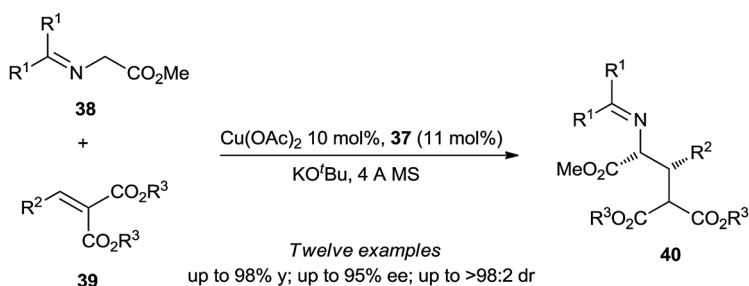


Fig. 12 Deng and co-workers' metallocene catalysts **36** and **37**.

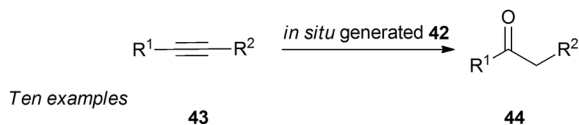
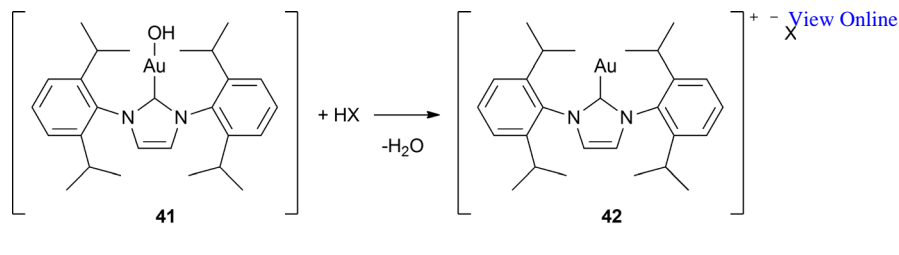
Ferrocene derivatives, as ever, feature heavily in the organometallic literature and their use in catalysis continues to be developed. One of the authors of this chapter co-authored two papers where ferrocene derivatives were used as catalysts during the period surveyed. The kinetic resolution of bulky heteroaryl secondary alcohols catalysed by **36** (Fig. 12),⁴⁰ and the 1,4 addition of **38** to **39** to give **40** was reported, catalysed by related ferrocene derivative **37** (Fig. 12, Scheme 7).⁴¹



Scheme 7 1,4 addition catalysed by metallocene **37**.

Nun *et al.* employed a gold hydroxide carbene **41** as a precatalyst which, upon treatment with HSbF_6 , delivered **42** which was used as an *in situ* generated catalyst for the transformation of a variety of alkynes **43** into ketones **44** (Scheme 8).⁴²

To round off this section a catalyst not for organic synthesis but water oxidation is discussed. Blackmore *et al.* developed a water oxidation catalyst using organometallic precursors.⁴³ Electrodeposition from aqueous solutions of Cp^* iridium complexes, compounds **45** and **46**, (Fig. 13), gave fine control over loading and



Scheme 8 Gold hydroxide carbene **41** catalyses the transformation of alkyne **43** to ketone **44**.

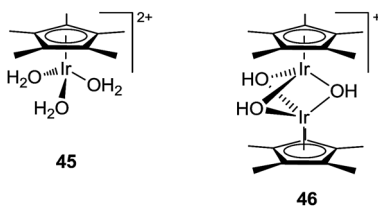


Fig. 13 Iridium complexes **45** and **46**.

preparation conditions of iridium oxide. The authors commented on how this method of preparing electrochemical oxidation catalysts is both direct and simple and importantly the properties are not dissimilar to the classically prepared, sputtered iridium oxide catalysts.

Dye sensitised solar cells

Background

The Dye-sensitized Solar Cell (DSC) is a low-cost photovoltaic technology and promising alternative to conventional inorganic materials-based approaches. For about two decades the material of choice for the sensitizer was based on ruthenium complexes and this aspect of DSCs has been reviewed in 2011.⁴⁴ Although dominated by organic dyes, the development of ruthenium sensitizers for dye-sensitized solar cells still attracts significant attention.

The most utilised design still revolves around the [Ru(dcbpy)(NCS)₂] fragment reminiscent of the original N3 dye (Fig. 14). The coordination sphere around the ruthenium is then completed with a bidentate ancillary ligand used to tune the photophysical (visible absorption) and electrochemical (energy levels) of the complex and make it suitable for sensitizing mesoporous titanium oxide, TiO₂, in the DSC.

A classical approach is to use a bipyridine-based ancillary ligand with extended π-system to improve absorption properties of the complex and harvest more of the incoming sunlight. The dyes **47** and **48** have the solubilising chains on the position 3,4 and 3 of the thiophene moiety to increase solubility.⁴⁵ Compared to N3,

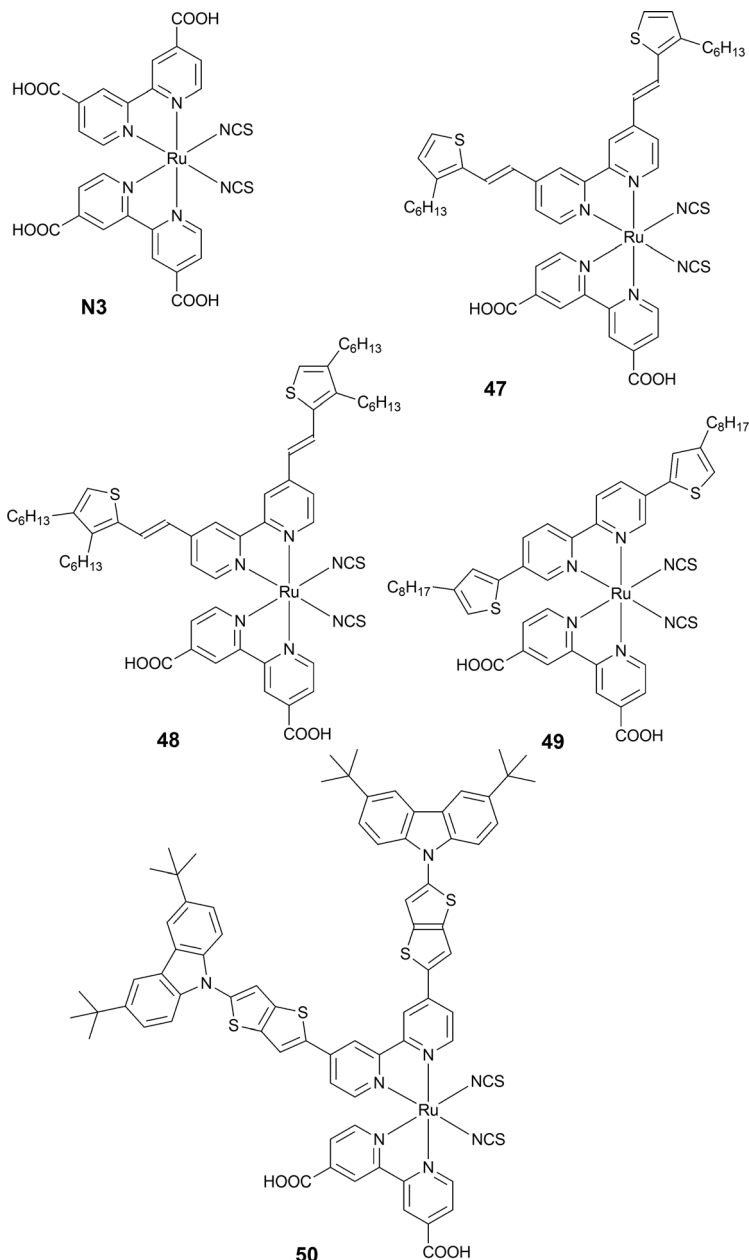


Fig. 14 Structures of the standard dye N3 and of dyes **47–50**.

the UV-visible absorption of the dyes is 15 nm red-shifted and the absorption coefficient is increased to $21,700 \text{ M}^{-1} \text{ cm}^{-1}$, resulting in solar-to-electrical energy conversion efficiency (η) of 7.7% for **48** compared to 6.05% for N3 (Fig. 14),⁴⁵ a similar design without the alkyl chains has also been reported, dye **49**.⁴⁶ The dye **50** possess carbazole-substituted thieno[3,2-*b*]thienyl antennas on the bipyridine, which results in absorption peak at 555 nm with absorption coefficient of $22400 \text{ M}^{-1} \text{ cm}^{-1}$.⁴⁷

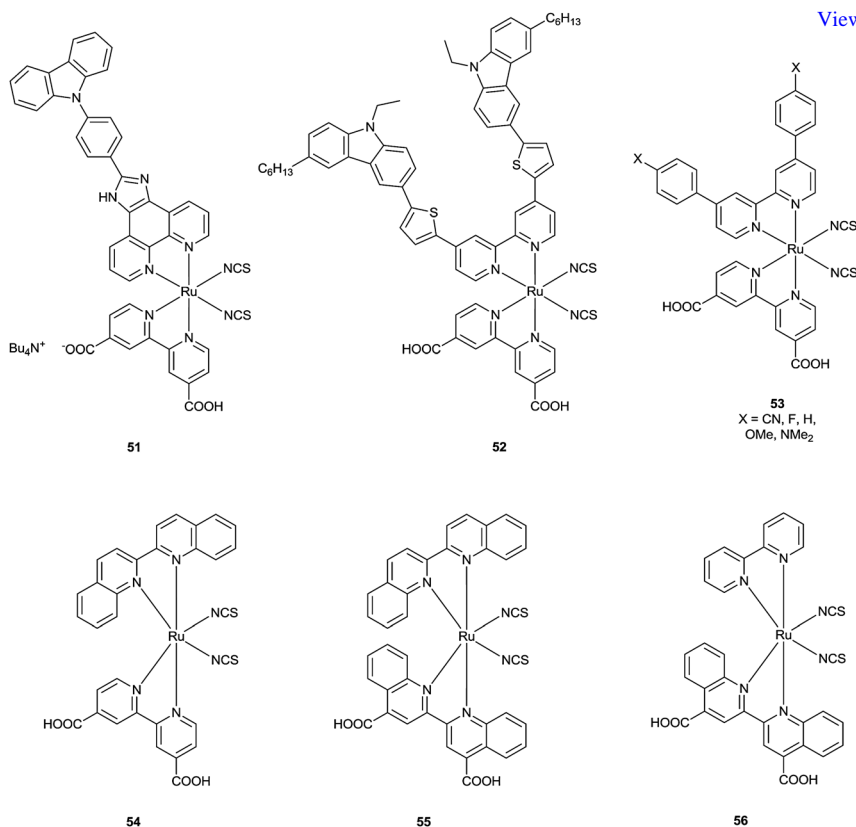


Fig. 15 Structures of dyes **51–56**.

The complex **51** has an imidazo-phenanthroline ligand bearing a phenylcarbazole moiety and achieved $\eta = 6.98\%$, compared to $\eta = 6.47\%$ for a N3-based device, mainly due to improved open-circuit voltage.⁴⁸ The sensitizer **52** performs poorly alone, although displaying improved absorption properties in solution compared to N3, due to the extended π -conjugation in the ancillary ligand.⁴⁹ Nevertheless, when employed as cosensitizer with an organic dye, devices with up to $\eta = 10.2\%$ were possible, while N3 in the same configuration achieved only 8.69% conversion efficiency. Han *et al.* reported an interesting systematic study of the impact of remote substituents on the properties of ruthenium complexes **53** and their performances in DSC.⁵⁰ The ancillary ligand is 4,4'-di(*p*-X-phenyl)-2,2'-bipyridine where X is cyanide –CN, fluorine –F, hydrogen –H, methoxy –OMe or dimethylamino –NMe₂ substituents. Changing the substituent from the electron-donating –NMe₂ to the electron-withdrawing –CN results in systematic bathochromic shifts of the absorption maxima. Optimum device performances were obtained for –OMe with $\eta = 8.3\%$, better than the N3-based device ($\eta = 7.8\%$). To enhance the red absorption, Johansson *et al.* have systematically studied biquinoline as ligand with lowered π^* orbitals compared to bipyridine in complexes **54**, **55**, and **56** (Fig. 15).⁵¹

Organometallics in DSCs

Ruthenium sensitizers based on cyclometallated ligands have gained significant attention in 2011. The rationale behind cyclometallation is to replace the mono-dentate and

ambidentate NCS to improve stability, while gaining the capability to fine tune the properties of the complex by grafting various substituents on the cyclometallated moiety, which is not possible with NCS (Fig. 16). Bomben *et al.* reported the use of bidentate ppy (ppy = 2-phenylpyridine) derivatives as direct replacement of the two NCS groups. With complex **57**, they report 7.3% conversion efficiency under

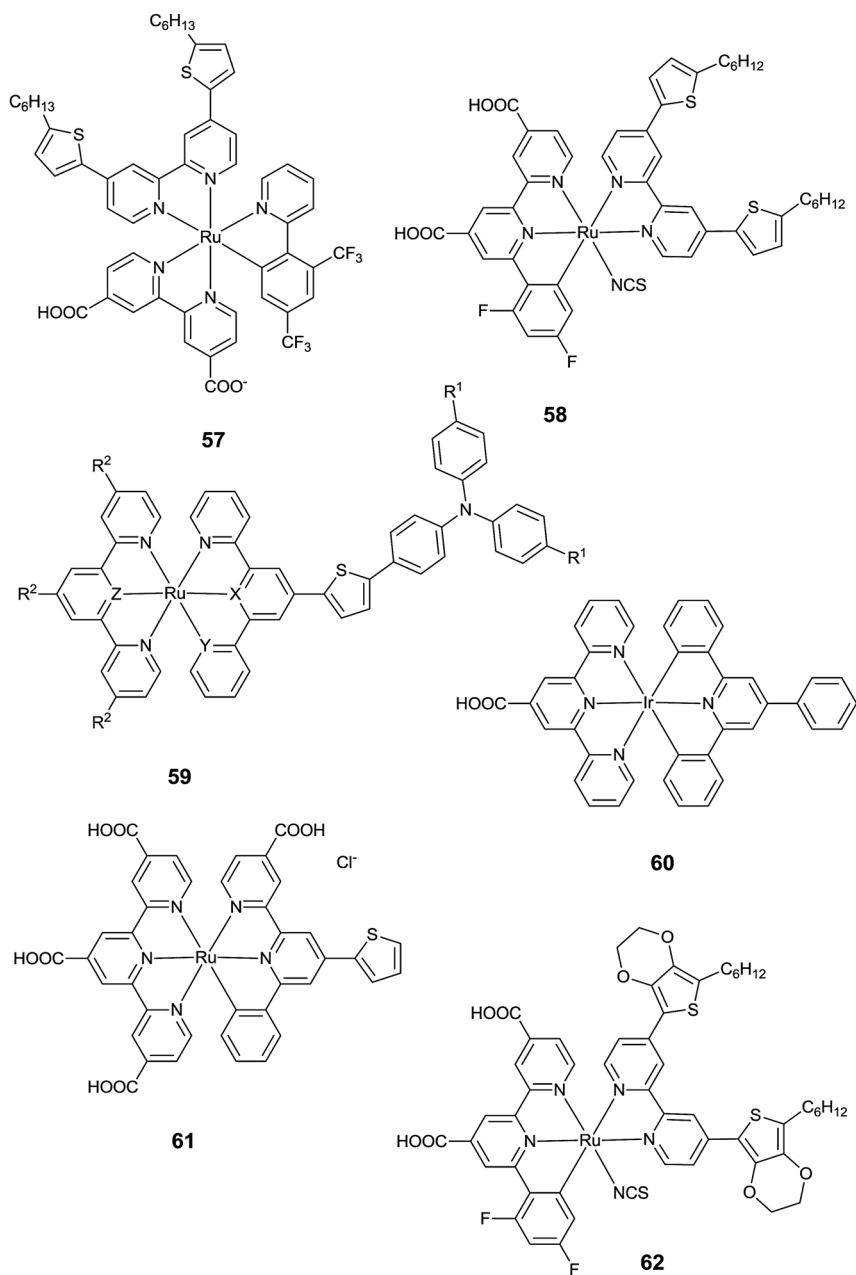


Fig. 16 Structures of cyclometallated ruthenium dyes **57–62**.

one sun illumination.⁵² The same group reported on the performance optimisation of the same family of complexes.⁵³ In complexes **58** and **62**, the cyclometallated moiety was introduced in a terdentate ligand, resulting in remarkable stability of 91% after 1000 hours light soaking at 60 °C.⁵⁴ Complex **61** was used in conjunction with cuprous iodide as electrolyte additive to reach $\eta = 5.7\%$.⁵⁵ Complex **59** was the base for a systematic study of the impact of the substituents and the cyclometallation position on ruthenium complex properties and performances in DSC.⁵⁶ A bis-cyclometallated iridium complex **60** was also reported for DSC.⁵⁷ Copper is another attractive metal for preparing sensitizers for DSC.⁵⁸

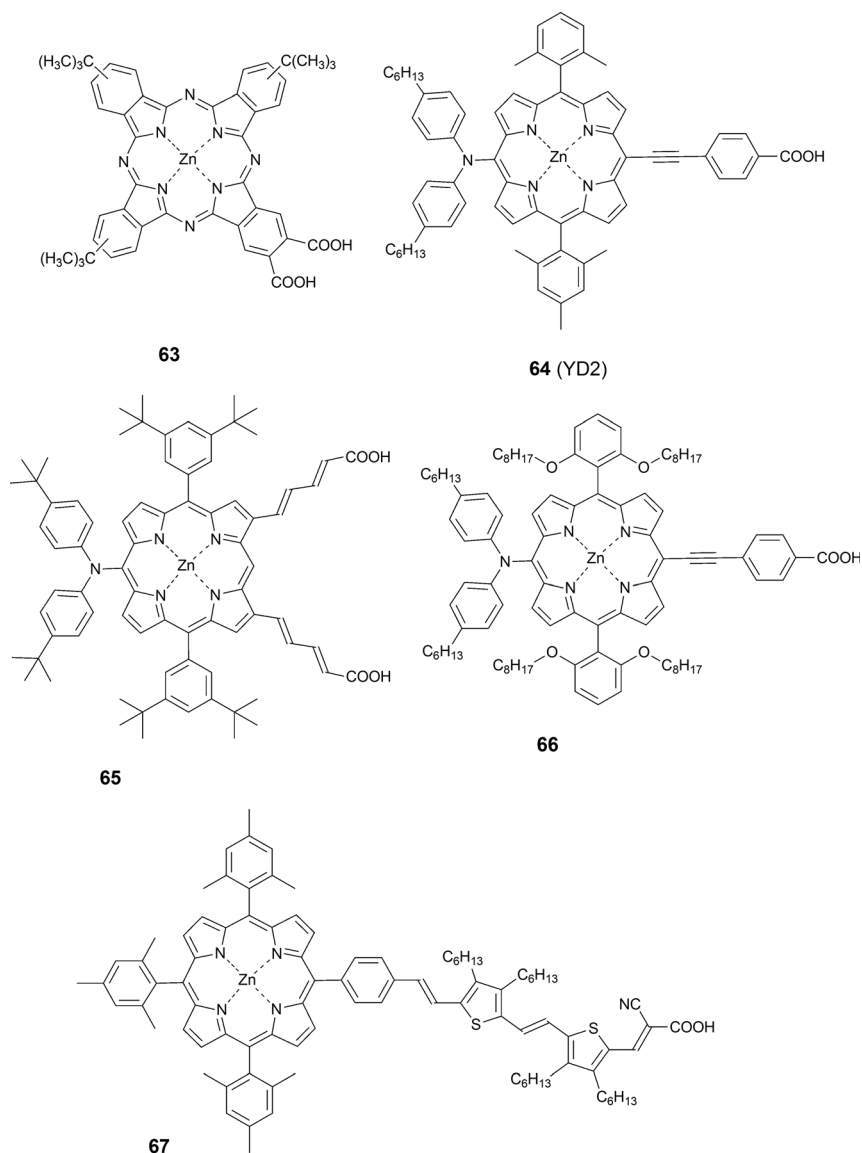


Fig. 17 Structures of dyes **63–67**.

Although sometimes classified as organic dyes, metallated porphyrins and phthalocyanines are an important class of sensitizers for DSC due to the remarkable absorption properties of these molecules. The dye **63** has been studied in detail,⁵⁹ and a systematic study of alkynyl-aromatic groups as replacements for the diphenylamine in dyes similar as **64** was reported.⁶⁰ The dye **65** is based on a similar design as **64** (YD2) with a diphenylamine directly attached to the porphyrin core, and possess two anchoring carboxylic acid groups.⁶¹ When used as co-sensitizer, a variant of **66** enabled 12.3% efficiency.⁶² The phthalocyanine **63** was studied to observe the impact of the anchoring group on the performances and stability of DSC.⁶³ The zinc porphyrin **67** having an oligothiophenevinylene moiety shows a three-fold increase in conversion efficiency compared to the parent dyes (Fig. 17).⁶⁴

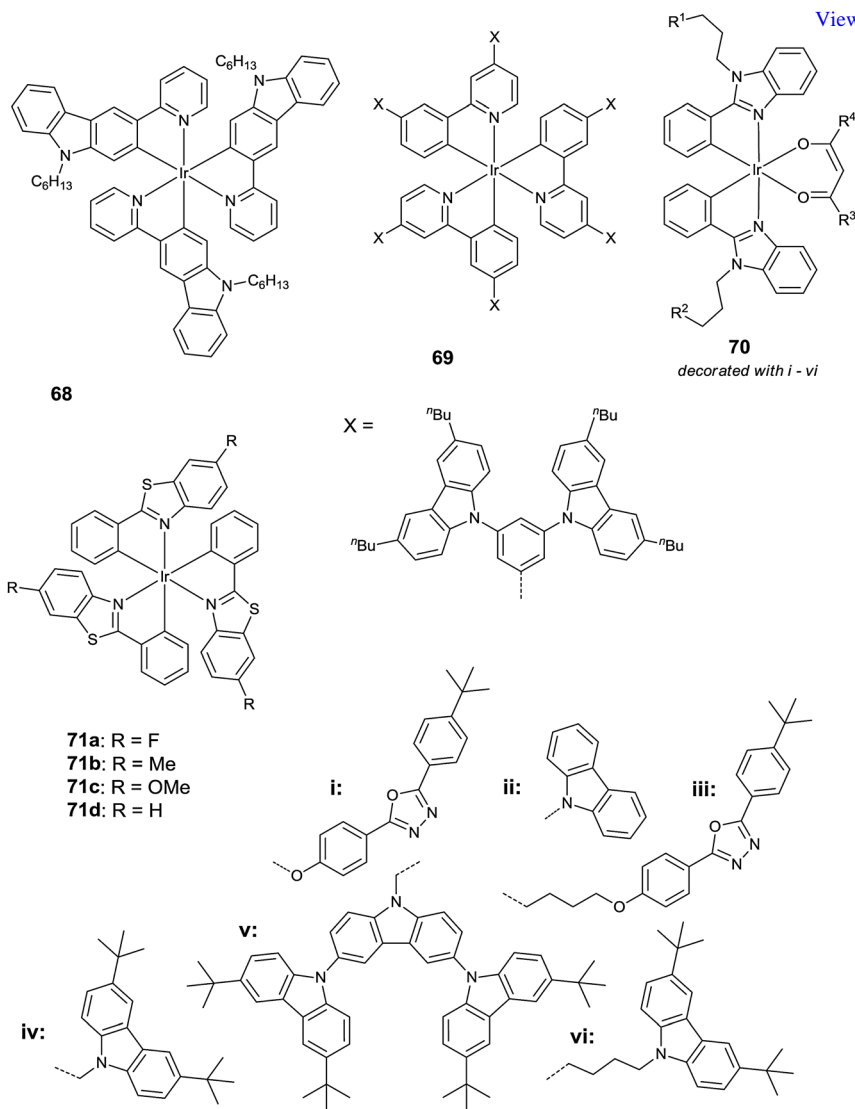
Besides the sensitizer, the redox shuttle is a paramount component of the DSC. Transition metal complexes have been explored as replacements of the iodide/triiodide classical couple.⁶⁵ Ferrocene⁶⁶ and copper⁶⁷ complexes have been explored. Nevertheless cobalt complexes are still the most studied transition-metal-based redox shuttles, in particular in combination with organic dyes.⁶⁸ As the electrochemical properties of the complexes are easily tuned, the impact of the driving force of the sensitizer regeneration has been studied,⁶⁹ and have also been used in p-type DSCs.⁷⁰ They could be successful with ruthenium-based sensitizer only when long alkyl chains were present on the sensitizer to avoid a detrimental electrostatic interaction between the cobalt complex and the ruthenium complex.⁷¹ Cobalt complexes have also been used as chemical dopant for solid-state DSCs.⁷²

Electroluminescence with metal complexes

Electroluminescence from phosphorescent metal complexes is a promising approach for low-cost and efficient future lighting. General aspects of the materials involved have been reviewed in 2011,⁷³ as well as reviews focused on white light emitting devices.⁷⁴

Due to their unique photophysical properties, high phosphorescent quantum yield, general photo-, electro- and chemical stability, and emitted colour tunability over the entire visible spectrum, cyclometallated iridium(III) complexes are still the most studied family of phosphorescent emitters for electroluminescence. A series of complexes based on **68** with various substituents on the pyridine was used to explore the effect of molecular dipole moment on the charge transport in solution processed devices.⁷⁵ Compound **69** possesses six carbazole-based dendrons to efficiently suppress intermolecular interactions in neat films resulting in photoluminescence quantum yield comparable to those obtained in diluted solution.⁷⁶ Complex **70** was substituted with various combinations of the charge transporting groups (i–vi, Fig. 18) in a self-hosting approach.⁷⁷ Varying the number of hole transporting carbazole and electron transporting oxadiazole allows for the optimisation of charge transport in the device for improved efficiency. Complexes **71a–d** have been used for efficient vacuum-processed orange OLEDs and reach efficiency of 87.9 cd A^{-1} .⁷⁸

The green emitting **72** and the orange emitting **73** feature a carbazole-modified acetylacetonate ancillary ligand and they achieve 36.8 cd A^{-1} and 10.8 cd A^{-1} , respectively, in vacuum-deposited OLEDs.⁷⁹ The amidinate ancillary ligand **dipba** (dipba = *N,N'*-diisopropylbenzamidinate) was used with difluorophenyl-pyridine **74** and benzoquinoline **75** main cyclometallated ligand for complexes highly phosphorescent in the solid state and with improved hole–electron transporting properties.⁸⁰ Hsieh *et al.* have reported a series of complexes (*e.g.* complex **76**, $\lambda_{\text{max}} = 456 \text{ nm}$) based on two



carbene-based main ligands and an anionic bidentate ancillary ligand from a deprotonated pyrazole or imidazole heterocycle for blue electroluminescence.⁸¹ Blue electroluminescence has been achieved with complexes **77–79** that are based on an unconventional tripodal bicyclicmetallated phosphite ligands, with CIE (x , y) coordinates as low as (0.169, 0.247) and efficiency as high as 11%.⁸² Efficient blue-green phosphorescent OLEDs, around 23 cd A⁻¹ at 1000 cd m⁻², have been obtained with complex **80** based on the tetraphenylimidodiphosphinate ancillary ligand.⁸³ With three fluorine substituents **81** results in greenish-blue electroluminescence.⁸⁴ The highly luminescent green emitting tetranuclear iridium complex **82** using cyanide bridging ligand is active as dopant in OLEDs and give green emission with excellent colour purity and maximum efficiency of about 10% (Fig. 19).⁸⁵

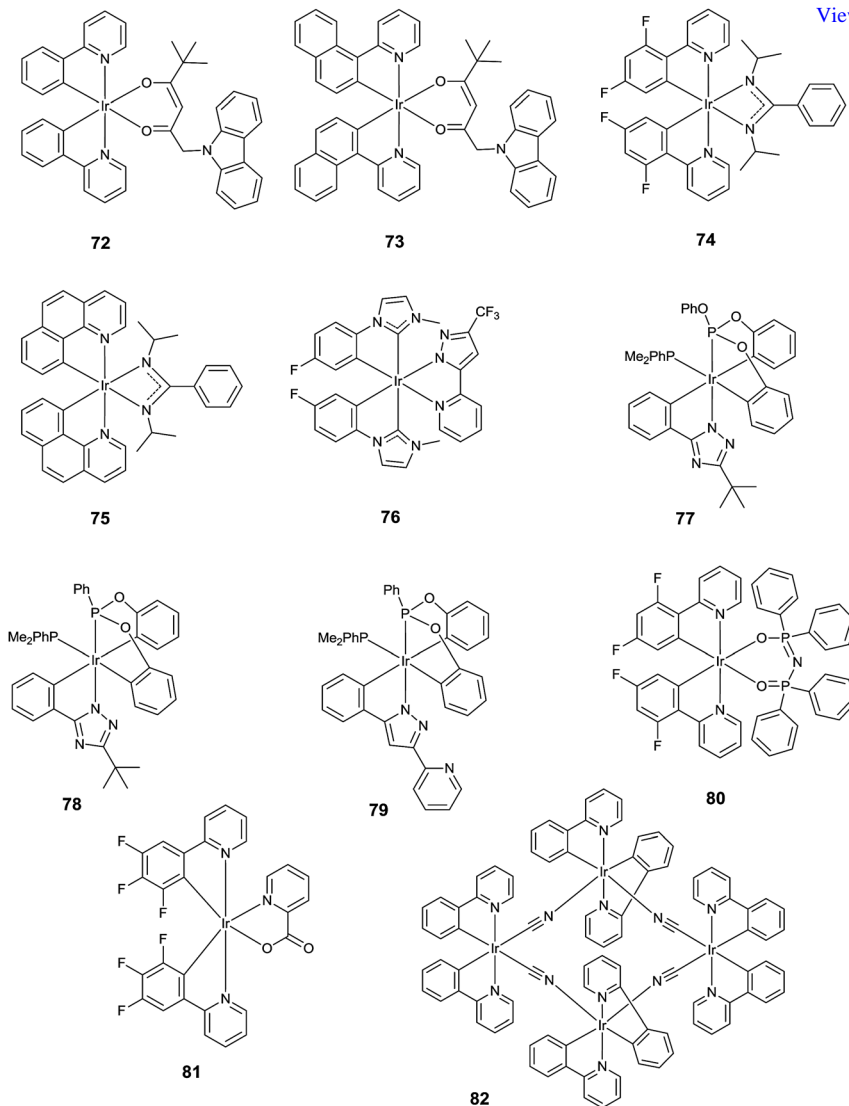


Fig. 19 Structures of emitters 72–82.

The complex **83** displays sharp red emission and devices using it reach efficiency of 25%.⁸⁶ Kim *et al.* studied complexes based on **84** and similar complexes with various alkyl groups to reduce self-quenching and fine tune the energy level, to achieve red OLED with 24.6% external efficiency.⁸⁷ Compound **85** is an efficient yellow phosphor with excellent colour purity and was used with the greenish-blue emitter **81** to obtain white light OLED.⁸⁸ Compound **86** (**dpa** = 2,2'-dipyridylamido) gives green emitting devices with high current efficiency of 123.5 cd A⁻¹.⁸⁹ Deep-red OLED are obtained with **89** although with limited efficiency.⁹⁰ Compound **87** gives excellent orange OLED with 27.2% external efficiency, and was used for white OLED with 24.1% external efficiency with CIE (*x*, *y*) coordinates (0.32, 0.41).⁹¹ Introduction of a phenylsulfonyl group provides interesting electron injection and

transporting properties.⁹² Complex **88** is based on cyclometallated naphthyl-phosphine chelate for orange electroluminescence (Fig. 20).⁹³

Charged iridium complexes are attracting attention for the use in light-emitting electrochemical cells (LECs) (Fig. 21). Pendant phenyl rings on the neutral ancillary

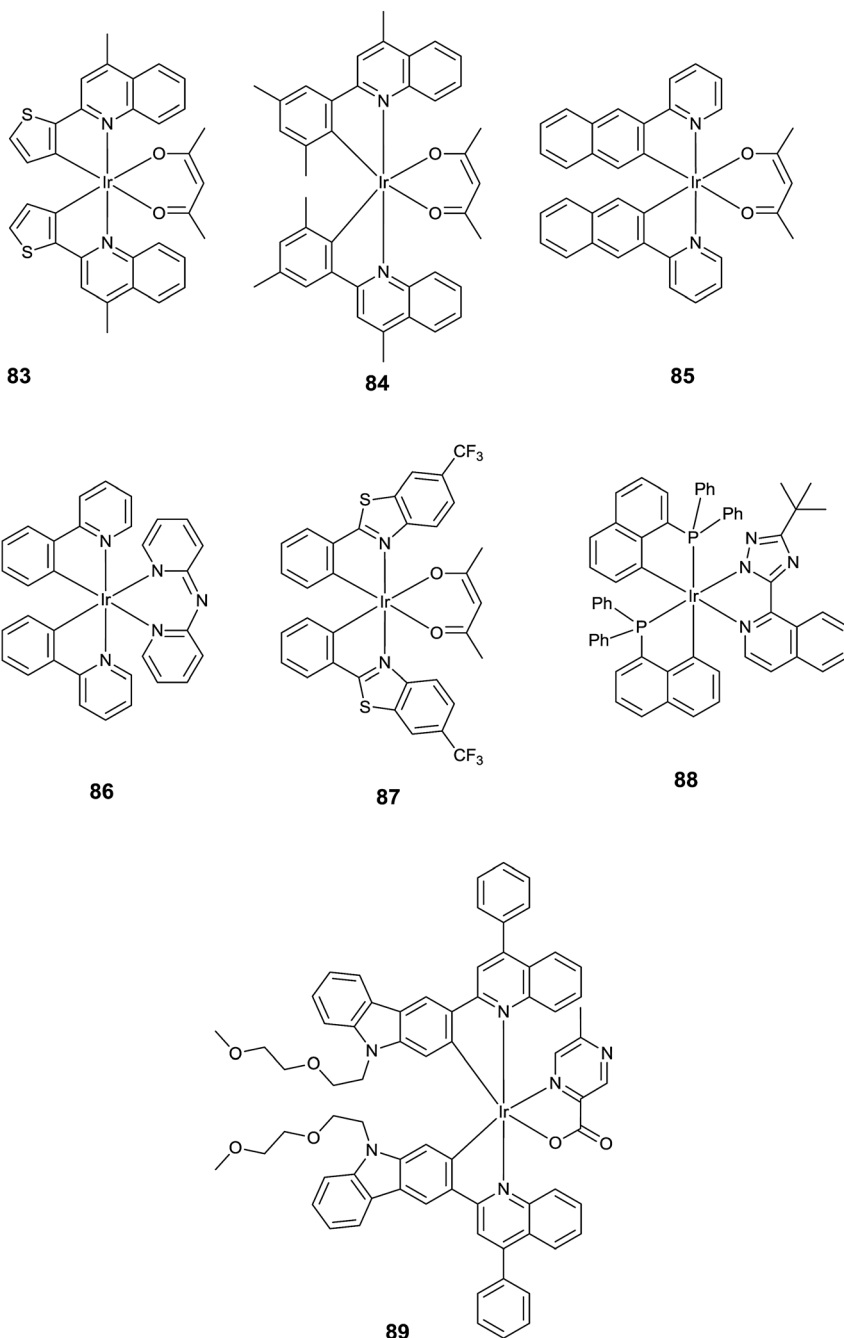


Fig. 20 Structures of emitters 83–89.

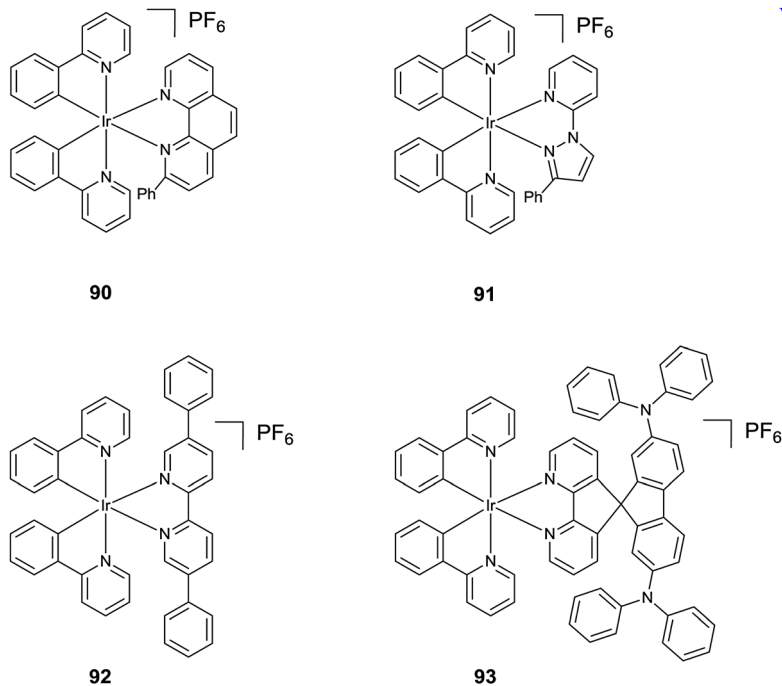


Fig. 21 Structures of charged emitters **90–93**.

ligands continue to provide an efficient design motif for improved efficiency, in the orange⁹⁴ for **90** and greenish-blue⁹⁵ for **91**. Introduction of bulky substituents is an approach used to reduce intermolecular interaction in LEC devices as in **92**.⁹⁶ Furthermore such bulky substituents can be advantageously replaced with functional bulky groups such as the electron donating diphenyl amines in **93**.⁹⁷ Soft salts based on cyclometallated iridium complexes for electroluminescence were also reported.⁹⁸

Platinum-based cyclometallated complexes are also often used for electroluminescence. Their square-planar geometry allows the formation of excimers useful for single dopant white electroluminescence (Fig. 22).⁹⁹ A series of complexes based on **94** (**95–97**) have been prepared, the substituents on the **acac** ligand do not modify the electronic properties of the complexes, but impact efficiency of the device through influencing the molecular packing.¹⁰⁰ Complex **98** was used for low efficiency red OLEDs.¹⁰¹ The dinuclear platinum complex **99** results in white emitting OLED while the same device based on the corresponding mononuclear platinum complex emits orange-red.¹⁰² A series of complexes based on **100** with a range of substituents on the ligand is reported.¹⁰³ **101** was used in an interesting study about the differences between photo- and electroluminescence.¹⁰⁴ Electroluminescence based on complexes with terdentate ligands using triazole, **102**,¹⁰⁵ and tetrazole¹⁰⁶ have been reported. Platinum porphyrins, such as **103**,¹⁰⁷ are used for NIR¹⁰⁷ and red emitting OLEDs.¹⁰⁸

Copper complexes, a low-cost alternative to iridium and platinum complexes, are also gaining attention for OLEDs as neutral mononuclear¹⁰⁹ or polynuclear complexes.¹¹⁰

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Additional systems of interest from the 2011 literature are also discussed.

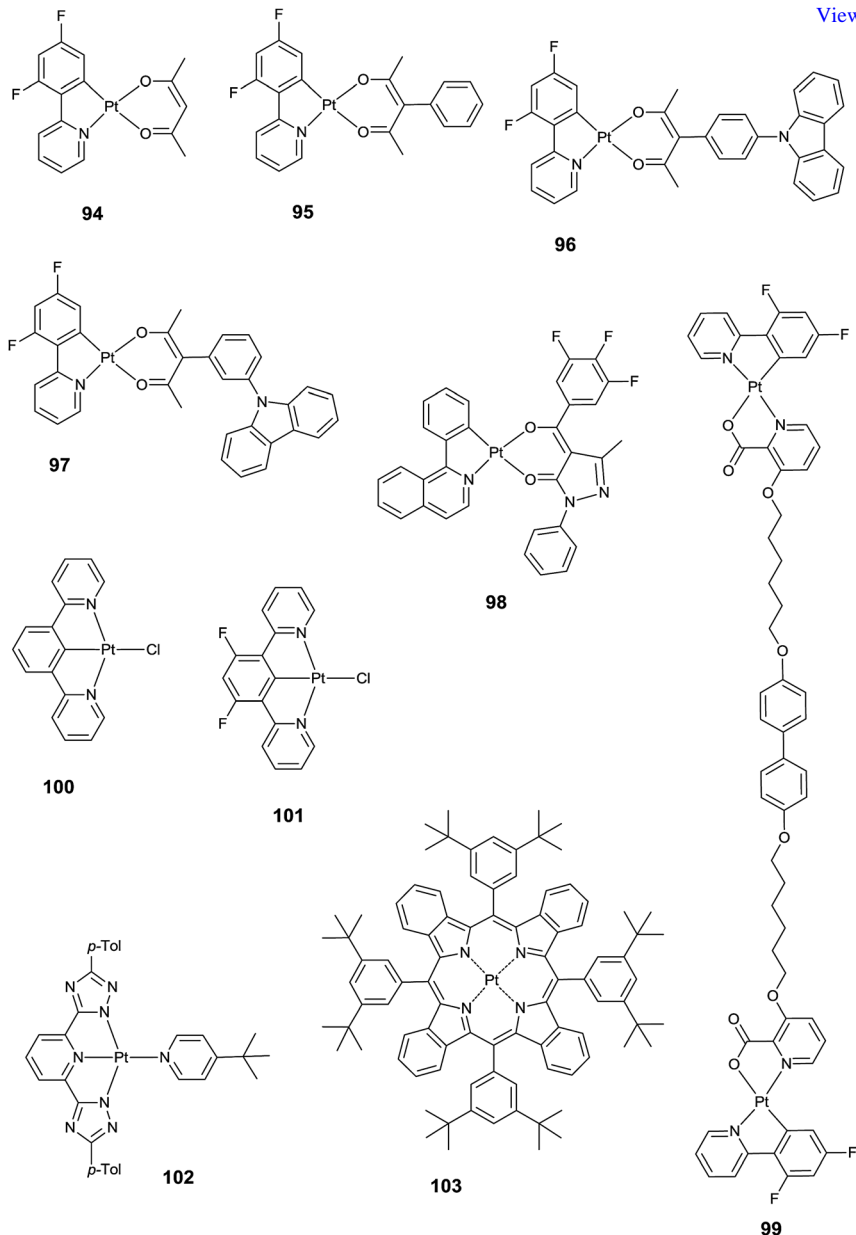


Fig. 22 Structures of emitters 94–103.

Organometallic compounds are used for bioimaging. Often the materials are based on standard cyclometallated iridium complexes similar to those reported in a previous section (OLEDs) with only small structural variations.¹¹¹ Rhenium(I)-tricarbonyl complexes can be used as a replacement of iridium complexes due to their ease of synthesis. The series of complexes **104–106** is based on a bis(phenanthridinylmethyl)amine (bpm) ligand (Fig. 23).¹¹² The complexes are orange emitters ($\lambda_{em} = 570$ nm). Complexes **104** and **106** have been tested for preliminary cell microscopy on

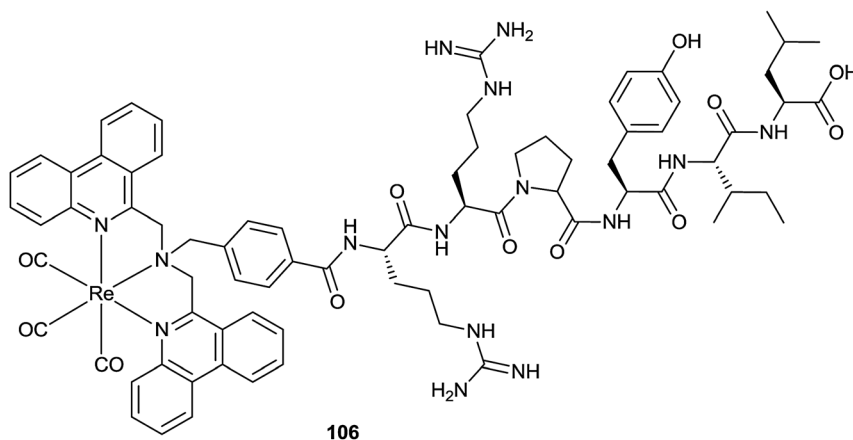
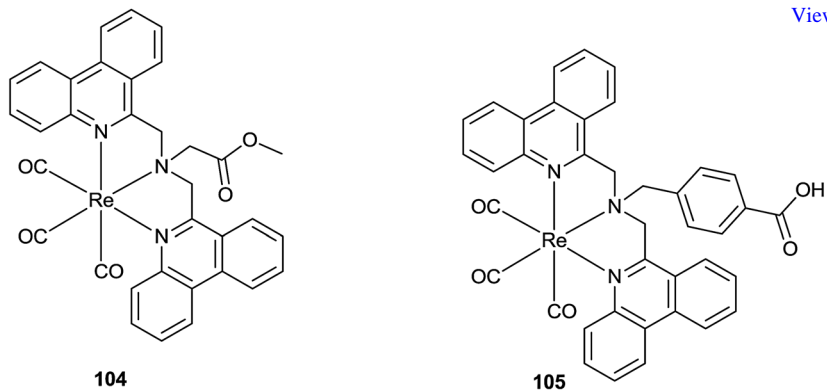
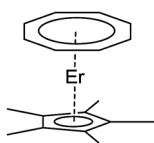


Fig. 23 Structures of emitters **104–106**.

various cell lines and both are taken up well and distributed in the cytosol. Interestingly, the peptide derivative **106** seems to present some degree of selectivity in the uptake mechanism, which is attributed to the attached peptide.

Hydrogen is a promising green fuel, however it is necessary to store it for use as a fuel. Theoretical calculations have been used to explore the possibility of using a vanadium-based organometallic complex, $V(C_3H_3)$, for a calculated uptake capacity up to 10.07 wt%.¹¹³

Single-ion magnets are crucial for the development of information storage, quantum computing, and molecular spintronics. A simple erbium complex, **107** (Fig. 24),



107

Fig. 24 Erbium organometallic complex **107**.

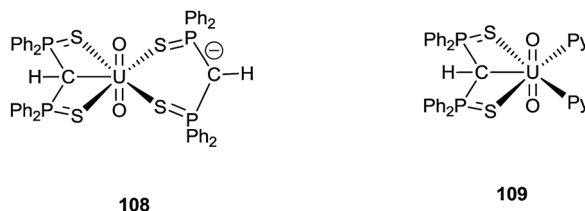


Fig. 25 Organouranium compounds **108** and **109**.

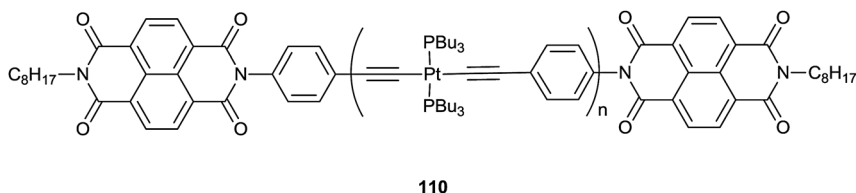


Fig. 26 Platinum containing molecular wire **110**.

sandwiched by two different aromatic ligands, cyclooctatetraenide ($C_8H_8^{2-}$, COT) and pentamethylcyclopentadienide ($C_5Me_5^-$, Cp*), presents a magnetisation hysteresis and two thermal relaxation processes with a high energy barrier.¹¹⁴

Uranium chemistry is undergoing significant development recently. Novel complexes, **108** and **109** (Fig. 25), have been reported having a rare example of uranium-carbon bond.¹¹⁵

Finally, a molecular wire, **110** (Fig. 26), incorporating platinum-acetylide repeating unit capped by naphthalene diimide end groups can transport both electron and triplet excitons.¹¹⁶

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