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Comparison of structure, log*P* and P388 cytotoxicity of some phenyl and ferrocenyl cyclic chalcone analogues. Application of RP-TLC for log*P* determination of the ferrocenyl analogues[¥]

Research Article

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Abstract: Cyclic chalcone analogues (**2**-**5**) and their ferrocenyl counterparts (**6**-**10**) were synthesized and their logP and P388 cyctotoxity were investigated. The structures of the newly synthesized compounds were confirmed by IR ^{1}H and ^{13}NMR spectroscopy. Comparison of conjugation and stereochemistry of the respective derivatives showed similar characteristics compared to ones with some higher degree of conjugation in the ferrocenyl series. Comparison of log*P* of the ferrocenyl derivatives determined by a validated RP-TLC method showed the ferrocenyl derivatives to have higher logP_{TLC}. The results demonstrate that the differences in three dimensional shape, conjugation and lipophilicity do not have strong influence on the P388 cytotoxicity of the investigated phenyl (**1**-**5**) and ferrocenyl (**6**-**10**) enones.

Keywords: *Cyclic chalcone analogues • Ferrocene • RP-TLC logP • P388 cytotoxicity • IR • NMR*

1. Introduction

Chalcones (**1**) are intermediary compounds of the biosynthetic pathway of a very large and widespread group of plant constituents known collectively as flavonoids [2]. Among the naturally occurring chalcones and their synthetic analogs, several compounds displayed cytotoxic (cell cell growth inhibitor) activity towards cultured tumor cells [3,4]. Recently we have synthesized and investigated *in vitro* antineoplastic activity of several synthetic chalcones and cyclic chalcone analogues with a variety of substitutions on their cynnamylidene moiety possessing *in vitro* cytotoxic activity against murine and human cancer cells [5-7]. Structure and stereochemistry of the synthesized compounds have been investigated by X-ray and NMR methods [5-9]. Our results showed that three dimentional shape, nature of the aromatic substituent and lipohilicity of the molecules could be considered to contribute to the variation of cytotoxic properties of the compounds [5-7].

In vivo investigation of the *in vitro* most active cyclic chalcone derivative, (*E*)-2-(4'-methoxybenzylide)-1-

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¥*Benzylidenebenzocyclanones, Part IX (Part VIII, see [1a]), and Study on ferrocenes, Part 24*

benzosuberone, showed the compound to effectively reduce the DMBA-induced expression of some onco/suppressor genes [10]. This observation led to recognition of strong CYP1A inhibitory effect of the compound and its structurally related analogues [11]. Investigation of (*E*)-2-(4'-methoxybenzylide)-1-benzosuberone and its methyl-substituted analogue showed the compounds to modify the cellular thiol status and cause apoptosis in *in vitro* Jurkat cells culture [12]. Investigation of their effects on mitochondrial function showed the compounds to decrease the reduced glutathione (GSH) level and to inhibit respiration [13].

Ferrocene is a lipophilic, electron donating entity, with no hydrogen donor or acceptor properties [14]. The ferrous ion can undergo reversible oxidation–reduction and the nature of the substituents on the ferrocene ring has a marked influence on this process [15]. The incorporation of a metallocene into compounds with medicinal application was rare prior to the 1980's. The low cytotoxicity of ferrocene, coupled with its lipophilicity and its electrochemical behavior, suggested that this compound could yield interesting results if incorporated into a known drug [16]. Indeed, there are several reported observations on increased efficacy of ferrocene analogues of known drugs. Ferrocifen, the ferrocene analogue of tamoxifen, has shown antiproliferation effects in both hormone-dependent and hormone-independent tumors. Tamoxifen and other established chemotherapeutic agents fail to exhibit such antiproliferative effects in both hormone-dependent and hormone-independent cell lines [17].

Ferrocene possesses immune-stimulatory properties [18]. Ferrocene derivatives have been reported to show antitumor activities [19]. Both effects have been reported to be associated with the redox activity of the ferrocene moiety [18,19]. Earlier results suggested that ferrocene analogues of benzylidene ketones could have as high cytotoxicity against the P388 leukemia cells as their benzylidene counterparts [20]. On the other hand, ferrocene analogues of chalcones have been reported as effective antiplasmodial agents [21]. Being conjugated molecules, replacement of the benzylidine for a ferrocenemethylene moiety can change the electron distribution, stereochemistry, reactivity and lipophilic character of the previously investigated benzylidene compounds.

Lipophilicity is one of the important physico-chemical parameters characterizing compounds related to their biological activities. Lipophilicity, measured as log*P*, is a key property in quantitative structure activity relationship (QSAR) studies. Several methods have been developed for experimental determination and prediction (calculation) of log*P* values [22]. In our previous publications we reported log*P* values of the cyclic chalcone analogues (*E*)- and (*Z*)-2-(X-benzylidene)-1-indanones (**1**), -tetralones (**2**) and -1-benzosuberones (**3**) determined by RP-TLC. Their experimental log*P* values were in the range 2.9 - 5.2 and had characteristic differences depending on ring size, configuration and substitution pattern of the compounds [23a-c].

Since replacement of phenyl moieties of biologically active compounds for ferrocenyl moieties is a frequently used approach in present-day drug research, there is an interest in a fast and reliable method for determination of lipophilicity of such compounds with predictable higher log*P* values than their phenyl counterparts [24]. In the present work we report the modification of our previously applied RP-TLC method optimized for determination of log*P* of chalcones and the above mentioned chalcone analogues. Details of the modified RP-TLC method are useful to determine $\log P_{TLC}$ value of lipophilic compounds, which were found to have values as high as log*P* 6, as shown in this paper.

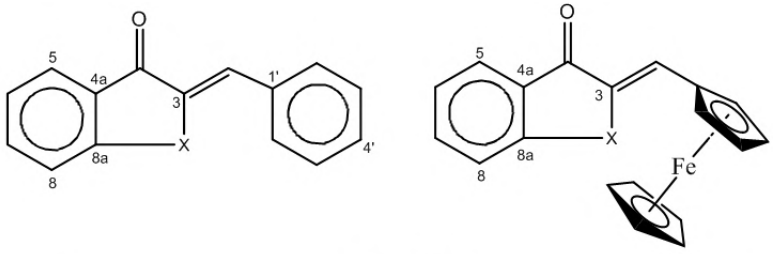
As a continuation of our previous work on the investigation of the antiproliferative effect of chalcones and their cyclic analogues, here we report on the comparative P388 cytotoxic activity of some (*E*)-2-benzylidene-1-benzocyclanones (**1**-**5**) and their ferrocene counterparts (**6**-**10**) (Fig. 1). In order to search for the physico-chemical characteristics that could be related to the observed biological effects, we compared stereochemistry, polarity of the enone fragment and lipophilic character of the two series of compounds.

2. Experimental procedure

2.1. Materials

(E)-2-phenylmethylene-1-benzocyclanones (**1**-**5**) and their ferrocenyl analogues (**6**-**10**) were synthesized by base-catalyzed Claisen-Schmidt condensation as described earlier [5-8,25]. Their structure and stereochemistry were determined by X-ray and NMR methods [5-8,25]. All the compounds were purified by column chromatography over Kieselgel 60 with toluene or toluene-ethanol 99:1 (v/v) as mobile phase. The compounds had melting points in accord with their literature values. The purity of the compounds was checked by TLC (silica gel, toluene or toluene-ethanol 4:1 (v/v) as mobile phase) and GC (HP 5890 Series II gas chromatograph; 25 m (0.32 mm x 0.17 µm) HP-5 capillary column).

The ^{1}H- and ^{13}C-NMR spectra were recorded in CDCl3 or DMSO-d_6 solution in 5 mm tubes at room temperature, on a Bruker DRX 500 spectrometer at 500 (^{1}H) and 125 (^{13}C) MHz, with the deuterium signal of



1	X = -(CH_2)-	**6**
2	X = -$(CH_2)_2$-	**7**
3	X = -$(CH_2)_3$-	**8**
4	X = -(OCH_2)-	**9**
5	X = -(SCH_2)-	**10**

Figure 1. Structure of the investigated compounds.

the solvent as the lock and TMS as internal standard. The standard Bruker microprogram NOEMULT.AU to generate NOE was used. DEPT spectra were run in a standard manner, using only the = 135° pulse to separate CH/CH_3 and CH_2 lines phased "up" and "down", respectively. The 2D-HSC spectra were obtained by using the standard Bruker pulse program HXCO.AU.

Spectral data of **4** and **5**: IR (in KBr disc, cm^{-1}): νC=O: 1667 (**4**), 1660 (**5**); γC_{Ar}H (condensed ring): 737 (**4**), 734 (**5**); γC_{Ar}H (phenyl): 755 (**4**), 762 (**5**); γ$C_{Ar}C_{Ar}$ (phenyl): 699 (**4**), 689 (**5**); νC-O: 1146 (**4**). ^{1}H-NMR (in $CDCl_3$, δ, ppm, δ_{TMS} = 0 ppm): CH_2, *s* (2H): 5.31 (**4**), 4.10 (**5**); CH (olefinic), *s* (1H): 7.86 (**4**), 7.78 (**5**); ArH-5, *dd* (1H): 8.01 (**4**), 8.19 (**5**); ArH-6, ~*t* (1H): 7.04 (**4**), 7.22 (**5**); ArH-8, ~*d* (1H): 6.94 (**4**), 7.29 (**5**); ArH-7 and ArH-3'-5' (phenyl), *m* (4H): ~7.4 (**4**); ArH-7 and ArH (phenyl), *m* (6H): ~7.4 (**5**); ArH-2',6', ~*d*, (2H): 7.28 (**4**).

Compounds in the calibration set (Table 1) have been described elsewhere [23a]. Three compounds of the validation set (progesterone, diazepam, itraconazol) were of pharmacopoeial grade. Prostaglandin E1 ethyl ester (PGEE) was generously supplied by Sanofi-Synthelabo-Chinoin Pharmaceutical Works (Budapest, Hungary). Methanol was HPLC grade. Other reagents were of analytical grade.

2.2. Methods

2.2.1. Determination of logP by the RP-TLC method

RP-TLC determination of the log*P* of the chalcone and ferrocene analogs was performed on 20×20 cm plates pre-coated with 0.25 mm layers of silanized silica gel $60F_{254}$ (Merck, Germany; #5747). Before use, the plates were washed with methanol (ascending development) and dried. The samples were dissolved in 1:1 methanol-chloroform (c=2 mg mL^{-1}) and these solutions (2 μL; 4 μg compound) were spotted on the plate. Methanol-water, 65+35 (*v/v*), was used as mobile phase. After development the plates were dried and the chromatograms were assessed visually under UV illumination (λ=254 nm) and after spraying with iodine. The log*P* values of compounds were calculated by use of the calibration equation. Each logP_{TLC} value is an average from four parallel measurements.

2.2.2. Cytotoxicity screens

The *in vitro* cytotoxicity screens was undertaken using the protocol described before [26]. Statistical comparisons were made by u-test (p = 0.05).

3. Results and discussion

Cytotoxicity testing results of compounds **1**-**10** towards murine P388 cells (Table 1) showed the corresponding phenyl and ferrocenyl compounds to possess cytotoxicity of the same degree with statistically different (p = 0.05) IC_{50} values for the **1**/**6**, **3**/**8** and **4**/**9** pairs. Comparison of the tetralone (**2** and **7**) derivatives with those of the corresponding 4-chromanones (**4** and **9**) and thiochroman-4-ones (**5** and **10**) showed a somewhat lower cytotoxicity in the phenyl series (**2**,**4** and **5**) but a moderately higher cytotoxicity in the ferrocenyl series (**7**,**9** and **10**). Change in size of the cycloalkanone ring of **2** and **7** caused a decrease of cytotoxicity in the phenyl series (**1** and **3**) and an increased cytotoxicity in the ferrocenyl (**6** and **8**) series. Thus, compounds **2**, **6** and **9** were found to be the most active derivatives in the two series (Table 1).

Earlier, steric structures of compounds **1**-**3** [9] and **6**-**10** [25] were investigated in detail showing that respective phenyl and ferrocene analogues adopt similar conformations. It was found that the five-membered **1**

Table 1. *In vitro* IC50 values (μM) against P388 murine leukemia cells, vC=O IR frequencies (in 0.05 M $CHCl_3$ solution) and logP_{TLC} values of compounds **1**-**10**.

Compound-	IC_{50}	*v*C=O	logP_{TLC}	Compound-	IC_{50}	*v*C=O	logP_{TLC}
1	65.9±3.5	1696	3.63±0.06*	**6**	9.9±0.3	1690	4.62±0.15
2	13.8±1.0	1667	4.14±0.04*	**7**	22.5±5.4	1661	5.23±0.04
3	21.0±0.3	1663	4.48±0.05*	**8**	15.3±0.6	1659	5.36±0.05
4	32.3±1.8	1673	3.98±0.02	**9**	10.7±2.6	1668	4.79±0.05
5	19.9±4.7	1663	4.43±0.04	**10**	17.0±0.5	1656	5.35±0.06

**See [23a]*

Table 2. LogP_{SF} and logP_{TLC} values of the compounds of the validation set.

Compound	logP_{SF}	logP_{TLC}	Δlog*P*
Diazepam	2.77	2.70	0.07
Progesterone	3.54	3.45	0.09
PGEE	4.08	4.02	0.06
Itraconazol	5.66	5.50	0.16

is almost perfectly planar while in **2** and **3** the planes of the enone and the phenyl moieties deviate from planarity [9]. Investigation of transmission of substituent effects in the three series lead to the same conclusions [27a,b]. Our results were found to be in accordance with the previously published X-ray investigation results of **1** [28] and **2** [29]. The similar IR and NMR data of **1**-**3** and **6**-**8** indicated similar conformation of the conjugated moieties of the respective compounds [25]. Similar results were obtained by comparing the stereochemistry of some (*E*)-2-ferrocenemethylene- and (*E*,*E*)-2,5-diferrocenemethylene-1-cycloalkanones as well as their benzylidene analogues [30]. Comparison of X-ray crystal data of the pyrone derivative **4** [31] and its substituted analogues [8] with those of *(E)*-2-benzylide-1-tetralone **(2)** [8, 29] indicated similar spatial structure of the enone moieties in the two series [8]. Comparison of the ^{1}H and ^{13}C NMR data of **4** and **5** (see Experimental) with those of the analogous **9** and **10** [25] indicates similar conformation of the respective phenyl (**4**,**5**) and ferrocenyl (**9**,**10**) compounds, with a non-planar cinnamoyl moiety.

On comparison of the IR vC=O frequencies of the two series we can see that the frequencies of the benzylidene ketones (**1**-**5**) are 4-7 cm^{-1} higher than those of the respective ferrocene (**6**-**10**) ones (Table 1). The lower vC=O frequency of **6**-**10** is due to the higher mass of the ferrocene moiety and the higher degree of conjugation of the compounds [25].

Lipophilicity of compounds **4**-**10** was studied by a slight modification of the previously optimized RP-TLC method used for determination of log*P* of chalcones and some cyclic chalcone analogues [23a-c]. For the RP-TLC log*P* determination a calibration curve (log*P* = *a* R_M + *b*) was set up using the previously selected set of compounds (calibration set) with known logP_{SF} values (Fig. 1) [23a]. Based on the results of four parallel chromatographic runs of compounds of the calibration set, the following calibration equation was obtained:

$$\log P = 4.721\ R_M + 1.795 \quad (n = 6;\ r^2 = 0.9959;\ s = 0.049;\ F = 972) \qquad (1)$$

Afterwards, the optimized chromatographic system was validated for log*P* measurements. For this reason, four drug molecules of known log*P* values (diazepam, progesterone, PGEE and itraconazol) were tested. Comparison of their logP_{TLC} values obtained in this study with their logP_{SF} data measured by shake-flask method showed excellent agreement (Table 2).

Further on, these four compounds have also been involved into the calibration set. This produced the following calibration equation, based on four parallel experiments:

$$\log P = 4.7927\ R_M + 1.7897 \quad (n = 10;\ r^2 = 0.9992;\ s = 0.042\quad F = 10\,017) \qquad (2)$$

Eq. 2 was used for the logP_{TLC} determination of compounds **4**-**10**. The logP_{TLC} data obtained by our optimized and validated RP-TLC method are collected in Table 1.

The logP_{TLC} data can provide a good basis for evaluation of structure-lipophilicity relationship of the examined compounds. Comparison of the logP_{TLC} value of the (*E*)-2-benzylidene-1-tetralone (**2**) with that of its oxygen (**4**) and sulfur (**5**) analogues showed that replacement of a ring carbon atom for oxygen slightly decreased (Δ log*P* = -0.16) while for sulfur substantially increased (ΔlogP_{TLC} = 0.44) the lipophilicity in the phenyl series. Such a comparison of log*P* value of **7** with the respective **9** and **10** indicated a higher decrease (ΔlogP_{TLC} = -0.29) for **9** and a lower increase (ΔlogP_{TLC} = 0.44) for **10** of the logP_{TLC} values of the respective ferrocene analogues (Table 1).

Comparison of the logP_{TLC} values of the corresponding phenyl (**1**-**5**) and ferrocenyl derivatives (**6**-**10**) showed similar relative values in both series having the ferrocene analogues some higher lipophilicity in each pair. The average of difference in the logP_{TLC} values of the ferrocenyl and the respective phenyl derivatives is 0.94 log*P* unit, which is similar to the reported difference (1.15 log*P* unit) between the log*P* value of ferrocene (log*P* 3.28) and benzene (log*P* 2.13) [24].

4. Conclusions

Cyclic chalcone analogues (**2**-**5**) and their ferrocenyl counterparts (**6**-**10**) were synthesized and their log*P* and P388 cytotoxities were investigated. Comparison of conjugation and stereochemistry of the respective derivatives showed similar characteristics with ones with some higher degree of conjugation in the ferrocenyl series. Comparison of log*P* of the ferrocenyl derivatives determined by a validated RP-TLC method showed the ferrocenyl derivatives to have higher logP_{TLC}. These results demonstrate that the differences in three-dimentional shape, conjugation and lipophilicity not to have a strong influence on the P388 cytotoxicity of the investigated phenyl (**1**-**5**) and ferrocenyl (**6**-**10**) enones. Further investigations in this direction would be useful in identifying the target of action for these compounds and, ultimately, the design of more effective antitumor drugs.

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