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Electroreductive Intermolecular Coupling of Uracils with Aromatic Ketones: Synthesis of 6-Substituted and *cis*-5,6-Disubstituted 5,6-Dihydro-1,3-dimethyluracils and Their Transformation to 6-Substituted 1,3-Dimethyluracils, *trans*-5,6-Disubstituted 5,6-Dihydro-1,3-dimethyluracils, and 4,5,5-Trisubstituted 3-Methyloxazolizin-2-ones

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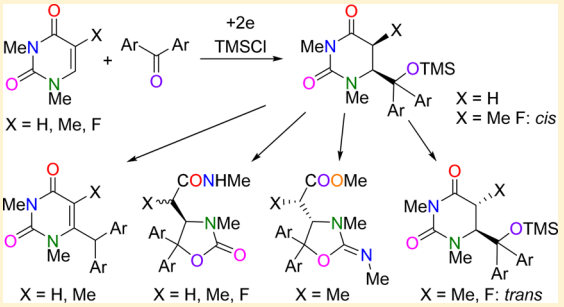
Supporting Information

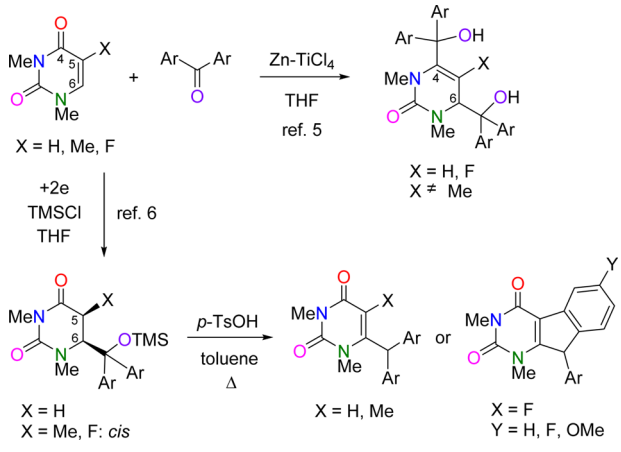
ABSTRACT: The electroreductive coupling of 1,3-dimethyluracil, thymine, and 5-fluorouracil with aromatic ketones in the presence of TMSCl gave 6-substituted and *cis*-5,6-disubstituted 5,6-dihydro-1,3-dimethyluracils. The dehydrotrimethylsiloxylation of the adducts afforded 6-substituted and 5,6-fused 1,3-dimethyluracils. The detrimethylsilylation of the adducts with TBAF or 1 M HCl−MeOH gave 4,5,5-trisubstituted 3-methyloxazolizin-2-ones or 3-methyloxazolizin-2-imines in addition to simply desilylated alcohols. The *cis*-5,6-disubstituted 5,6-dihydro-1,3-dimethyluracils were isomerized to the corresponding *trans*-isomers by heating in the presence of cat. DMAP. The *cis*- and *trans*-5,6-disubstituted 5,6-dihydro-1,3-dimethyluracils were assigned by the coupling constants $J_{5,6}$ of their ^{1}H NMR spectra.



INTRODUCTION

To date, a number of 5- and 6-substituted uracils have been investigated as pharmacologically and biologically active compounds, since they are analogues of primary nucleic acid bases.[1,2] Therefore, the selective synthesis of 5- and 6-substituted uracils attracts much interest from the synthetic chemists.[3,4] In this context, we reported the reductive two-to-one coupling of benzophenones with 1,3-dimethyluracils by low-valent titanium as the first example of the reductive coupling of uracils with carbonyl compounds (Scheme 1).[5] In addition, we recently reported the electroreductive one-to-one coupling between aromatic ketones and 1,3-dimethyluracils to give 6-substituted 5,6-dihydro-1,3-dimethyluracils and their transformation to 6-substituted 1,3-dimethyluracils (X = H, Me) or 5,6-fused 1,3-dimethyluracils (X = F).[6] It is noted that *cis*-5,6-disubstituted 5,6-dihydro-1,3-dimethyluracils were formed stereoselectively from 1,3-dimethylthymine (X = Me) and 5-fluoriuracil (X = F). In this paper, we report our further study on the electroreductive coupling of 1,3-dimethyluracils with aromatic ketones and the dehydrotrimethylsiloxylation of the adducts. Moreover, we found that the adducts can be transformed to 4,5,5-trisubstituted 3-methyloxazolizin-2-ones, 3-methyloxazolizin-2-imines, and *trans*-5,6-disubstituted 5,6-dihydro-1,3-dimethyluracils (Scheme 2). Successive ring-closure and opening of the adducts proceeded by treatment with TBAF in THF or HCl in MeOH to give *N*-methyl-2-(3-methyl-2-oxo-5,5-diaryloxazolidin-4-yl)acetamides (X = H, Me, F) or methyl 3-methyl-2-(methylimino)-5,5-diaryloxazolidin-4-yl)-propanoates (X = Me), respectively. Furthermore, *cis*-5,6-disubstituted 5,6-dihydro-1,3-dimethyluracils (X = Me, F) were

Scheme 1. Previous Works: Reductive Coupling of Uracils with Benzophenones



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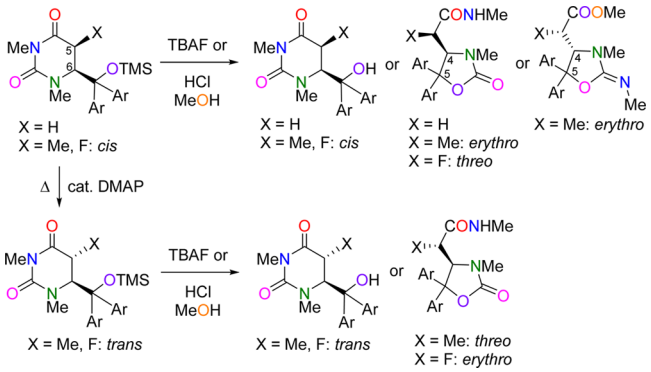
Scheme 2. Transformation of 6-Substituted (X = H) and *cis*-5,6-Disubstituted 5,6-Dihydro-1,3-Dimethyluracils (X = Me, F)



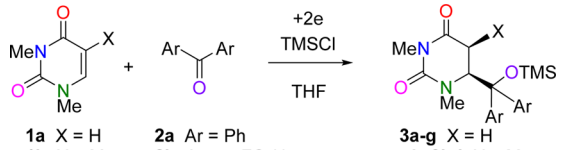
isomerized to the corresponding *trans*-isomers by heating at 150 °C in the presence of cat. DMAP. These results provide a new method for the stereoselective synthesis of *cis*- and *trans*-5,6-disubstituted 5,6-dihydro-1,3-dimethyluracils. The both geometric isomers were readily assigned by the coupling constants $J_{5,6}$ of their ^{1}H NMR spectra.

RESULTS AND DISCUSSION

Electroreductive Coupling of Uracils with Aromatic Ketones. The electroreduction of 1,3-dimethyluracils **1a**−**c** and benzophenones **2a**−**g** (2 equiv) was carried out in THF in the presence of TMSCl (5 equiv) and TEA (5 equiv) using a Pt cathode to give 6-substituted 1,3-dimethyl-5,6-dihydrouracils **3a**−**n** as the adducts (Table 1).[6] As the cathode material, Pt, Pb, Au, Ag, Cu, Zn, and Sn afforded almost the same yields of **3a** (72−77%) in the reaction of **1a** and **2a**. The presence of TMSCl is indispensable for the electroreductive coupling,[7] since no cross-coupled product was produced by the electroreduction of **1a** and **2a** in the absence of TMSCl; 1,1,2,2-tetraphenylethane-1,2-diol was obtained as an only product by the pinacol coupling of **2a**. On the other hand, the presence of TEA is not crucial for the reductive coupling but brought about steady results. The role of TEA is probably to neutralize hydrogen chloride generated from TMSCl and trace amounts of water remaining in the solvent and reagents. From 1,3-dimethylthymine (**1b**) and 1,3-dimethyl-5-fluorouracil (**1c**), *cis*-5,6-disubstituted 1,3-dimethyl-5,6-dihydrouracils ***cis*-3h**−**n** were produced with complete stereoselectivity (runs 8−14). The stereostructures of ***cis*-3h**−**n** were determined by X-ray crystallographic and ^{1}H NMR analyses (vide infra).

The electroreductive coupling of **1a**−**c** with alkyl aryl ketones **4a**−**e** was also effected under the same conditions (Table 2).[6]

Table 1. Electroreductive Coupling of Uracils with Benzophenones



1a X = H
1b X = Me
1c X = F
(1 mmol)

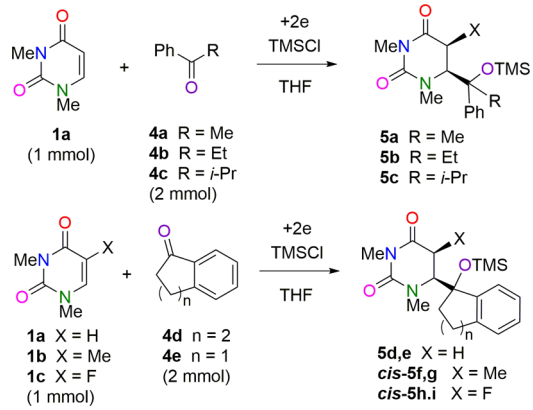
2a Ar = Ph
2b Ar = *p*-FC_6H_4
2c Ar = *p*-$MeOC_6H_4$
2d dibenzosuberone
2e dibenzosuberenone
2f anthrone
2g xanthone
(2 mmol)

3a-g X = H
***cis*-3h-j** X = Me
***cis*-3k-n** X = F

run	1	2	3	% yield of 3[a]
1	**1a**	**2a**	**3a**	77[b]
2	**1a**	**2b**	**3b**	62[b]
3	**1a**	**2c**	**3c**	57[b]
4	**1a**	**2d**	**3d**	65[b]
5	**1a**	**2e**	**3e**	58
6	**1a**	**2f**	**3f**	44
7	**1a**	**2g**	**3g**	52
8	**1b**	**2a**	***cis*-3h**	63[b]
9	**1b**	**2b**	***cis*-3i**	45[b]
10	**1b**	**2c**	***cis*-3j**	80[b]
11	**1c**	**2a**	***cis*-3k**	67[b]
12	**1c**	**2b**	***cis*-3l**	54[b]
13	**1c**	**2c**	***cis*-3m**	49[b]
14	**1c**	**2d**	***cis*-3n**	68[b]

[a]Isolated yields. [b]Reported data in ref 6.

Table 2. Electroreductive Coupling of Uracils with Alkyl Aryl Ketones



1a
(1 mmol)

4a R = Me
4b R = Et
4c R = *i*-Pr
(2 mmol)

5a R = Me
5b R = Et
5c R = *i*-Pr

1a X = H
1b X = Me
1c X = F
(1 mmol)

4d n = 2
4e n = 1
(2 mmol)

5d,e X = H
***cis*-5f,g** X = Me
***cis*-5h.i** X = F

run	1	4	5	% yield (dr) of 5[a]
1	**1a**	**4a**	**5a**	52 (50:50)[b]
2	**1a**	**4b**	**5b**	49 (55:45)[b]
3	**1a**	**4c**	**5c**	71 (67:33)[b]
4	**1a**	**4d**	**5d**	75 (73:27)[b,c]
5	**1a**	**4e**	**5e**	52 (55:45)[c]
6	**1b**	**4d**	***cis*-5f**	76 (85:15)[c,d]
7	**1b**	**4e**	***cis*-5g**	53 (67:33)[c,d]
8	**1c**	**4d**	***cis*-5h**	60 (70:30)[c,d]
9	**1c**	**4e**	***cis*-5i**	42 (55:45)[c,d]

[a]Isolated yields. [b]Reported data in ref 6. [c]Major isomers were *erythro*. [d]Both isomers were *cis*.

All of the products **5a**−**i** were formed as mixtures of two diastereomers. Fortunately, it was confirmed by X-ray crystallographic analysis of the corresponding detrimethylsilylated alcohols as described below that the major isomers of **5e** (X = H) and **5h** (X = F) were *erythro* and *cis-erythro*, respectively, and the minor isomer of **5g** (X = Me) was *cis-threo*. These results suggest that the major isomers of **5d**−**i** formed from cyclic ketones, 1-tetralone (**4d**) and 1-indanone (**4e**), were *erythro* (runs 4−9) and the both isomers of **5f**−**i** (X = Me, F) were *cis* (runs 6−9). The *cis*-stereoconfiguration of both isomers of **5f**−**i** was also supported by ^{1}H NMR analysis (vide infra).

As described in the previous report,[6] the *cis*-stereoselective formation of **3h**−**n** and **5f**−**i** can be explained by the assumption that the protonation to the 5-position of 6-substituted silyl enol

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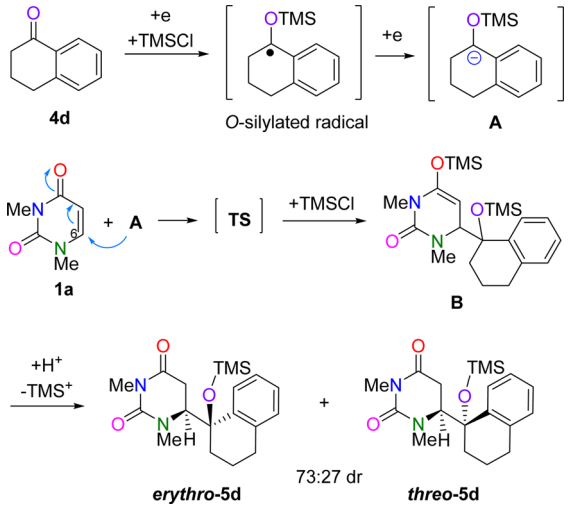
ethers occurs from the less-hindered side, that is, the opposite side of the 6-substituent predominantly. Next, the presumed reaction mechanism of the electroreductive coupling of **1a** with **4d** is illustrated in Scheme 3, according to the reported

Scheme 3. Presumed Reaction Mechanism of Electroreductive Coupling of 1a with 4d



mechanism.[6] Carbanion **A** is generated by the two-electron transfer to **4d** and *O*-trimethylsilylation, The nucleophilic addition of **A** proceeds at the 6-position of **1a** through transition states **TS** and subsequent *O*-silylation produces silyl enol ether **B**. During workup, the desilylation of the silyl enol ether moiety in **B** affords **5d**. Therefore, we calculated the transition states **TS** to give ***erythro***- and ***threo*-5d** by the DFT method at the B3LYP/6-311+G(2d,p) level using the IEFPCM model in THF to elucidate the *erythro*-selectivity in the electroreductive coupling of **1a** with **4d**. As exhibited in Figure 1, two transition

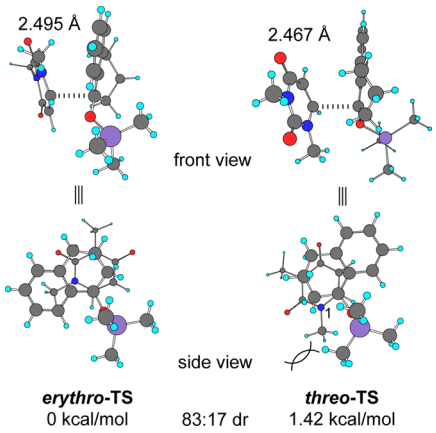


Figure 1. Optimized structures and relative energies of ***erythro*-TS** and ***threo*-TS** calculated at the B3LYP/6-311+G(2d,p) level using the IEFPCM model in THF.

states ***erythro*-TS** and ***threo*-TS** were found and ***erythro*-TS** is lower in energy than ***threo*-TS** (1.42 kcal/mol corresponding to 83:17 dr). The energy difference is probably due to the steric repulsion between the trimethylsiloxy group and 1-methyl group in ***threo*-TS**. Although the calculation results somewhat overestimate the diastereomeric ratio compared to the experimental result (73:27 dr), the *erythro*-selectivity in the formation of **5d** is supported by the DFT calculations.

Dehydrotrimethylsiloxylation of the Adducts. The results of the dehydrotrimethylsiloxylation of **3a−n** by reflux in a benzenoid solvent in the presence of cat. TsOH are summarized in Table 3. From **3a−j** (X = H, Me) except for **3f**, the corresponding 6-diarylmethyl-1,3-dimethyluracils **6a−e,g** (X = H) and **6h−j** (X = Me) were obtained in moderate to high yields (runs 1−5 and 7−10). From **3f**, 6-(9-anthracenyl)-5,6-dihydouracil **6f** was formed as a product (run 6). However, 5,6-fused 1,3-dimethyluracils **7k−n** were given by the reactions of ***cis*-3k−n** (X = F) under the same conditions (runs 11−14).

The results of the dehydrotrimethylsiloxylation of **5a−i** are shown in Table 4. From **5a−c** derived from acetophenones **4a−c**, 6-alkenyl-1,3-dimethyl-5,6-dihydrouracils **8a−c** were obtained as the major products together with 6-alkyl-1,3-dimethyluracils **9a−c** (runs 1−3). In contrast, the reactions of **5d−i** formed from cyclic ketones **4d,e** selectively yielded **8d−i** (runs 4−9). Dehydrogenation of **8d** and ***cis*-8f,h** with DDQ gave 6-(1-naphthyl)-1,3-dimethyl-5,6-dihydrouracils **10d** and ***cis*-10f,h**, respectively (Scheme 4). The stereoconfiguration of ***cis*-8f,h** was completely retained in ***cis*-10f,h**.

Detrimethylsilylation of the Adducts with TBAF. The results of the detrimethylsilylation of **3a−n** with TBAF in THF are summarized in Table 5. The reactions were typically performed until almost all of **3a−n** were consumed. The treatment of **3a−e** (X = H) at 25 °C for 15 min gave 4-substituted 5,5-diaryloxazolidin-2-ones **12a−e** (runs 1−5), while simply detrimethylsilylated alcohols **11f,g** were obtained from **3f,g** (X = H) under the same conditions (runs 6 and 7). The reaction of ***cis*-3h** (X = Me) at 25 °C for 15 min afforded almost *trans*-isomerized **11h** and a diastereomeric mixture of **12h** in 26% (3:97 dr) and 58% (70:30 dr) yields, respectively (run 8). When the reaction was carried out at 0 °C for 15 min, ***cis*-11h** was obtained as the major product (86:14 dr) in 88% yield (run 9). The prolonged reaction time (12 h) at 0 °C brought about considerable isomerization of *cis*-**11h** to *trans*-**11h** (72%, 31:69 dr) and slight formation of **12h** (15%, 80:20 dr) from **11h** (run 10). The desilylation of ***cis*-3i,j** (X = Me) was slower than that of ***cis*-3h** (runs 11−13). The treatment of ***cis*-3i,j** at 25 °C for 12 h afforded completely *trans*-isomerized **11i,j** as minor products and diastereomeric mixtures of **12i,j** as major products (runs 11 and 13). From ***cis*-3k−n** (X = F), **12k−n** were obtained by treatment at 25 °C for 15 min as single stereoisomers (runs 14−17). Since the stereostructure of the obtained **12k−n** was determined to be *threo* by X-ray crystallography, the stereoconfiguration of ***cis*-3k−n** was completely reflected in ***threo*-12k−n**.

The results of the detrimethylsilylation of **5d−i** with TBAF in THF are shown in Table 6. The treatment of both isomers of **5d,e** (X = H) gave the corresponding desilylated alcohols **13d,e** (runs 1−4). While *trans*-isomers of ***erythro*-13f,g** and diastereomeric mixtures of oxazolin-2-ones **14f,g** were formed from ***cis-erythro*-5f,g** (runs 5 and 7), only *trans*-isomerized ***threo*-13f,g** were obtained from ***cis-threo*-5f,g** (runs 6 and 8). In contrast, the reactions of both isomers of ***cis*-5h,i** (X = F) afforded **14h,i** selectively (runs 9−11). Although the stereostructures of **14i** obtained from ***cis-erythro***- and ***cis-threo*-5i** could not be confirmed (runs 10 and 11), they were assumed to be *erythro-threo* and *threo-threo*, respectively, from the completely stereoselective formation of ***threo*-12k−n** (runs 14−17 in Table 5).

The presumed reaction mechanism of the transformation of **3** to **12** is shown in Scheme 5. Detrimethylsilylation of **3** with

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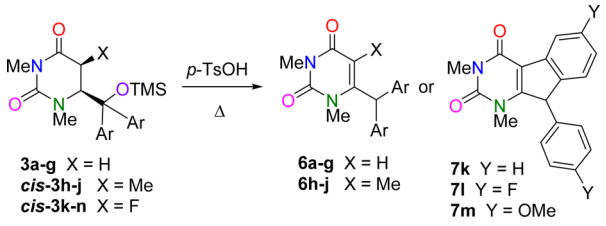
Table 3. Dehydrotrimethylsiloxylation of 3a−n to 6a−j or 7a−d



run	3	solvent	time (h)	6	% yield of 6[a]	7	% yield of 7[a]
1	**3a**	toluene	12	**6a**	86[b]		
2	**3b**	toluene	12	**6b**	82[b]		
3	**3c**	toluene	1	**6c**	93[b]		
4	**3d**	toluene	12	**6d**	94[b]		
5	**3e**	toluene	12	**6e**	66		
6	**3f**	xylene	1	**6f**[c,d]	62		
7	**3g**	xylene	24	**6g**	94		
8	***cis*-3h**	toluene	12	**6h**	83[b]		
9	***cis*-3i**	toluene	12	**6i**	69[b]		
10	***cis*-3j**	toluene	2	**6j**	95[b]		
11	***cis*-3k**	toluene	12			**7k**	70[b]
12	***cis*-3l**	toluene	12			**7l**	51[b]
13	***cis*-3m**	toluene	1			**7m**	72[b]
14	***cis*-3n**	benzene	12			**7n**	90[b]

[a]Isolated yields. [b]Reported data in ref 6. [c]The structure of **6f** is shown below. [d]

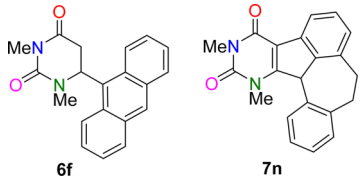
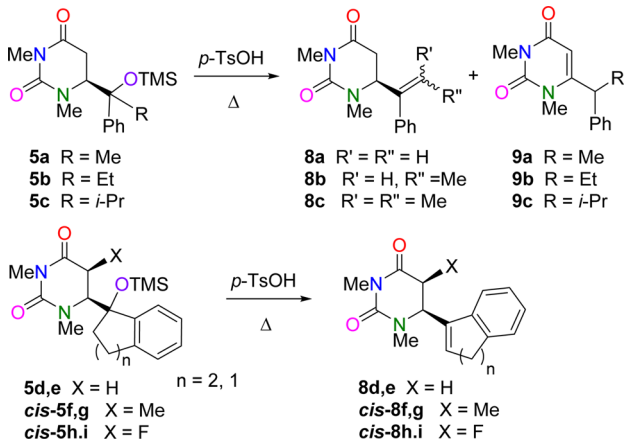


Table 4. Dehydrotrimethylsiloxylation of 5a−i to 8a−i or 9a−c



run	5	solvent	time (h)	8	% yield (gr) of 8[a]	9	% yield of 9[a]
1	**5a**	xylene	12	**8a**	81[b]	**9a**	6[b]
2	**5b**	xylene	12	**8b**	50 (70:30)[b]	**9b**	8[b]
3	**5c**	xylene	72	**8c**	28[b]	**9c**	11[b]
4	**5d**[c]	xylene	2	**8d**	89[b]		
5	**5e**[c]	toluene	2	**8e**	90		
6	***cis*-5f**[c]	toluene	12	***cis*-8f**	85		
7	***cis*-5g**[c]	toluene	3	***cis*-8g**	73		
8	***cis*-5h**[c]	toluene	3	***cis*-8h**	78		
9	***cis*-5i**[c]	toluene	2	***cis*-8i**	76		

[a]Isolated yields. [b]Reported data in ref 6. [c]Diastereomeric mixtures obtained in Table 2.

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Scheme 4. Dehydration of 8d,f,h to 10d,f,h

8d X = H
***cis*-8f** X = Me
***cis*-8h** X = F

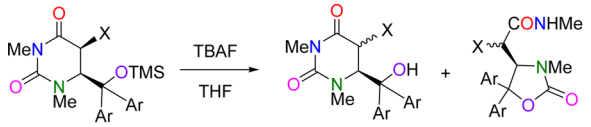
10d 86%
***cis*-10f** 90%
***cis*-10h** 93%

TBAF generates *O*-anion **C**. Intramolecular nucleophilic addition of the *O*-anion to the carbonyl group at the 2-position in **C** forms bicyclo[3.2.1] *O*-anion **D**. Ring-opening of the six-membered ring in **D** and subsequent protonation of the resultant anion **E** produce oxazolizin-2-ones **12**. When the stereoconfiguration of **3h−n** (X = Me, F) is retained, *cis*-isomers of **3h−n** are transformed to ***erythro*-12h−j** (X = Me) and ***threo*-12k−n** (X = F). The relative energies of **E** to **C** (Ar = Ph, X = H, Me, F) calculated by the DFT method at the B3LYP/6-311+G(2d,p) level using the IEFPCM model in THF are summarized in Table 7. These results show that anions **Ea,h,k** are much lower in energy than anions **Ca,h,k** and, therefore, suggest the spontaneous transformation from **C** to **E**. It seems to be possible that an alternative intramolecular nucleophilic addition of the *O*-anion to the carbonyl group at the 4-position in **Ca** and subsequent ring-opening of the resultant **Da′** gives **Ea′**. However, **Ea′** is higher in energy than **Ca** (3.20 kcal/mol). This result shows that the alternative route from **Ca** to **Ea′** is unlikely. Since the transformation of ***cis*-11h−j** (X = Me) to ***erythro*-12h−j** is slow compared with those of ***cis*-11a−e** (X = H) and ***cis*-11k−n** (X = F) probably due to steric and electronic effects of the 5-Me group in ***cis*-11h−j**, the isomerization of ***cis*-11h−j** to ***trans*-11h−j** occurs. Unsurprisingly, ***trans*-Ch** (Ar = Ph, X = Me) is much lower in energy (−5.78 kcal/mol) than ***cis*-Ch**, and this result elucidates the straightforward isomerization of ***cis*-11h−j** and ***cis*-13f,g** to their *trans*-isomers. The transformation of ***trans*-11h−j** with TBAF gave ***threo*-12h−j** as described below.

Detrimethylsilylation of the Adducts with 1 M HCl in MeOH. The results of the detrimethylsilylation of **3a−g** (X = H) and ***cis*-3k−n** (X = F) with 1 M HCl in MeOH at 25 or 0 °C are summarized in Table 8. The reactions were carried out until almost all of **3** was consumed. Except for **3c** and **3f**, the corresponding desilylated alcohols **11a,b,d,e,g,k−n** were obtained in good to high yields (runs 1, 2, 5, 6, 8−12). From **3c** (Ar = 4-MeOC$_6$H$_4$), methyl ether **11c′** (23%) was also formed with **11c** (63%) by the substitution of **11c** with methanol even at 0 °C for 30 min (run 3). Although the desilylation needed prolonged reaction time (6 h), **11c** was formed as the sole product (85%) by treatment with 1 M HCl aq/dioxane (1/1) at 25 °C (run 4). In the reaction of **3f**, dehydrated product **6f** (26%) was also obtained with **11f** (35%) even at 0 °C for 30 min (run 7). Stereoconfiguration of ***cis*-3k−n** was completely retained in ***cis*-11k−n** (runs 9−12).

On the contrary, ***cis*-3h,i** (X = Me) were transformed to ***cis*-11h,i**, *trans*-3,4-disubstituted 5,5-diaryl-γ-butyrolactones ***trans*-15h,i**, and 4-substituted 5,5-diaryloxazolidin-2-imines **16h,i** by treatment with 1 M HCl in MeOH depending on the reaction conditions (Table 9). The treatment of ***cis*-3h** with 1 M HCl in MeOH at 0 °C for 12 h gave ***cis*-11h** (50%) and ***trans*-15h** (25%) (run 1). The reaction at 25 °C accelerated the isomerization of ***cis*-11h** to ***trans*-15h** and brought about the formation of **16h** (runs 2 and 3). Under the same conditions, the product converged with **16h** (90%) after 120 h (run 4). The transformation of ***cis*-3h** to **16h** was completed at reflux temperature within 3 h (run 5). The treatment of ***cis*-3i** at 0 °C for 8 h gave ***cis*-11i** (42%) and ***trans*-15i** (24%) (run 6) and that

Table 5. Detrimethylsilylation of 3a−n to 11f−j, 12a−e, and 12h−m with TBAF



3a-g X = H
***cis*-3h-j** X = Me
***cis*-3k-n** X = F

11f,g X = H
11h-j X = Me

12a-e X = H
12h-j X = Me
12k-n X = F

run	3	temp. (°C)	time	11	% yield of 11[a] (*cis*:*trans*)	12	% yield of 12[a] (dr)
f1	**3a**	25	15 min			**12a**	87
2	**3b**	25	15 min			**12b**	69
3	**3c**	25	15 min			**12c**	83
4	**3d**	25	15 min			**12d**	84
5	**3e**	25	15 min			**12e**	85
6	**3f**	25	15 min	**11f**	81		
7	**3g**	25	15 min	**11g**	88		
8	***cis*-3h**	25	15 min	**11h**	26 (3:97)	**12h**	58 (70:30)[b]
9	***cis*-3h**	0	15 min	**11h**	88 (86:14)		
10	***cis*-3h**	0	12 h	**11h**	72 (31:69)	**12h**	15 (80:20)[b]
11	***cis*-3i**	25	12 h	**11i**	34 (<1:99)	**12i**	54 (78:22)[b]
12	***cis*-3j**	25	15 min	**11j**	90 (29:71)		
13	***cis*-3j**	25	12 h	**11j**	19 (<1:99)	**12j**	67 (45:55)[b]
14	***cis*-3k**	25	15 min			**12k**	63 (>99:1)[c]
15	***cis*-3l**	25	15 min			**12l**	49 (>99:1)[c]
16	***cis*-3m**	25	15 min			**12m**	82 (>99:1)[c]
17	***cis*-3n**	25	15 min			**12n**	82 (>99:1)[c]

[a]Isolated yields. [b]*erythro*:*threo* in parentheses. [c]Obtained as *threo* only.

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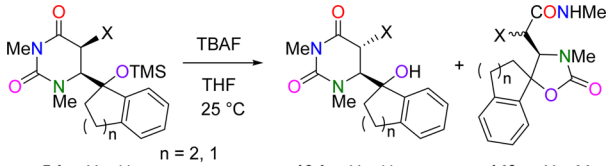
Table 6. Detrimethylsilylation of 5d–i to 13d–g and 14f–i with TBAF



5d,e X = H
***cis*-5f,g** X = Me
***cis*-5h.i** X = F

13d,e X = H
***trans*-13f,g** X = Me

14f,g X = Me
14h,i X = F

run	5 (*cis*-5f–i)	time	13 (*trans*-13f,g)	% yield of 13[a]	14	% yield of 14[a] (dr)
1	*erythro*-5d	15 min	*erythro*-13d	91		
2	*threo*-5d	15 min	*threo*-13d	65		
3	*erythro*-5e	15 min	*erythro*-13e	78		
4	*threo*-5e	15 min	*threo*-13e	79		
5	*erythro*-5f	30 min	*erythro*-13f	41[b]	*erythro*-14f	50 (68:32)
6	*threo*-5f	30 min	*threo*-13f	70[b]		
7	*erythro*-5g	2 h	*erythro*-13g	63[b]	*erythro*-14g	15 (73:27)
8	*threo*-5g	2 h	*threo*-13g	58[b]		
9	5h[c]	15 min			14h	70 (60:40)
10	*erythro*-5i	15 min			*erythro*-14i	83 (>99:1)
11	*threo*-5i	15 min			*threo*-14i	68 (>99:1)

[a]Isolated yields. [b]Obtained as *trans* only. [c]*erythro*:*threo* = 70:30.

Scheme 5. Presumed Reaction Mechanism of the Transformation of 3 to 12

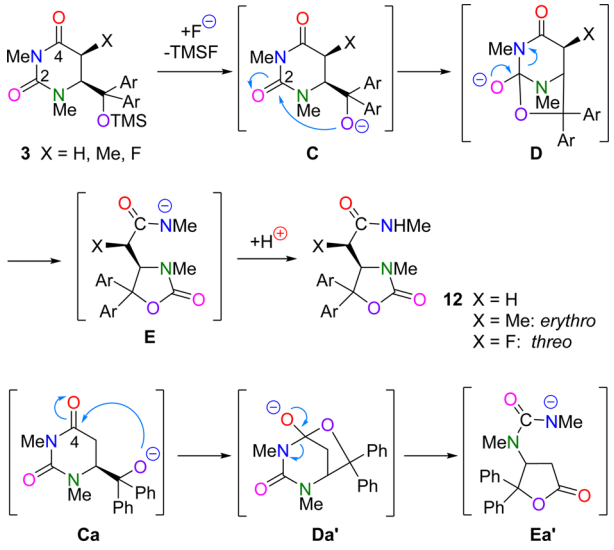


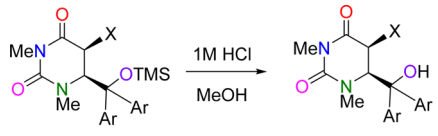
Table 7. Relative Energies of E to C (Ar = Ph, X = H, Me, F) Calculated at the B3LYP/6-311+G(2d,p) Level Using the IEFPCM Model in THF

X	C	E	relative energy of E to C (kcal/mol)
H	Ca	Ea	−5.71
H	Ca	Ea′	3.20
Me	*cis*-Ch[a]	*erythro*-Eh	−6.71
F	*cis*-Ck	*threo*-Ek	−10.09

[a]Relative energy to ***trans*-Ch** is 5.78 kcal/mol.

at 25 °C or reflux temperature did **16i** (89% or 90%) as a sole product (runs 7 and 8). Similarly to the reaction of **3c**, methyl ether ***cis*-11j′** was formed as a major product (52%) with a small amount of ***cis*-11j** (8%) and **16j** (11%) from ***cis*-3j** under the conditions at 0 °C for 6 h (run 9). The alcohol ***cis*-11j** was obtained predominantly (59%) with a small amount of ***trans*-15j**

Table 8. Detrimethylsilylation of 3a–g and 3k–n to 11a–g and 11k–n with 1 M HCl in MeOH

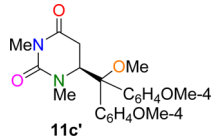


3a-g X = H
***cis*-3k-n** X = F

11a-g X = H
***cis*-11k-n** X = F

run	3	temp. (°C)	time	11	% yield of 11[a]
1	3a	25	15 min	11a	93
2	3b	25	30 min	11b	92
3	3c	0	30 min	11c	63[b,e]
4	3c	25	6 h[c]	11c	85
5	3d	0	30 min	11d	81
6	3e	25	3 h	11e	70
7	3f	0	30 min	11f	35[d]
8	3g	0	30 min	11g	88
9	*cis*-3k	25	2 h	*cis*-11k	91
10	*cis*-3l	25	6 h	*cis*-11l	93
11	*cis*-3m	25	12 h	*cis*-11m	80
12	*cis*-3n	25	6 h	*cis*-11n	84

[a]Isolated yields. [b]Obtained with **11c′** (23%). [c]In 1 M HCl aq/dioxane (1/1). [d]Obtained with **6f** (26%). [e]



11c'

(15%) by treatment with 1 M HCl aq/dioxane (1/1) at 25 °C for 12 h (run 10). The products **11h–j**, **15h–j**, and **16h–j** were all formed as single stereoisomers, and the stereostructures of **11h–j** and **15h–j** were confirmed to be *cis* and *trans*, respectively, by X-ray and ^{1}H NMR analyses. Although the stereoconfiguration of **16h–j** could not be determined, it seemed that the *erythro*-isomers of **16h–j** were obtained exclusively with retaining the stereochemistry. Moreover, it is probable that *Z*-imines of **16h–j** (***erythro*-Z**) were formed

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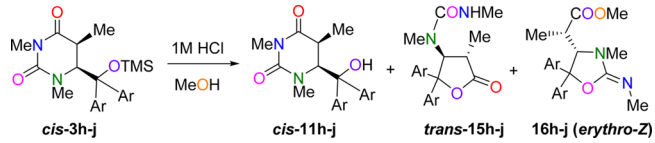
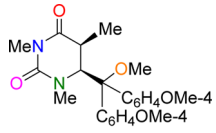
Table 9. Detrimethylsilylation of *cis*-3h–j to *cis*-11h–j, *trans*-15h–j, and 16h–j with 1 M HCl in MeOH



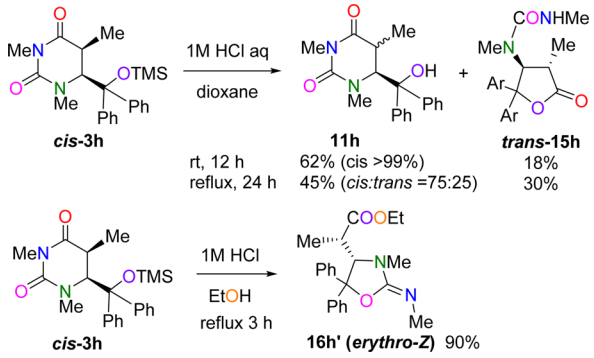
run	*cis*-3	temp. (°C)	time (h)	*cis*-11	% yield of 11[a]	*trans*-15	% yield of 15[a]	16	% yield of 16[a]
1	**3h**	0	1	**11h**	50	**15h**	25		
2	**3h**	25	4	**11h**	25	**15h**	49	**16h**	16
3	**3h**	25	12	**11h**	15	**15h**	24	**16h**	57
4	**3h**	25	120					**16h**	90
5	**3h**	reflux	3					**16h**	90
6	**3i**	0	8	**11i**	42	**15i**	24		
7	**3i**	25	72					**16i**	89
8	**3i**	reflux	3					**16i**	90
9	**3j**	0	6	**11j**	8[b,d]			**16j**	11
10	**3j**	25	12[c]	**11j**	59	**15j**	15		

[a]Isolated yields. [b]Obtained with **11j′** (52%). [c]In 1 M HCl aq/dioxane (1/1). [d]



preferentially, since *Z*-imines are expected to be thermodynamically more stable than *E*-imines. Even after the reaction of ***cis*-3h** was carried out in refluxing 1 M HCl aq/dioxane (1/1) for 24 h, the carboxylic acid corresponding to **16h** could not be obtained; ***cis*-11h** (45%) and ***trans*-15h** (30%) were afforded (Scheme 6). In contrast, the corresponding ethyl ester **16′** (90%) was formed from ***cis*-3h** after reflux in 1 M HCl–EtOH for 3 h.

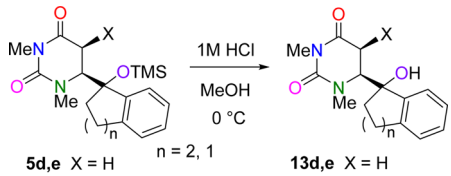
Scheme 6. Treatment of *cis*-3h with Refluxing 1 M HCl aq/Dioxane (1/1) and 1 M HCl–EtOH



On the other hand, the treatment of **5d–j** with 1 M HCl in MeOH at 0 °C for 1–3 h gave the corresponding desilylated alcohols **13d–j** selectively (Table 10). From ***cis*-5f–i** (X = Me, F), *cis*-isomers of **13f–i** were formed exclusively with keeping the stereostructure of ***cis*-5f–i** (runs 5–11).

The presumed reaction mechanism of the transformation of ***cis*-3h** to ***trans*-15h** and **16h** (***erythro*-Z**) is shown in Scheme 7. Initially, acid-catalyzed detrimethylsilylation of ***cis*-3h** in MeOH generates alcohol ***cis*-11h**. After protonation to the carbonyl group at the 4-position in ***cis*-11h**, intramolecular nucleophilic addition of the hydroxy group to the 4-position forms bicyclo[3.2.1] cation **G**. After proton migration to the nitrogen at the 3-position in **G**, ring-opening of the six-membered ring in

Table 10. Detrimethylsilylation of 5d–i to 13d–i with 1 M HCl in MeOH



run	5 (*cis*-5f–i)	time (h)	13 (*cis*-13f–i)	% yield of 13[a]
1	***erythro*-5d**	1	***erythro*-13d**	82
2	***threo*-5d**	1	***threo*-13d**	65
3	***erythro*-5c**	2	***erythro*-13e**	87
4	***threo*-5e**	2	***threo*-13e**	83
5	***erythro*-5f**	1	***erythro*-13f**	80
6	***threo*-5f**	1	***threo*-13f**	74
7	***erythro*-5g**	1	***erythro*-13g**	68
8	***threo*-5g**	2	***threo*-13g**	61
9	**5h**[b]	3	**13h**	77[c]
10	***erythro*-5i**	1	***erythro*-13i**	70
11	***threo*-5i**	1	***threo*-13i**	64

[a]Isolated yields. [b]*erythro:threo* = 70:30. [c]*erythro:threo* = 73:27.

the resultant **H** to **I** and subsequent deprotonation from **I** afford γ-lactone ***trans*-15h**. Under reflux conditions, intramolecular nucleophilic substitution of the urea carbonyl oxygen atom to the 5-position in the protonated ***trans*-15h** (**I**) proceeds through carbocation **J** to give carboxylic acid **L** after deprotonation of the resultant **K**. Finally, acid-catalyzed esterification of **L** produces methyl ester **16h**. The relative energies of ***cis*-11h**, ***trans*-11h**, ***trans*-15h**, **L** (***erythro*-Z** and ***erythro*-E**), and **16h** (***erythro*-Z**) were calculated by the DFT method at the B3LYP/6-311+G(2d,p) level using the IEFPCM model in MeOH and are shown in Table 11. The calculation results show that ***trans*-15h** is thermodynamically more stable (4.49 kcal/mol) than ***cis*-11h**, whereas **L** (***erythro*-Z**) is much more unstable (18.63 kcal/

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Scheme 7. Presumed Reaction Mechanism of the Transformation of *cis*-3h to *trans*-15h and 16h

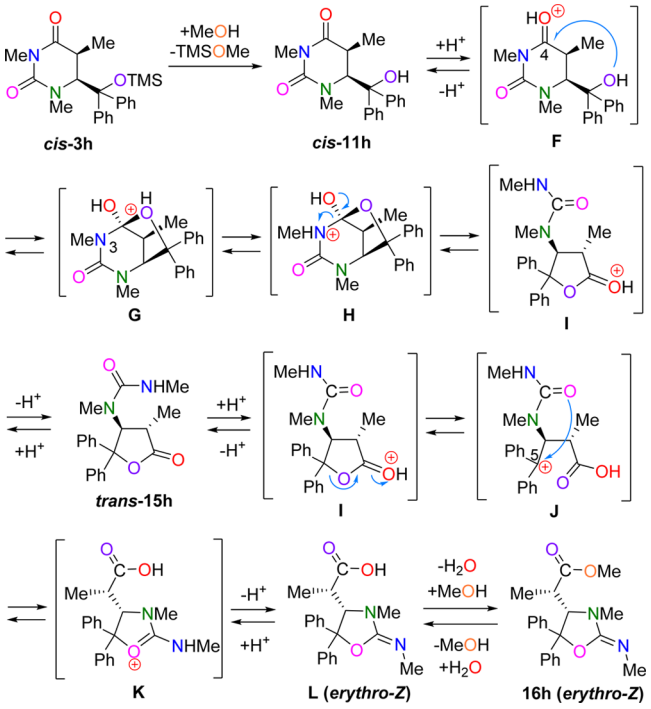


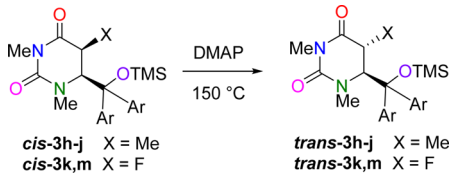
Table 11. Relative Energies of 11h, 15h, L, and 16h Calculated at the B3LYP/6-311+G(2d,p) Level Using the IEFPCM Model in MeOH

	relative energy (kcal/mol)
cis-11h	0
trans-11h	−4.41
trans-15h	−4.49
L (*erythro-Z*)	14.14
L (*erythro-E*)	20.15
6h (*erythro-Z*) − MeOH + H_2O	11.58

mol) than ***trans*-15h**. As expected above, **L** (***erythro-Z***) is more stable (5.97 kcal/mol) than **L** (***erythro-E***). Accordingly, in the reaction of **11h** with 1 M HCl aq/dioxane (1/1) (Scheme 6), **L** was not formed at all. Predictably, ***trans*-11h** is more stable (4.41 kcal/mol) than ***cis*-11h**, and therefore, isomerization of ***cis*-11h** to ***trans*-11h** was observed under the reflux conditions in 1 M HCl aq/dioxane (1/1), as shown in Scheme 6. Under the conditions in 1 M HCl−MeOH, the equilibrium between ***cis*-11h**, ***trans*-15h**, **L**, and **16h** was completely moved to **16h** by esterification of **L** (runs 4 and 5 in Table 9). The driving force of the isomerization of ***cis*-11h−j** to ***trans*-15h−j** seems to be release of steric hindrance, since this type of isomerization could not be observed for **3a−g**, ***cis*-3k−n** (Table 8), and ***trans*-11h−j** (vide infra).

Isomerization of *cis*-5,6-Disubstituted 1,3-Dimethyl-5,6-dihydrouracils to *trans*-Isomers and Their Desilylation. The results of the isomerization of ***cis*-3h−k,m** (X = Me, F) to the corresponding *trans*-isomers ***trans*-3h−k,m** by heating at 150 °C in the presence of cat. DMAP are summarized in Table 12. The isomerization of ***cis*-3h−j** (X = Me) was completed after heating for 24 h (runs 1−3), while that of ***cis*-3k,m** (X = F) was finished within 8 h (runs 4 and 5). Similarly, the isomerization of ***cis*-8f,h** and ***cis*-10f** was effected under the

Table 12. Isomerization from 3h−k,m to *trans*-3h−k,m

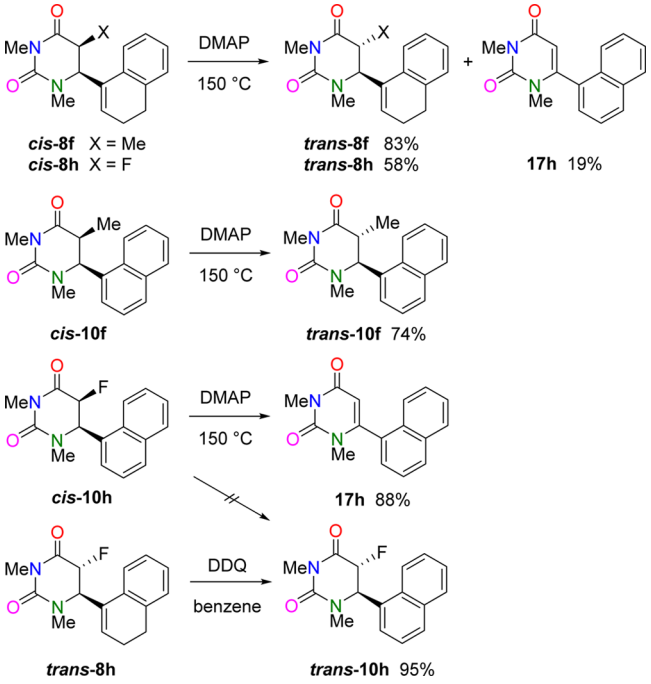


run	*cis*-3	time (h)	*trans*-3	% yield of *trans*-3[a]
1	***cis*-3h**	24	***trans*-3h**	67
2	***cis*-3i**	24	***trans*-3i**	63
3	***cis*-3j**	24	***trans*-3j**	75
4	***cis*-3k**	8	***trans*-3k**	70
5	***cis*-3m**	8	***trans*-3m**	82

[a]Isolated yields.

same conditions (Scheme 8). However, a dehydrofluorinated product **17h** was the only product in the reaction of ***cis*-10h**

Scheme 8. Isomerization of 8f,h and 10f,h



under the same conditions. Incidentally, ***trans*-10h** was obtained by dehydrogenation of ***trans*-8h** with DDQ.

The results of detrimethylsilylation of ***trans*-3h−k,m** with TBAF are shown in Table 13. Whereas mixtures of ***trans*-11h−j** and ***threo*-12h−j** (X = Me) were obtained from ***trans*-3h−j** (runs 1−3), ***erythro*-12k,m** (X = F) were only products from ***trans*-3k,m** (runs 4 and 5). The stereoconfiguration of ***trans*-3h−k,m** was completely reflected in ***threo*-12h−j** and ***erythro*-12k,m**. On the other hand, detrimethylsilylation of ***trans*-3h−k,m** with 1 M HCl aq/dioxane (1/1) at 25 °C selectively gave ***trans*-11h−k,m** in high yields (Table 14). The isomerization of ***trans*-11h−j** to γ-lactone **15** as described above could not be observed at all.

Assignment of Geometric Isomers of 5,6-Disubstituted 1,3-Dimethyl-5,6-dihydrouracils. Table 15 exhibits ^{1}H NMR chemical shifts of 6-H and coupling constants between 5-H and 6-H ($J_{5,6}$) of 5,6-*cis*- and *trans*-substituted 5,6-dihydro-1,3-dimethyluracils obtained in this paper. These results indicate

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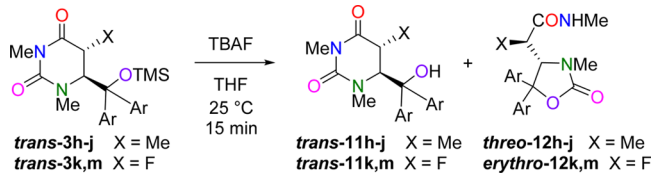
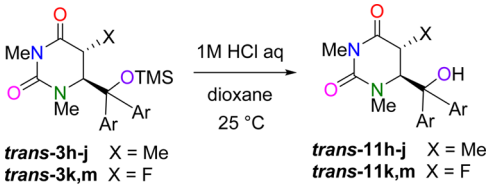
Table 13. Detrimethylsilylation of *trans*-3h−l,k,m with TBAF



run	*trans*-3	*trans*-11	% yield of *trans*-11[a]	12	% yield of 12[a]
1	*trans*-3h	*trans*-11h	56	*threo*-12h	34
2	*trans*-3i	*trans*-11i	63	*threo*-12i	25
3	*trans*-3j	*trans*-11j	52	*threo*-12j	28
4	*trans*-3k			*erythro*-12k	87
5	*trans*-3m			*erythro*-12m	92

[a]Isolated yields.

Table 14. Detrimethylsilylation of *trans*-3h−k,m with 1 M HCl aq/Dioxane (1/1)



run	*trans*-3	time (h)	*trans*-11	% yield of *trans*-11[a]
1	*trans*-3h	12	*trans*-11h	96
2	*trans*-3i	12	*trans*-11i	94
3	*trans*-3j	12	*trans*-11j	84
4	*trans*-3k	8	*trans*-11k	95
5	*trans*-3m	8	*trans*-11m	92

[a]Isolated yields.

that the $J_{5,6}$ values of 5,6-disubstituted uracils are within 5.3−8.0 Hz for *cis* and 0−2.6 Hz for *trans*. Consequently, the geometric structure of 5,6-disubstituted 5,6-dihydrouracils can readily be assigned by the $J_{5,6}$ values of their ^{1}H NMR spectra.

CONCLUSION

The electroreductive intermolecular coupling of 1,3-dimethyluracil (**1a**), thymine (**1b**), and 5-fluorouracil (**1c**) with benzophenones **2a−g** and alkyl aryl ketones **4a−e** in the presence of TMSCl in THF proceeded at the 6-position of **1a−c** to give adducts **3a−n** and **5a−i**, respectively. The adducts **3h−n** and **5f−i** obtained from **1b** and **1c** were formed as *cis*-isomers stereoselectively. Furthermore, the adducts **5d−i** derived from cyclic alkyl aryl ketones **4d** and **4e** were obtained *erythro*-selectively. Treatment of **3a−j** obtained from **1a** and **1b** with refluxing cat. *p*-TsOH/toluene or xylene gave 6-diarylmethyl-1,3-dimethyluracils **6a−j**. In contrast, the same treatment of **3k−n** obtained from **1c** afforded 5,6-fused 1,3-dimethyluracils **7k−n**. The adducts **5a−i** were transformed to 6-alkenyl-5,6-dihydro-1,3-dimethyluracils **8a−i** by reflux in *p*-TsOH/xylene or toluene. Treatment of **3a−f**, ***cis*-3k−n**, and ***cis*-5h,i** obtained from **1a** and **1c** with TBAF in THF gave 4-substituted 5,5-diaryloxazolidin-2-ones **12a−e**, ***threo*-12k−n**, and ***threo*-14h,i**, respectively. On the other hand, the same treatment of ***cis*-3h−j** obtained from **1b** afforded *trans*-isomerized alcohols ***trans*-11h−j** and diastereomeric mixtures of **12h−j**. The same treatment of ***cis*-5f,g** obtained from **1b** also produced *trans*-isomerized alcohols ***trans*-13f,g**. Treatment of the adducts **3** and **5** except for ***cis*-3h−j** with 1 M HCl−MeOH gave the corresponding desilylated alcohols **11** and **13** with completely retaining their stereochemistry. The same treatment of ***cis*-3h−j** afforded 3,4-

Table 15. ^{1}H NMR Chemical Shifts of 6-H and Coupling Constants ($J_{5,6}$) of 5,6-*cis*- and *trans*-Substituted 1,3-Dimethyl-5,6-dihydrouracils

	cis		*trans*	
	6-H (δ)	$J_{5,6}$ (Hz)	6-H (δ)	$J_{5,6}$ (Hz)
3h	4.45	6.3	4.09	0[a]
11h	4.24	5.8	3.93	0[a]
3i	4.41	6.3	4.03	0
11i	4.16	5.4[a]	3.86	0
3j	4.38	6.9	4.02	0
11j	4.13	5.9	3.83	0[a]
11j′	4.41	6.7[a]		
3k	4.96	8.0	4.61	0
11k	4.71	6.9	4.44	0
3l	4.90	7.7[a]		
11l	4.67	6.9		
3m	4.88	8.0	4.53	0
11m	4.63	6.9	4.34	0
3n	4.71	7.7[a]		
11n	4.61	6.7[a]		
***erythro*-5f**	3.77	6.3		
***threo*-5f**	3.52	5.3		
8f	4.72	7.0	4.21	0
10f	5.45	7.3	4.95	2.3
***erythro*-13f**	3.90	6.3[a]	3.40	0
***threo*-13f**	3.90	6.0	3.41	0
***erythro*-5g**	3.68	6.2		
***threo*-5g**	3.22	5.9		
8g	4.56	6.9		
***erythro*-13g**	3.75	6.1	3.36	1.1
***threo*-13g**	3.37	5.6[a]	3.07	0[a]
***erythro*-5h**	3.97	6.7		
***threo*-5h**	3.84	6.2		
8h	5.03	6.9[a]	4.83	2.5
10h	5.75	7.5	5.54	2.6
***erythro*-13h**	4.22	6.9[a]		
***threo*-13h**	3.93	6.6		
***erythro*-5i**	3.99	6.9		
***threo*-5i**	3.54	6.7		
8i	4.93	6.9[a]		
***erythro*-13i**	4.13	7.0		
***threo*-13i**	3.66	6.9		

[a]Confirmed by X-ray crystallography.

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disubstituted-5,5-diaryl-γ-butyrolactones ***trans*-15h−j** and 4-substituted 5,5-diaryloxazolidin-2-imines **16h−j** (***erythro*-Z**) depending on the reaction conditions. These types of transformations were observed only in the reaction of highly sterically hindered ***cis*-3h−j**. Isomerization of ***cis*-3h−k,m** and ***cis*-8f,h** to the corresponding *trans*-isomers was effected by heating in the presence of cat. DMAP. The geometric structure of 5,6-disubstituted 5,6-dihydro-1,3-dimethyluracils was assigned by the $J_{5,6}$ values of their ^{1}H NMR spectra.

EXPERIMENTAL SECTION

General Methods. Column chromatography was performed on silica gel 60. THF was freshly distilled from sodium benzophenone ketyl radical. DMF, TMSCl, and TEA were distilled from CaH_2.

Typical Procedure for Electroreductive Coupling. A 0.3 M solution of Bu_4NClO_4 in THF (15 mL) was placed in the cathodic chamber of a divided cell (40 mL beaker, 3 cm diameter, 6 cm height) equipped with a platinum cathode (5 × 5 cm^2), a platinum anode (2 × 1 cm^2), and a ceramic cylindrical diaphragm (1.5 cm diameter). A 0.3 M solution of Et_4NOTs in DMF (4 mL) was placed in the anodic chamber (inside the diaphragm). 1,3-Dimethylpyrimidine-2,4(1*H*,3*H*)-dione (**1a**) (140 mg, 1.0 mmol), benzophenone (**2a**) (368 mg, 2.0 mmol), TMSCl (0.64 mL, 5 mmol), and TEA (0.70 mL, 5 mmol) were added to the cathodic chamber. After 400 C of electricity was passed at a constant current of 200 mA at 25 °C under a nitrogen atmosphere, the catholyte was evaporated *in vacuo*. The residue was dissolved in diethyl ether (20 mL), and the insoluble solid was filtered off. After removal of the solvent *in vacuo*, the residue was purified by column chromatography on silica gel (hexanes−EtOAc) to give **3a** (305 mg) in 77% yield. Compounds **3a−d**, ***cis*-3h−n**, and **5a−d** were already reported.[6]

*1,3-Dimethyl-6-(5-((trimethylsilyl)oxy)-5H-dibenzo[a,d][7]annulen-5-yl)dihydropyrimidine-2,4(1H,3H)-dione (**3e**).* Colorless paste (244 mg, 58%) R_f 0.5 (hexanes−ethyl acetate, 1:1); IR (ATR) 1707, 1655, 1512, 1483, 993, 980, 943, 912, 880, 835, 806, 797, 764, 756, 727, 683, 662 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 0.36 (s, 9H), 2.08 (d, 1H, *J* = 17.0 Hz), 2.32 (dd, 1H, *J* = 7.9, 17.0 Hz), 3.16 (s, 3H), 4.13 (d, 1H, *J* = 7.9 Hz), 6.89 (s, 2H), 7.32−7.38 (m, 4H), 7.41−7.49 (m, 2H), 7.78−7.83 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 3.3 (q), 27.1 (q), 31.6 (t), 37.2 (q), 56.3 (d), 88.6 (s), 127.4 (d), 127.6 (d), 127.8 (d), 127.9 (d), 128.3 (d), 128.7 (d), 130.6 (d), 130.7 (d), 131.5 (d), 132.3 (d), 133.3 (s), 138.7 (s), 140.6 (s), 154.2 (s), 169.3 (s); HRMS (ESI, ion trap) calcd for $C_{24}H_{29}N_2O_3Si$ (M + H$^+$) 421.1947; found 421.1945.

*1,3-Dimethyl-6-(9-((trimethylsilyl)oxy)-9,10-dihydroanthracen-9-yl)dihydropyrimidine-2,4(1H,3H)-dione (**3f**).* White solid (180 mg, 44%); R_f 0.3 (hexanes−ethyl acetate, 2:1); mp 186−188 °C; IR (ATR) 1709, 1663, 1558, 1541, 1506, 1481, 951, 945, 920, 899, 878, 868, 843, 775, 768, 754, 721, 689, 673, 669 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ −0.21 (s, 9H), 2.33 (d, 1H, *J* = 17.2 Hz), 2.49 (dd, 1H, *J* = 8.0, 17.2 Hz), 2.52 (s, 3H), 3.12 (s, 3H), 3.69 (d, 1H, *J* = 8.0 Hz), 4.04 (d, 1H, *J* = 20.5 Hz), 4.18 (d, 1H, *J* = 20.5 Hz), 7.27−7.37 (m, 6H), 7.57−7.60 (m, 1H), 7.65−7.68 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 1.3 (q), 26.1 (q), 31.4 (t), 33.4 (t), 38.4 (q), 65.9 (d), 78.8 (s), 125.7 (d), 126.1 (d), 126.5 (d), 127.32 (d), 127.34 (d), 127.6 (d), 127.8 (d), 133.0 (s), 133.3 (s), 136.0 (s), 137.7 (s), 152.9 (s), 167.8 (s). Anal. Calcd for $C_{23}H_{28}N_2O_3Si$: C, 67.61; H, 6.91; N, 6.86. Found: C, 67.57; H, 6.90; N, 6.75.

*1,3-Dimethyl-6-(9-((trimethylsilyl)oxy)-9H-xanthen-9-yl)dihydropyrimidine-2,4(1H,3H)-dione (**3g**).* White solid (213 mg, 52%); R_f 0.3 (hexanes−ethyl acetate, 2:1); mp 176−178 °C; IR (ATR) 1711, 1663, 1601, 1574, 1506, 1474, 961, 928, 903, 880, 870, 843, 758, 750, 689, 673 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ −0.16 (s, 9H), 2.31 (d, 1H, *J* = 17.1 Hz), 2.53 (dd, 1H, *J* = 8.4, 17.1 Hz), 2.59 (s, 3H), 3.19 (s, 3H), 3.68 (d, 1H, *J* = 8.4 Hz), 7.13−7.22 (m, 4H), 7.31−7.39 (m, 2H), 7.46−7.49 (m, 1H), 7.53−7.56 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 1.5 (q), 26.5 (q), 31.4 (t), 38.8 (q), 65.9 (d), 74.3 (s), 116.5 (d), 116.7 (d), 121.6 (s), 122.7 (d), 123.2 (d), 123.8 (s), 126.7 (d), 127.8 (d), 129.7 (d), 129.9 (d), 149.8 (s), 149.9 (s), 153.3 (s), 167.4 (s). Anal. Calcd for $C_{22}H_{26}N_2O_4Si$: C, 64.36; H, 6.38; N, 6.82. Found: C, 64.41; H, 6.40; N, 6.73.

(R)-1,3-Dimethyl-6-((S*)-1-((trimethylsilyl)oxy)-2,3-dihydro-1H-inden-1-yl)dihydropyrimidine-2,4(1H,3H)-dione (**erythro-5e**).* Colorless paste (99 mg, 29%); R_f 0.5 (hexanes−ethyl acetate, 1:1); IR (ATR) 1709, 1659, 1477, 993, 980, 947, 926, 910, 881, 868, 837, 754, 725, 687, 675 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ −0.06 (s, 9H), 2.19−2.26 (m, 1H), 2.33−2.38 (m, 1H), 2.70 (dd, 1H, *J* = 8.0, 16.7 Hz), 2.81 (dd, 1H, *J* = 1.0, 16.7 Hz), 2.84 (s, 3H), 2.87−2.98 (m, 2H), 3.14 (s, 3H), 3.53 (dd, 1H, *J* = 1.0, 8.0 Hz), 7.18−7.27 (m, 4H); ^{13}C NMR ($CDCl_3$) δ 1.7 (q), 26.7 (q), 29.4 (t), 31.7 (t), 37.8 (t), 38.8 (q), 62.3 (d), 88.8 (s), 124.7 (d), 125.2 (d), 126.4 (d), 128.9 (d), 142.0 (s), 143.5 (s), 153.8 (s), 168.4 (s); HRMS (ESI, ion trap) calcd for $C_{18}H_{27}N_2O_3Si$ (M + H$^+$) 347.1791; found 347.1789.

(R)-1,3-Dimethyl-6-((R*)-1-((trimethylsilyl)oxy)-2,3-dihydro-1H-inden-1-yl)dihydropyrimidine-2,4(1H,3H)-dione (**threo-5e**).* Colorless paste (81 mg, 23%); R_f 0.35 (hexanes−ethyl acetate, 1:1); IR (ATR) 1709, 1651, 1516, 1474, 980, 945, 883, 870, 835, 768, 756, 727, 698, 686, 671 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ −0.08 (s, 9H), 2.14−2.28 (m, 2H), 2.43 (s, 3H), 2.80−2.87 (m, 3H), 2.96 (dd, 1H, *J* = 9.0, 15.9 Hz), 3.25 (dd, 1H, *J* = 2.1, 6.2 Hz), 7.19−7.29 (m, 4H); ^{13}C NMR ($CDCl_3$) δ 1.4 (q), 27.0 (q), 28.9 (t), 33.1 (t), 38.0 (t), 38.1 (q), 60.7 (d), 88.7 (s), 124.7 (d), 125.1 (d), 126.8 (d), 128.7 (d), 141.1 (s), 144.0 (s), 154.1 (s), 169.5 (s); HRMS (ESI, ion trap) calcd for $C_{18}H_{27}N_2O_3Si$ (M + H$^+$) 347.1791; found 347.1789.

(5R,6R*)-1,3,5-Trimethyl-6-((S*)-1-((trimethylsilyl)oxy)-1,2,3,4-tetrahydronaphthalen-1-yl)dihydropyrimidine-2,4(1H,3H)-dione (**cis-erythro-5f**).* Colorless paste (242 mg, 65%); R_f 0.45 (hexanes−ethyl acetate, 2:1); IR (ATR) 1709, 1653, 1520, 1485, 943, 914, 903, 885, 858, 837, 770, 758, 741, 687, 662 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ −0.13 (s, 9H), 1.47 (d, 3H, *J* = 7.3 Hz), 1.49−1.60 (m, 1H), 1.71−1.90 (m, 2H), 2.04−2.11 (m, 1H), 2.28 (s, 3H), 2.66−2.72 (m, 2H), 3.05−3.13 (m, 1H), 3.17 (s, 3H), 3.77 (d, 1H, *J* = 6.3 Hz), 7.03−7.06 (m, 1H), 7.16−7.23 (m, 2H), 7.54−7.57 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 1.6 (q), 13.6 (q), 20.0 (t), 27.3 (q), 29.3 (t), 33.3 (t), 37.8 (q), 39.5 (d), 68.1 (d), 77.3 (s), 125.6 (d), 127.6 (d), 128.2 (d), 128.8 (d), 137.3 (s), 140.0 (s), 153.8 (s), 172.6 (s); HRMS (ESI, ion trap) calcd for $C_{20}H_{31}N_2O_3Si$ (M + H$^+$) 375.2104; found 375.2101.

(5R,6R*)-1,3,5-Trimethyl-6-((R*)-1-((trimethylsilyl)oxy)-1,2,3,4-tetrahydronaphthalen-1-yl)dihydropyrimidine-2,4(1H,3H)-dione (**cis-threo-5f**).* Colorless paste (43 mg, 11%); R_f 0.4 (hexanes−ethyl acetate, 1:1); IR (ATR) 1709, 1663, 1483, 955, 918, 910, 876, 837, 770, 750, 725, 687, 667 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ −0.10 (s, 9H), 1.46 (d, 3H, *J* = 7.0 Hz), 1.81−1.91 (m, 2H), 2.01−2.06 (m, 1H), 2.06 (s, 3H), 2.25−2.36 (m, 1H), 2.80−2.85 (m, 2H), 2.95−3.02 (m, 1H), 3.25 (s, 3H), 3.52 (d, 1H, *J* = 5.3 Hz), 7.02−7.05 (m, 1H), 7.11−7.23 (m, 3H); ^{13}C NMR ($CDCl_3$) δ 1.6 (q), 13.7 (q), 20.3 (t), 27.3 (q), 28.4 (t), 35.8 (t), 36.3 (q), 39.9 (d), 66.2 (d), 79.1 (s), 126.0 (d), 127.5 (d), 128.0 (d), 128.4 (d), 135.9 (s), 139.3 (s), 154.1 (s), 172.5 (s); HRMS (ESI, ion trap) calcd for $C_{20}H_{31}N_2O_3Si$ (M + H$^+$) 375.2104; found 375.2102.

(5R,6R*)-1,3,5-Trimethyl-6-((S*)-1-((trimethylsilyl)oxy)-2,3-dihydro-1H-inden-1-yl)dihydropyrimidine-2,4(1H,3H)-dione (**cis-erythro-5g**).* Colorless paste (128 mg, 36%); R_f 0.45 (hexanes−ethyl acetate, 2:1); IR (ATR) 1709, 1665, 1474, 928, 901, 876, 837, 806, 781, 756, 735, 704, 687, 669 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ −0.10 (s, 9H), 1.48 (d, 3H, *J* = 7.3 Hz), 2.04−2.13 (m, 1H), 2.27−2.33 (m, 1H), 2.37 (s, 3H), 2.56−2.64 (m, 1H), 2.83−2.91 (m, 1H), 3.04−3.11 (m, 1H), 3.13 (s, 3H), 3.68 (d, 1H, *J* = 6.2 Hz), 7.18−7.28 (m, 3H), 7.31−7.33 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 1.7 (q), 13.2 (q), 27.3 (q), 29.7 (t), 36.3 (t), 39.1 (q), 39.7 (d), 67.0 (d), 88.1 (s), 124.5 (d), 125.1 (d), 126.1 (d), 128.7 (d), 142.0 (s), 145.5 (s), 153.4 (s), 172.6 (s); HRMS (ESI, ion trap) calcd for $C_{19}H_{29}N_2O_3Si$ (M + H$^+$) 361.1947; found 361.1945.

(5R,6R*)-1,3,5-Trimethyl-6-((R*)-1-((trimethylsilyl)oxy)-2,3-dihydro-1H-inden-1-yl)dihydropyrimidine-2,4(1H,3H)-dione (**cis-threo-5g**).* Colorless paste (63 mg, 17%); R_f 0.2 (hexanes−ethyl acetate, 2:1); IR (ATR) 1709, 1659, 1477, 964, 916, 899, 837, 772, 752, 725, 681, 669 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ −0.11 (s, 9H), 1.49 (d, 3H, *J* = 6.9 Hz), 2.09−2.17 (m, 1H), 2.11 (s, 3H), 2.47−2.53 (m, 1H), 2.79−2.88 (m, 1H), 2.93−3.01 (m, 1H), 3.22 (d, 1H, *J* = 5.9 Hz), 3.22 (s, 3H), 7.06−7.10 (m, 1H), 7.18−7.21 (m, 1H), 7.23−7.27 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 1.5 (q), 13.3 (q), 27.3 (q), 29.2 (t), 36.9 (q), 39.2 (d), 39.9 (t), 65.2 (d), 88.4 (s), 124.5 (d), 125.5 (d), 127.1 (d), 128.4 (d), 141.1

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(s), 145.1 (s), 154.0 (s), 172.4 (s); HRMS (ESI, ion trap) calcd for $C_{19}H_{29}N_2O_3Si$ (M + H^+) 361.1947; found 361.1944.

(5R,6S*)-5-Fluoro-1,3-dimethyl-6-(1-((trimethylsilyl)oxy)-1,2,3,4-tetrahydronaphthalen-1-yl)dihydropyrimidine-2,4(1H,3H)-dione (**cis-5h**).* Colorless paste (227 mg, 60%, 70:30 dr); R_f 0.35 (hexanes−ethyl acetate, 2:1); IR (ATR) 1719, 1670, 953, 899, 839, 795, 748, 723, 691, 665 cm^{-1}; ^1H NMR ($CDCl_3$) δ −0.042 (s, 3H), −0.037 (s, 6H), 1.83−1.95 (m, 2.67H), 2.03−2.10 (m, 1.33H), 2.28−2.34 (m, 0.67H), 2.58−2.63 (m, 0.33H), 2.74−2.90 (m, 2H), 3.02 (s, 2H), 3.20 (s, 2H), 3.25 (s, 1H), 3.84 (d, 0.33H, J = 6.2 Hz), 3.97 (d, 0.67H, J = 6.7 Hz), 5.20 (dd, 0.67H, J = 6.7, J_{HF} = 47.0 Hz), 5.27 (dd, 0.33H, J = 6.2, J_{HF} = 47.1 Hz), 7.04−7.09 (m, 1H), 7.14−7.27 (m, 2.33H), 7.47−7.51(m, 0.67H); ^{13}C NMR ($CDCl_3$) δ major: 1.7 (q), 19.4 (t), 27.1 (q), 28.3 (t), 34.9 (t), 39.3 (q), 64.9 (d, J_{CCF} = 18.0 Hz), 78.2 (s, J_{CCCF} = 2.4 Hz), 83.3 (d, J_{CF} = 200.3 Hz), 124.9 (d), 127.6 (d), 128.1 (d), 128.6 (d), 136.5 (s), 138.1 (s), 153.1 (s), 166.9 (s, J_{CCF} = 21.6 Hz), minor: 1.4 (q), 19.9 (t), 27.1 (q), 27.9 (t), 34.3 (t), 36.7 (q), 63.4 (d, J_{CCF} = 19.2 Hz), 77.9 (s, J_{CCCF} = 2.4 Hz), 83.9 (d, J_{CF} = 196.1 Hz), 126.0 (d), 127.4 (d), 127.6 (d), 128.5 (d), 135.8 (s), 138.4 (s), 152.8 (s), 166.9 (s, J_{CCF} = 21.0 Hz); HRMS (ESI, ion trap) calcd for $C_{19}H_{28}FN_2O_3Si$ (M + H^+) 379.1853; found 379.1851.

(5R,6S*)-5-Fluoro-1,3-dimethyl-6-((S*)-1-((trimethylsilyl)oxy)-2,3-dihydro-1H-inden-1-yl)dihydropyrimidine-2,4(1H,3H)-dione (**cis-erythro-5i**).* Colorless paste (84 mg, 23%); R_f 0.65 (hexanes−ethyl acetate, 1:1); IR (ATR) 1734, 1670, 1474, 991, 951, 893, 839, 752, 725, 683 cm^{-1}; ^1H NMR ($CDCl_3$) δ −0.05 (s, 9H), 2.15−2.23 (m, 1H), 2.54−2.61 (m, 1H), 2.71 (s, 3H), 2.84−2.93 (m, 1H), 2.98−3.07 (m, 1H), 3.32 (s, 3H), 3.99 (d, 1H, J = 6.9 Hz), 5.21 (dd, 1H, J = 6.9, J_{HF} = 47.0 Hz), 7.15−7.25 (m, 4H); ^{13}C NMR ($CDCl_3$) δ 1.8 (q), 26.9 (q), 29.1 (t), 39.1 (t), 39.4 (q), 65.4 (d, J_{CCF} = 21.6 Hz), 83.5 (d, J_{CF} = 197.0 Hz), 88.8 (s), 125.1 (d), 125.3 (d), 126.1 (d), 129.0 (d), 142.7 (s), 142.9 (s), 152.7 (s), 165.1 (s, J_{CCF} = 21.6 Hz); HRMS (ESI, ion trap) calcd for $C_{18}H_{25}FN_2O_3Si$ (M + H^+) 365.1697; found 365.1695.

(5R,6S*)-5-Fluoro-1,3-dimethyl-6-((R*)-1-((trimethylsilyl)oxy)-2,3-dihydro-1H-inden-1-yl)dihydropyrimidine-2,4(1H,3H)-dione (**cis-threo-5i**).* Colorless paste (69 mg, 19%); R_f 0.55 (hexanes−ethyl acetate, 1:1); IR (ATR) 1728, 1717, 1676, 1665, 1474, 939, 908, 839, 804, 772, 752, 725, 694, 679 cm^{-1}; ^1H NMR ($CDCl_3$) δ −0.06 (s, 9H), 2.16−2.22 (m, 1H), 2.20 (s, 3H), 2.66−2.71 (m, 1H), 2.80−2.87 (m, 1H), 2.92−3.01 (m, 1H), 3.24 (s, 3H), 3.54 (d, 1H, J = 6.7 Hz), 5.28 (dd, 1H, J = 6.7, J_{HF} = 47.0 Hz), 7.15−7.30 (m, 4H); ^{13}C NMR ($CDCl_3$) δ 1.8 (q), 27.3 (q), 29.0 (t), 37.7 (q), 38.6 (t), 62.7 (d, J_{CCF} = 20.4 Hz), 83.2 (d, J_{CF} = 195.5 Hz), 87.1 (s), 124.7 (d), 125.2 (d), 127.1 (d), 128.7 (d), 141.3 (s), 144.3 (s), 153.0 (s), 167.0 (s, J_{CCF} = 21.6 Hz); HRMS (ESI, ion trap) calcd for $C_{18}H_{25}FN_2O_3Si$ (M + H^+) 365.1697; found 365.1696.

Typical Procedure for Elimination of Trimethylsilanol from the Adducts. A solution of **3a** (198 mg, 0.5 mmol) and *p*-TsOH (10 mg) in toluene (10 mL) was refluxed under a nitrogen atmosphere for 12 h. After the solvent was removed *in vacuo*, the residue was purified by column chromatography on silica gel (hexanes−EtOAc) to give **6a** (132 mg) in 86% yield. Compounds **6a−e**, **6h−j**, **7k−n**, **8a−d**, and **9a−c** were already reported.[6]

*6-(Anthracen-9-yl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (**6f**).* Colorless paste (169 mg, 62%); R_f 0.4 (hexanes−ethyl acetate, 2:1); IR (ATR) 1707, 1655, 1526, 993, 891, 862, 843, 793, 758, 731, 702, 683, 662 cm^{-1}; ^1H NMR ($CDCl_3$) δ 2.77 (s, 3H), 2.93 (dd, 1H, J = 5.3, 17.6 Hz), 3.41 (s, 3H), 3.69 (dd, 1H, J = 13.8, 17.6 Hz), 6.23 (dd, 1H, J = 5.3, 13.8 Hz), 7.48−7.61 (m, 4H), 8.05−8.10 (m, 2H), 8.23−8.27 (m, 1H), 8.51−8.56 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 28.2 (q), 33.0 (q), 37.2 (t), 51.6 (d), 121.7 (d), 123.7 (d), 125.0 (d), 125.1 (d), 126.7 (s), 126.9 (d), 127.3 (d), 129.1 (s), 129.7 (d), 129.8 (d), 130.1 (d), 130.2 (s), 131.4 (s), 131.7 (s), 154.8 (s), 168.5 (s); HRMS (ESI, ion trap) calcd for $C_{20}H_{19}N_2O_2$ (M + H^+) 319.1447; found 319.1446.

*1,3-Dimethyl-6-(9H-xanthen-9-yl)pyrimidine-2,4(1H,3H)-dione (**6g**).* Colorless paste (150 mg, 94%); R_f 0.6 (hexanes−ethyl acetate, 1:1); IR (ATR) 1703, 1653, 1616, 1570, 1479, 989, 905, 860, 849, 824, 772, 762, 746, 700, 685, 667 cm^{-1}; ^1H NMR ($CDCl_3$) δ 2.91 (s, 3H), 3.38 (s, 3H), 5.41 (s, 1H), 5.96 (s, 1H), 7.06−7.10 (m, 2H), 7.11−7.15 (m, 4H), 7.30−7.35 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 28.0 (q), 32.0 (q), 43.4 (d), 104.9 (d), 117.1 (d), 117.5 (s), 123.8 (d), 127.7 (d), 129.6 (d), 150.0 (s), 152.7 (s), 152.9 (s), 162.0 (s); HRMS (ESI, ion trap) calcd for $C_{19}H_{17}N_2O_3$ (M + H^+) 321.1239; found 321.1237.

*6-(1H-Inden-3-yl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (**8e**).* Pale yellow solid (115 mg, 90%); R_f 0.4 (hexanes−ethyl acetate, 1:1); mp 116−118 °C; IR (ATR) 1763, 1746, 1705, 1647, 999, 968, 951, 914, 804, 772, 754, 721, 700, 682, 669 cm^{-1}; ^1H NMR ($CDCl_3$) δ 3.05 (dd, 1H, J = 3.6, 16.6 Hz), 3.09 (s, 3H), 3.12 (dd, 1H, J = 7.0, 16.6 Hz), 3.22 (s, 3H), 3.39 (brs, 2H), 4.60−4.64 (m, 1H), 6.22−6.24 (m, 1H), 7.23−7.34 (m, 3H), 7.48−7.51 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 27.4 (q), 34.8 (q), 35.8 (t), 37.6 (t), 55.2 (d), 118.5 (d), 124.2 (d), 125.4 (d), 126.2 (d), 129.5 (d), 140.4 (s), 141.7 (s), 144.6 (s), 153.9 (s), 167.9 (s); HRMS (ESI, ion trap) calcd for $C_{15}H_{16}N_2O_2$ (M + H^+) 257.1290; found 257.1287.

(5R,6R*)-6-(3,4-Dihydronaphthalen-1-yl)-1,3,5-trimethyldihydropyrimidine-2,4(1H,3H)-dione (**cis-8f**).* White solid (121 mg, 85%); R_f 0.3 (hexanes−ethyl acetate, 2:1); mp 159−160 °C; IR (ATR) 1701, 1649, 1597, 1508, 1474, 826, 816, 775, 754, 741, 712, 673, 660 cm^{-1}; ^1H NMR ($CDCl_3$) δ 0.99 (d, 3H, J = 7.0 Hz), 2.12−2.30 (m, 2H), 2.67−2.71 (m, 2H), 3.03 (s, 3H), 3.13−3.20 (m, 1H), 3.26 (s, 3H), 4.72 (d, 1H, J = 7.0 Hz), 5.82 (dd, 1H, J = 3.5, 5.9 Hz), 7.17−7.28 (m, 4H); ^{13}C NMR ($CDCl_3$) δ 11.9 (q), 22.7 (t), 27.6 (q), 28.0 (t), 34.7 (q), 40.2 (d), 56.8 (d), 121.4 (d), 126.2 (d), 126.6 (d), 127.2 (d), 128.1 (d), 132.2 (s), 134.1 (s), 136.3 (s), 154.1 (s), 170.8 (s). Anal. Calcd for $C_{17}H_{20}N_2O_2$: C, 71.81; H, 7.09; N, 9.85. Found: C, 71.80; H, 7.13; N, 9.76.

(5R,6R*)-6-(1H-Inden-3-yl)-1,3,5-trimethyldihydropyrimidine-2,4(1H,3H)-dione (**cis-8g**).* Colorless paste (99 mg, 73%); R_f 0.45 (hexanes−ethyl acetate, 1:1); IR (ATR) 1707, 1661, 1479, 974, 918, 843, 833, 770, 756, 721, 667 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.15 (d, 3H, J = 7.5 Hz), 3.02 (s, 3H), 3.20−3.27 (m, 1H), 3.29 (s, 3H), 3.39 (brs, 2H), 4.56 (d, 1H, J = 6.9 Hz), 6.23 (t, 1H, J = 1.9 Hz), 7.22−7.28 (m, 1H), 7.29−7.34 (m, 2H), 7.46−7.50 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 11.9 (q), 27.7 (q), 35.0 (q), 38.1 (t), 40.3 (d), 56.0 (d), 118.8 (d), 124.1 (d), 125.3 (d), 126.2 (d), 131.1 (d), 139.9 (s), 143.9 (s), 144.0 (s), 154.0 (s), 171.1 (s); HRMS (ESI, ion trap) calcd for $C_{16}H_{19}N_2O_2$ (M + H^+) 271.1447; found 271.1445.

(5R,6S*)-6-(3,4-Dihydronaphthalen-1-yl)-5-fluoro-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (**cis-8h**).* White solid (112 mg, 78%); R_f 0.5 (hexanes−ethyl acetate, 1:1); mp 152−154 °C; IR (ATR) 1722, 1676, 1508, 1474, 941, 912, 897, 835, 767, 762, 752, 733, 714, 689, 669 cm^{-1}; ^1H NMR ($CDCl_3$) δ 2.21−2.37 (m, 2H), 2.66−2.79 (m, 2H), 3.06 (s, 3H), 3.26 (s, 3H), 5.03 (d, 1H, J = 6.9 Hz), 5.45 (dd, 1H, J = 6.9, J_{HF} = 48.1 Hz), 5.83 (t, 1H, J = 4.6 Hz), 7.14−7.25 (m, 4H); ^{13}C NMR ($CDCl_3$) δ 22.8 (t), 27.71 (t), 27.73 (q), 35.1 (q), 56.3 (d, J_{CCF} = 24.0 Hz), 84.6 (d, J_{CF} = 193.1 Hz), 122.4 (d), 126.2 (d), 127.4 (d), 127.5 (d), 127.8 (d), 129.2 (s), 133.1 (s), 136.2 (s), 153.0 (s), 165.2 (s, J_{CCF} = 21.6 Hz). Anal. Calcd for $C_{16}H_{17}FN_2O_2$: C, 66.65; H, 5.94; N, 9.72. Found: C, 66.71; H, 5.96; N, 9.66.

(5R,6S*)-5-Fluoro-6-(1H-inden-3-yl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (**cis-8i**).* White solid (104 mg, 76%); R_f 0.55 (hexanes−ethyl acetate, 1:1); mp 172−173 °C; IR (ATR) 1730, 1670, 1607, 1504, 974, 961, 922, 881, 858, 789, 779, 766, 752, 745, 718, 698, 677, 669 cm^{-1}; ^1H NMR ($CDCl_3$) δ 3.09 (s, 3H), 3.26 (s, 3H), 3.41 (s, 2H), 4.93 (brd, 1H, J = 6.9 Hz), 5.47 (dd, 1H, J = 6.9, J_{HF} = 47.7 Hz), 6.27 (s, 1H), 7.22−7.27 (m, 1H), 7.29−7.33 (m, 1H), 7.37−7.41 (m, 1H), 7.44−7.47 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 27.7 (q), 35.3 (q), 38.1 (t), 55.2 (d, J_{CCF} = 24.6 Hz), 84.7 (d, J_{CF} = 193.1 Hz), 119.8 (d), 123.8 (d), 125.5 (d), 126.2 (d), 131.7 (d), 137.1 (s), 143.0 (s), 144.0 (s), 152.9 (s), 165.3 (s, J_{CCF} = 21.0 Hz). Anal. Calcd for $C_{15}H_{15}FN_2O_2$: C, 65.68; H, 5.51; N, 10.21. Found: C, 65.66; H, 5.52; N, 10.13.

Dehydrogenation of 8d. To a solution of **8d** (54 mg, 0.20 mmol) in benzene (5 mL) was added DDQ (57 mg, 0.25 mmol), and the mixture was refluxed for 1 h. After filtration, the solvent was removed *in vacuo*. The residue was purified by column chromatography on silica gel (hexanes−EtOAc) to give **10d**[6] (46 mg) in 86% yield.

(5R,6R*)-1,3,5-Trimethyl-6-(naphthalen-1-yl)dihydropyrimidine-2,4(1H,3H)-dione (**cis-10f**).* Colorless paste (51 mg, 90%); R_f 0.55 (hexanes−ethyl acetate, 1:1); IR (ATR) 1705, 1655, 1597, 1508, 1477, 932, 799, 775, 752, 737, 712, 675 cm^{-1}; ^1H NMR ($CDCl_3$) δ 0.95 (d,

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3H, J = 7.0 Hz), 2.97 (s, 3H), 3.37 (s, 3H), 3.38−3.45 (m, 1H), 5.45 (d, 1H, J = 7.3 Hz), 7.16−7.20 (m, 1H), 7.41−7.46 (m, 1H), 7.51−7.60 (m, 2H), 7.82−7.85 (m, 1H), 7.89−7.92 (m, 1H), 8.04−8.08 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 11.5 (q), 27.9 (q), 35.0 (q), 40.3 (d), 56.6 (d), 121.9 (d), 123.3 (d), 125.79 (d), 125.81 (d), 126.6 (d), 129.2 (d), 129.3 (d), 132.1 (s), 132.2 (s), 133.6 (s), 154.0 (s), 170.8 (s); HRMS (ESI, ion trap) calcd for $C_{17}H_{19}N_2O_2$ (M + H^+) 283.1447; found 283.1445.

(5R,6S*)-5-Fluoro-1,3-dimethyl-6-(naphthalen-1-yl)dihydropyrimidine-2,4(1H,3H)-dione* (***cis-10h***). Colorless paste (53 mg, 93%); R_f 0.45 (hexanes−ethyl acetate, 1:1); IR (ATR) 1730, 1670, 1599, 1508, 1477, 908, 885, 799, 775, 750, 727 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 3.06 (s, 3H), 3.34 (s, 3H), 5.65 (dd, 1H, J = 7.5, J_{HF} = 47.5 Hz), 5.75 (dd, 1H, J = 1.7, 7.5 Hz), 7.08−7.12 (m, 1H), 7.43−7.47 (m, 1H), 7.51−7.60 (m, 2H), 7.86−7.91 (m, 2H), 7.99−8.04 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 27.9 (q), 35.3 (q), 56.6 (d, J_{CCF} = 25.2 Hz), 84.5 (d, J_{CF} = 191.9 Hz), 122.9 (d), 123.0 (d), 125.2 (d), 126.0 (d), 126.5 (d), 127.9 (s), 128.8 (d), 130.0 (d), 132.4 (s), 133.9 (s), 153.2 (s), 165.0 (s, J_{CCF} = 21.6 Hz); HRMS (ESI, ion trap) calcd for $C_{16}H_{16}FN_2O_2$ (M + H^+) 287.1196; found 287.1195.

Treatment of the Adducts with TBAF. To a solution of **3a** (99 mg, 0.25 mmol) in THF (5 mL) was added 1 M TBAF in THF (0.25 mL), and the solution was stirred at 25 °C under a nitrogen atmosphere for 15 min. After addition of AcOH (15 mg, 0.25 mmol), the solvent was removed *in vacuo*. The residue was purified by column chromatography on silica gel (hexanes−EtOAc) to give **12a** (71 mg) in 87% yield.

Treatment of the Adducts with 1 M HCl−MeOH. To a solution of **3a** (99 mg, 0.25 mmol) in MeOH (2.5 mL) was added 2 M HCl in MeOH (2.5 mL), and the solution was stirred at 25 °C for 15 min. After the solvent was removed *in vacuo*, the residue was purified by column chromatography on silica gel (hexanes−EtOAc) to give **11a** (75 mg) in 93% yield.

6-(Hydroxydiphenylmethyl)-1,3-dimethyldihydropyrimidine-2,4-(1H,3H)-dione (***11a***). White solid (71 mg, 93%); R_f 0.35 (hexanes−ethyl acetate, 1:1); mp 197−198 °C; IR (ATR) 3340, 1701, 1641, 1520, 1489, 982, 941, 920, 770, 758, 748, 706, 696, 687, 669 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 2.52 (s, 3H), 2.80−2.92 (m, 3H), 2.95 (s, 3H), 4.30 (d, 1H, J = 6.7 Hz), 7.26−7.42 (m, 10H); ^{13}C NMR ($CDCl_3$) δ 27.0 (q), 32.8 (t), 38.3 (q), 61.2 (d), 81.0 (s), 126.5 (d), 126.6 (d), 127.6 (d), 127.7 (d), 128.2 (d), 128.4 (d), 142.8 (s), 143.3 (s), 154.0 (s), 168.9 (s). Anal. Calcd for $C_{19}H_{20}N_2O_3$: C, 70.35; H, 6.21; N, 8.64. Found: C, 70.30; H, 6.17; N, 8.52.

6-(Bis(4-fluorophenyl)(hydroxy)methyl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (***11b***). White solid (83 mg, 92%); R_f 0.25 (hexanes−ethyl acetate, 1:1); mp 218−219 °C; IR (ATR) 3451, 3335, 1692, 1647, 1603, 1503, 1491, 980, 951, 833, 816, 804, 773, 758, 734, 696, 677, 669 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 2.55 (s, 3H), 2.77 (d, 1H, J = 16.8 Hz), 2.90 (dd, 1H, J = 7.8, 16.8 Hz), 2.94 (s, 3H), 4.24 (d, 1H, J = 7.8 Hz), 6.99−7.10 (m, 4H), 7.29−7.34 (m, 2H), 7.34−7.39 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 26.5 (q), 32.4 (t), 38.1 (q), 60.8 (d), 79.8 (s), 114.4 (d, J_{CCF} = 20.7 Hz), 114.5 (d, J_{CCF} = 21.0 Hz), 128.3 (d, J_{CCCF} = 7.5 Hz), 128.5 (d, J_{CCCF} = 8.1 Hz), 138.5 (s, J_{CCCCF} = 3.0 Hz), 139.1 (s, J_{CCCCF} = 3.0 Hz), 153.5 (s), 161.4 (s, J_{CF} = 246.8 Hz), 161.5 (s, J_{CF} = 247.4 Hz), 168.3 (s). Anal. Calcd for $C_{19}H_{18}F_2N_2O_3$: C, 63.33; H, 5.03; N, 7.77. Found: C, 63.31; H, 5.05; N, 7.70.

6-(Hydroxybis(4-methoxyphenyl)methyl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (***11c***). White solid (82 mg, 85%); R_f 0.4 (hexanes−ethyl acetate, 1:2); mp 198−199 °C; IR (ATR) 3447, 3379, 1695, 1641, 1607, 1582, 1508, 1491, 980, 951, 932, 916, 897, 826, 812, 800, 787, 756, 739, 692, 681, 669 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 2.43 (brs, 1H), 2.63 (s, 3H), 2.86−2.90 (m, 2H), 2.93 (s, 3H), 3.79 (s, 3H), 3.81 (s, 3H), 4.22 (dd, 1H, J = 2.9, 5.7 Hz), 6.83−6.91 (m, 4H), 7.24−7.28 (m, 2H), 7.28−7.33 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 27.0 (q), 32.9 (t), 38.5 (q), 55.17 (q), 55.21 (q), 61.3 (d), 80.6 (s), 113.6 (d), 113.7 (d), 127.9 (d), 128.0 (d), 134.7 (s), 135.3 (s), 154.0 (s), 158.96 (s), 158.98 (s), 168.9 (s). Anal. Calcd for $C_{21}H_{24}N_2O_5$: C, 65.61; H, 6.29; N, 7.29. Found: C, 65.57; H, 6.33; N, 7.23.

6-(5-Hydroxy-10,11-dihydro-5H-dibenzo[a,d][7]annulen-5-yl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (***11d***). White solid (71 mg, 81%); R_f 0.4 (hexanes−ethyl acetate, 1:1); mp 231−232 °C; IR (ATR) 3329, 1717, 1634, 1489, 984, 974, 964, 920, 772, 756, 729, 692, 677 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 2.32 (d, 1H, J = 2.2 Hz), 2.34 (s, 3H), 2.51(dd, 1H, J = 7.3, 16.5 Hz), 2.58 (d, 1H, J = 16.5 Hz), 2.95−3.07 (m, 4H), 3.17 (s, 3H), 3.27−3.34 (m, 1H), 3.36−3.44 (m, 1H), 4.04 (d, 1H, J = 7.3 Hz), 7.09−7.13 (m, 1H), 7.18−7.35 (m, 5H), 7.79−7.83 (m, 1H), 7.87−7.91 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 27.1 (q), 32.0 (t), 34.6 (t), 35.3 (t), 37.2 (q), 62.8 (d), 82.2 (s), 126.97 (d), 127.00 (d), 127.9 (d), 128.2 (d), 128.5 (d), 128.7 (d), 130.1 (d), 131.0 (d), 139.0 (s), 139.3 (s), 140.0 (s), 141.1 (s), 154.1 (s), 169.6 (s). Anal. Calcd for $C_{21}H_{22}N_2O_3$: C, 71.98; H, 6.33; N, 7.99. Found: C, 71.95; H, 6.32; N, 7.90.

6-(5-Hydroxy-5H-dibenzo[a,d][7]annulen-5-yl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (***11e***). White solid (61 mg, 70%); R_f 0.3 (hexanes−ethyl acetate, 1:1); mp 246−248 °C; IR (ATR) 3458, 1701, 1655, 1483, 974, 935, 910, 814, 806, 795, 760, 727, 694, 673 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 2.02 (d, 1H, J = 16.6 Hz), 2.15 (s, 3H), 2.36 (dd, 1H, J = 8.0, 16.6 Hz), 3.05 (d, 1H, J = 1.2 Hz), 3.19 (s, 3H), 4.34 (d, 1H, J = 8.0 Hz), 7.04 (s, 2H), 7.32−7.39 (m, 4H), 7.43−7.50 (m, 2H), 7.72−7.76 (m, 1H), 7.81−7.85 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 27.2 (q), 31.8 (t), 37.0 (q), 53.5 (d), 80.5 (s), 124.6 (d), 124.9 (d), 127.6 (d), 127.8 (d), 129.4 (d), 129.6 (d), 129.7 (d), 129.8 (d), 131.2 (d), 132.0 (s), 132.2 (d), 133.1 (s), 138.7 (s), 140.3 (s), 154.5 (s), 169.5 (s). Anal. Calcd for $C_{21}H_{20}N_2O_3$: C, 72.40; H, 5.79; N, 8.04. Found: C, 72.36; H, 5.81; N, 7.95.

6-(9-Hydroxy-9,10-dihydroanthracen-9-yl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (***11f***). White solid (68 mg, 81%); R_f 0.4 (hexanes−ethyl acetate, 1:1); mp 250−251 °C; IR (ATR) 3337, 1709, 1636, 1522, 1481, 976, 970, 947, 937, 914, 887, 772, 760, 725, 698, 679 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 2.32 (s, 1H), 2.55 (dd, 1H, J = 7.6, 17.2 Hz), 2.62 (s, 3H), 2.64 (dd, 1H, J = 1.2, 17.2 Hz), 2.79 (s, 3H), 3.59 (dd, 1H, J = 1.2, 7.6 Hz), 3.97 (d, 1H, J = 20.1 Hz), 4.15 (d, 1H, J = 20.1 Hz), 7.29−7.38 (m, 6H), 7.72−7.75 (m, 1H), 7.77−7.80 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 26.6 (q), 31.7 (t), 34.4 (t), 38.0 (q), 63.8 (d), 76.0 (s), 125.9 (d), 126.1 (d), 126.7 (d), 127.0 (d), 127.5 (d), 127.8 (d), 128.0 (d), 128.1 (d), 134.6 (s), 138.1 (s), 138.3 (s), 153.3 (s), 168.6 (s). Anal. Calcd for $C_{20}H_{20}N_2O_3$: C, 71.41; H, 5.99; N, 8.33. Found: C, 71.47; H, 6.01; N, 8.24.

6-(9-Hydroxy-9H-xanthen-9-yl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (***11g***). Colorless paste (74 mg, 88%); R_f 0.3 (hexanes−ethyl acetate, 1:1); IR (ATR) 3362, 1705, 1647, 1601, 1574, 1483, 1474, 914, 895, 870, 816, 754, 731, 692, 677, 669 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 2.45 (dd, 1H, J = 1.2, 17.3 Hz), 2.52 (dd, 1H, J = 7.8, 17.3 Hz), 2.52 (s, 3H), 2.78 (brs, 1H), 3.00 (s, 3H), 3.59 (dd, 1H, J = 1.2, 7.8 Hz), 7.14−7.24 (m, 4H), 7.31−7.39 (m, 2H), 7.56−7.60 (m, 1H), 7.65−7.69 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 26.6 (q), 31.4 (t), 38.4 (q), 64.1 (d), 71.7 (s), 116.4 (d), 116.6 (d), 123.3 (s), 123.4 (d), 123.8 (d), 124.5 (s), 126.0 (d), 126.6 (d), 129.7 (d), 129.9 (d), 150.7 (s), 150.8 (s), 153.2 (s), 167.7 (s); HRMS (ESI, ion trap) calcd for $C_{19}H_{19}N_2O_4$ (M + H^+) 339.1345; found 339.1342.

(5R,6R*)-6-(Hydroxydiphenylmethyl)-1,3,5-trimethyldihydropyrimidine-2,4(1H,3H)-dione* (***cis-11h***). White solid (42 mg, 50%); R_f 0.4 (hexanes−ethyl acetate, 1:1); mp 104−106 °C; IR (ATR) 3510, 3416, 3238, 1701, 1659, 1491, 976, 881, 831, 777, 762, 752, 719, 704, 694, 669 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 0.86 (d, 3H, J = 7.5 Hz), 2.22 (s, 3H), 2.71 (brs, 1H), 3.00−3.07 (m, 1H), 3.25 (s, 3H), 4.24 (d, 1H, J = 5.8 Hz), 7.24−7.38 (m, 6H), 7.42−7.46 (m, 2H), 7.49−7.53 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 13.2 (q), 27.9 (q), 37.3 (q), 39.4 (d), 66.5 (d), 81.2 (s), 125.9 (d), 126.8 (d), 127.5 (d), 127.7 (d), 128.2 (d), 128.3 (d), 143.4 (s), 143.6 (s), 154.1 (s), 173.0 (s). Anal. Calcd for $C_{20}H_{22}N_2O_3$: C, 70.99; H, 6.55; N, 8.28. Found: C, 70.91; H, 6.52; N, 8.10.

(5R,6S*)-6-(Hydroxydiphenylmethyl)-1,3,5-trimethyldihydropyrimidine-2,4(1H,3H)-dione* (***trans-11h***). White solid (42 mg, 50%); R_f 0.4 (hexanes−ethyl acetate, 1:1); mp 184−186 °C; IR (ATR) 3429, 1694, 1649, 1489, 999, 880, 820, 756, 746, 739, 696, 660 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 1.34 (d, 3H, J = 7.3 Hz), 2.51 (brs, 1H), 2.54 (s, 3H), 2.96 (q, 1H, J = 7.3 Hz), 2.99 (s, 3H), 3.93 (s, 1H), 7.27−7.42 (m, 10H); ^{13}C NMR ($CDCl_3$) δ 17.7 (q), 27.1 (q), 37.7 (d), 39.0 (d), 68.6 (d), 81.1 (s), 126.2 (d), 126.5 (d), 127.7 (d), 127.8 (d), 128.3 (d),

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128.5 (d), 142.9 (s), 143.5 (s), 153.7 (s), 172.9 (s). Anal. Calcd for $C_{20}H_{22}N_2O_3$: C, 70.99; H, 6.55; N, 8.28. Found: C, 70.93; H, 6.58; N, 8.16.

(5R,6R*)-6-(Bis(4-fluorophenyl)(hydroxy)methyl)-1,3,5-trimethyldihydropyrimidine-2,4(1H,3H)-dione (**cis-11i**).* White solid (39 mg, 42%); R_f 0.55 (hexanes−ethyl acetate, 1:2); mp 220−222 °C; IR (ATR) 3397, 1701, 1655, 1599, 1504, 1483, 839, 822, 814, 804, 772, 754, 696, 669 cm^{-1}; ^1H NMR ($CDCl_3$) δ 0.85 (d, 3H, J = 7.3 Hz), 2.27 (s, 3H), 2.93 (brs, 1H), 3.01−3.07 (m, 1H), 3.21 (s, 3H), 4.16 (d, 1H, J = 5.4 Hz), 6.98−7.03 (m, 2H), 7.11−7.16 (m, 2H), 7.21−7.26 (m, 2H), 7.46−7.51 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 13.3 (q), 27.6 (q), 37.5 (q), 39.4 (d), 66.6 (d), 80.9 (s), 115.30 (d, J_{CCF} = 21.6 Hz), 115.32 (d, J_{CCF} = 21.6 Hz), 127.7 (d, J_{CCCF} = 8.4 Hz), 128.7 (d, J_{CCCF} = 7.8 Hz), 139.17 (s, J_{CCCCF} = 3.6 Hz), 139.22 (s, J_{CCCCF} = 3.6 Hz), 154.0 (s), 162.1 (s, J_{CF} = 248.3 Hz), 162.3 (s, J_{CF} = 248.3 Hz), 172.8 (s). Anal. Calcd for $C_{20}H_{20}F_2N_2O_3$: C, 64.16; H, 5.38; N, 7.48. Found: C, 64.13; H, 5.36; N, 7.45.

(5R,6S*)-6-(Bis(4-fluorophenyl)(hydroxy)methyl)-1,3,5-trimethyldihydropyrimidine-2,4(1H,3H)-dione (**trans-11i**).* White solid (32 mg, 34%); R_f 0.6 (hexanes−ethyl acetate, 1:2); mp 230−231 °C; IR (ATR) 3410, 1711, 1647, 1601, 1503, 1497, 1489, 997, 982, 880, 837, 824, 806, 772, 758, 667, 658 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.35 (d, 3H, J = 7.4 Hz), 2.58 (s, 3H), 2.86 (s, 1H), 2.88 (q, 1H, J = 7.4 Hz), 2.96 (s, 3H), 3.86 (s, 1H), 7.00−7.10 (m, 4H), 7.26−7.30 (m, 2H), 7.31−7.36 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 17.8 (q), 27.2 (q), 37.7 (d), 39.3 (q), 68.7 (d), 80.7 (s), 115.5 (d, J_{CCF} = 21.6 Hz), 115.6 (d, J_{CCF} = 21.6 Hz), 128.2 (d, J_{CCCF} = 8.4 Hz), 128.4 (d, J_{CCCF} = 8.4 Hz), 138.5 (s, J_{CCCCF} = 2.7 Hz), 139.1 (s, J_{CCCCF} = 2.7 Hz), 153.5 (s), 162.2 (s, J_{CF} = 248.9 Hz), 162.3 (s, J_{CF} = 248.3 Hz), 172.6 (s). Anal. Calcd for $C_{20}H_{20}F_2N_2O_3$: C, 64.16; H, 5.38; N, 7.48. Found: C, 64.10; H, 5.38; N, 7.44.

(5R,6R*)-6-(Hydroxybis(4-methoxyphenyl)methyl)-1,3,5-trimethyldihydropyrimidine-2,4(1H,3H)-dione (**cis-11j**).* Colorless paste (59 mg, 59%); R_f 0.4 (hexanes−ethyl acetate, 1:2); IR (ATR) 3379, 1699, 1647, 1607, 1578, 1508, 1489, 908, 881, 822, 799, 789, 772, 758, 727, 669 cm^{-1}; ^1H NMR ($CDCl_3$) δ 0.87 (d, 3H, J = 6.9 Hz), 2.28 (s, 3H), 2.61 (brs, 1H), 2.98−3.05 (m, 1H), 3.23 (s, 3H), 3.77 (s, 3H), 3.84 (s, 3H), 4.13 (d, 1H, J = 5.9 Hz), 6.80−6.85 (m, 2H), 6.92−6.96 (m, 2H), 7.15−7.19 (m, 2H), 7.37−7.41 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 13.2 (q), 27.5 (q), 37.3 (q), 39.4 (d), 55.1 (q), 55.2 (q), 66.6 (d), 80.7 (s), 113.4 (d), 113.5 (d), 127.1 (d), 128.1 (d), 135.8 (s), 135.9 (s), 154,1 (s), 158.6 (s), 158.9 (s), 173.0 (s); HRMS (ESI, ion trap) calcd for $C_{22}H_{27}N_2O_5$ ($M + H^+$) 399.1920; found 399.1918.

(5R,6R*)-6-(Methoxybis(4-methoxyphenyl)methyl)-1,3,5-trimethyldihydropyrimidine-2,4(1H,3H)-dione (**cis-11j′**).* White solid (54 mg, 52%); R_f 0.3 (hexanes−ethyl acetate, 1:1); mp 193−195 °C; IR (ATR) 1705, 1659, 1609, 1576, 1508, 1489, 988, 964, 943, 903, 885, 843, 827, 806, 799, 777, 764, 752, 729, 667 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.00 (d, 3H, J = 7.5 Hz), 2.47 (s, 3H), 2.67 (s, 3H), 2.88−2.96 (m, 1H), 3.17 (s, 3H), 3.83 (s, 3H), 3.84 (s, 3H), 4.41 (d, 1H, J = 6.7 Hz), 6.87−6.95 (m, 4H), 7.32−7.37 (m, 2H), 7.40−7.46 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 13.4 (q), 27.2 (q), 39.2 (d), 39.9 (q), 50.3 (q), 55.17 (q), 55.22 (q), 67.2 (d), 86.6 (s), 113.2 (d), 113.4 (d), 127.8 (s), 129.1 (s), 130.1 (d), 130.7 (d), 153.7 (s), 159.0 (s), 159.3 (s), 170.8 (s). Anal. Calcd for $C_{23}H_{28}N_2O_5$: C, 66.97; H, 6.84; N, 6.79. Found: C, 66.95; H, 6.87; N, 6.72.

(5R,6S*)-6-(Hydroxybis(4-methoxyphenyl)methyl)-1,3,5-trimethyldihydropyrimidine-2,4(1H,3H)-dione (**trans-11j**).* White solid (19 mg, 19%); R_f 0.45 (hexanes−ethyl acetate, 1:2); mp 217−218 °C; IR (ATR) 3385, 1697, 1647, 1609, 1582, 1508, 1489, 910, 891, 829, 802, 756, 729, 683, 669 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.34 (d, 3H, J = 7.2 Hz), 2.49 (brs, 1H), 2.62, (s, 3H), 2.93 (q, 1H, J = 7.2 Hz), 2.95 (s, 3H), 3.79 (s, 3H), 3.81 (s, 3H), 3.83 (s, 1H), 6.83−6.86 (m, 2H), 6.87−6.91 (m, 2H), 7.20−7.24 (m, 2H), 7.25−7.29 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 17.8 (q), 27.2 (q), 37.8 (d), 39.2 (q), 55.2 (q), 55.3 (q), 68.8 (d), 80.8 (s), 113.7 (d), 113.8 (d), 127.7 (d), 127.9 (d), 134.9 (s), 135.7 (s), 153.7 (s), 159.07 (s), 159.10 (s), 172.9 (s). Anal. Calcd for $C_{22}H_{26}N_2O_5$: C, 66.32; H, 6.58; N, 7.03. Found: C, 66.28; H, 6.57; N, 6.96.

(5R,6S*)-5-Fluoro-6-(hydroxydiphenylmethyl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (**cis-11k**).* White solid (78 mg, 91%); R_f 0.35 (hexanes−ethyl acetate, 1:1); mp 240−242 °C; IR (ATR) 3451, 3381, 1728, 1653, 1489, 951, 914, 899, 866, 793, 770, 752, 727, 698 cm^{-1}; ^1H NMR ($CDCl_3$) δ 2.30 (s, 3H), 2.65 (s, 1H), 3.16 (s, 3H), 4.71 (d, 1H, J = 6.9 Hz), 5.35 (dd, 1H, J = 6.9 Hz, J_{HF} = 47.1 Hz), 7.21−7.45 (m, 10H); ^{13}C NMR ($CDCl_3$) δ 27.5 (q), 38.2 (q), 64.0 (d, J_{CCF} = 21.6 Hz), 80.7 (s), 83.1 (d, J_{CF} = 196.1 Hz), 126.1 (d), 126.8 (d), 127.9 (d), 128.0 (d), 128.4 (d), 143.3 (s), 143.6 (s), 152.8 (s), 166.8 (s). Anal. Calcd for $C_{19}H_{19}FN_2O_3$: C, 66.66; H, 5.59; N, 8.18. Found: C, 66.73; H, 5.66; N, 8.05.

(5R,6S*)-6-(Bis(4-fluorophenyl)(hydroxy)methyl)-5-fluoro-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (**cis-11l**).* White solid (88 mg, 93%); R_f 0.25 (hexanes−ethyl acetate, 1:1); mp 235 °C; IR (ATR) 3402, 1727, 1649, 1603, 1506, 1489, 993, 930, 905, 870, 831, 810, 773, 756, 733, 716 cm^{-1}; ^1H NMR ($CDCl_3$) δ 2.34 (s, 3H), 2.69 (s, 1H), 3.17 (s, 3H), 4.67 (d, 1H, J = 6.9 Hz), 5.41 (dd, 1H, J = 6.9 Hz, J_{HF} = 47.0 Hz), 6.99−7.04 (m, 2H), 7.05−7.11 (m, 2H), 7.18−7.22 (m, 2H), 7.38−7.43 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 26.0 (q), 37.0 (q), 62.5 (d, J_{CCF} = 21.0 Hz), 78.2 (s), 81.8 (d, J_{CF} = 195.5 Hz), 113.2 (d, J_{CCF} = 21.0 Hz), 113.6 (d, J_{CCF} = 21.6 Hz), 127.4 (d, J_{CCCF} = 7.2 Hz), 128.0 (d, J_{CCCF} = 8.4 Hz), 138.8 (s), 139.1 (s, J_{CCCCF} = 2.4 Hz), 151.8 (s), 160.3 (s, J_{CF} = 245.3 Hz), 160.7 (s, J_{CF} = 246.5 Hz), 165.8 (s, J_{CCF} = 21.6 Hz). Anal. Calcd for $C_{19}H_{17}F_3N_2O_3$: C, 60.32; H, 4.53; N, 7.40. Found: C, 60.29; H, 4.54; N, 7.26.

(5R,6S*)-5-Fluoro-6-(hydroxybis(4-methoxyphenyl)methyl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (**cis-11m**).* White solid (80 mg, 80%); R_f 0.3 (hexanes−ethyl acetate, 1:1); mp 245−246 °C; IR (ATR) 3478, 1724, 1653, 1609, 1580, 1508, 1489, 995, 905, 866, 831, 824, 810, 795, 781, 754, 737 cm^{-1}; ^1H NMR ($CDCl_3$) δ 2.34 (s, 3H), 2.62 (brs, 1H), 3.16 (s, 3H), 3.78 (s), 3.81 (s), 4.63 (d, 1H, J = 6.9 Hz), 5.38 (dd, 1H, J = 6.9 Hz, J_{HF} = 47.0 Hz), 6.81−6.85 (m, 2H), 6.87−6.91 (m, 2H), 7.12−7.16 (m, 2H), 7.31−7.35 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 26.1 (q), 37.0 (q), 54.1 (q), 62.8 (d, J_{CCF} = 20.4 Hz), 78.3 (s), 82.0 (d, J_{CF} = 195.5 Hz), 111.8 (d), 112.1 (d), 126.7 (d), 127.3 (d), 135.3 (s), 135.6 (s). 151.9 (s), 157.1 (s), 157.5 (s), 166.1 (s, J_{CCF} = 21.6 Hz). Anal. Calcd for $C_{21}H_{23}FN_2O_5$: C, 62.68; H, 5.76; N, 6.96. Found: C, 62.70; H, 5.77; N, 6.89.

(5R,6S*)-5-Fluoro-6-(5-hydroxy-10,11-dihydro-5H-dibenzo[a,d]-[7]annulen-5-yl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (**cis-11n**).* White solid (77 mg, 84%); R_f 0.4 (hexanes−ethyl acetate, 1:1); mp 242−244 °C; IR (ATR) 3497, 1713, 1670, 1506, 1477, 924, 910, 893, 870, 864, 789, 775, 762, 752, 723, 712, 669 cm^{-1}; ^1H NMR ($CDCl_3$) δ 2.33 (s, 3H), 2.69 (brd, 1H, J = 2.4 Hz), 2.97−3.12 (m, 2H), 3.24 (s, 3H), 3.29−3.36 (m, 1H), 3.47−3.55 (m, 1H), 4.61 (dd, 1H, J = 2.3, 6.7 Hz), 5.13 (dd, 1H, J = 6.7 Hz, J_{HF} = 46.4 Hz), 7.08−7.13 (m, 1H), 7.18−7.32 (m, 5H), 7.72−7.76 (m, 1H), 7.76−7.82 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 26.3 (q), 32.9 (t), 34.1 (t), 36.2 (q), 64.1 (d, J_{CCF} = 19.2 Hz), 79.2 (s), 82.7 (d, J_{CF} = 198.5 Hz), 125.0 (d), 125.4 (d), 126.91 (d), 126.94 (d), 127.6 (d), 128.1 (d), 128.5 (d), 130.3 (d), 137.5 (s), 137.6 (s), 139.4 (s), 141.3 (s), 152.2 (s), 166.4 (s, J_{CCF} = 20.4 Hz). Anal. Calcd for $C_{21}H_{21}FN_2O_3$: C, 68.47; H, 5.75; N, 7.60. Found: C, 68.49; H, 5.78; N, 7.52.

*N-Methyl-2-(3-methyl-2-oxo-5,5-diphenyloxazolidin-4-yl)acetamide (**12a**).* White solid (71 mg, 87%); R_f 0.3 (ethyl acetate); mp 144−145 °C; IR (ATR) 3366, 1746, 1661, 1545, 1495, 986, 945, 928, 918, 841, 797, 766, 760, 754, 700, 671, 656 cm^{-1}; ^1H NMR ($CDCl_3$) δ 2.03 (dd, 1H, J = 8.3, 15.2 Hz), 2.31 (dd, 1H, J = 5.0, 15.2 Hz), 2.50 (d, 3H, J = 5.0 Hz), 2.87 (s, 3H), 4.73 (brs, 1H), 5.08 (dd, 1H, J = 5.0, 8.3 Hz), 7.22−7.44 (m, 8H), 7.65−7.69 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 26.3 (q), 29.5 (q), 36.8 (t), 62.3 (d), 86.7 (s), 126.1 (d), 126.8 (d), 127.9 (d), 128.0 (d), 128.47 (d), 128.53 (d), 138.5 (s), 142.1 (s), 156.4 (s), 169.7 (s). Anal. Calcd for $C_{19}H_{20}N_2O_3$: C, 70.35; H, 6.21; N, 8.64. Found: C, 70.31; H, 6.22; N, 8.55.

*2-(5,5-Bis(4-fluorophenyl)-3-methyl-2-oxooxazolidin-4-yl)-N-methylacetamide (**12b**).* White solid (62 mg, 69%); R_f 0.2 (ethyl acetate); mp 197−199 °C; IR (ATR) 3275, 1748, 1670, 1645, 1601, 1578, 1506, 1477, 957, 926, 849, 835, 812, 795, 775, 758, 723, 714, 706, 662 cm^{-1}; ^1H NMR ($CDCl_3$) δ 2.00 (dd, 1H, J = 8.5, 15.3 Hz), 2.36 (dd, 1H, J = 4.9, 15.3 Hz), 2.55 (d, 3H, J = 5.0 Hz), 2.87 (s, 3H), 5.05

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(dd, 1H, J = 4.9, 8.5 Hz), 5.06 (brs, 1H), 6.95−7.01 (m, 2H), 7.08−7.13 (m, 2H), 7.25−7.30 (m, 2H), 7.63−7.68 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 26.2 (q), 29.5 (q), 36.5 (t), 62.5 (d), 85.9 (s), 114.9 (d, J_{CCF} = 21.6 Hz), 115.6 (d, J_{CCF} = 21.6 Hz), 128.0 (d, J_{CCCF} = 8.4 Hz), 129.0 (d, J_{CCCF} = 8.4 Hz), 134.3 (s, J_{CCCCF} = 2.7 Hz), 137.9 (s, J_{CCCCF} = 3.3 Hz), 156.0 (s), 162.2 (s, J_{CF} = 249.2 Hz), 162.6 (s, J_{CF} = 248.6 Hz), 169.4 (s). Anal. Calcd for $C_{19}H_{18}F_2N_2O_3$: C, 63.33; H, 5.03; N, 7.77. Found: C, 63.29; H, 5.04; N, 7.72.

*2-(5,5-Bis(4-methoxyphenyl)-3-methyl-2-oxooxazolidin-4-yl)-N-methylacetamide (**12c**).* White solid (80 mg, 83%); R_f 0.15 (hexanes−ethyl acetate, 1:2); mp 105−107 °C; IR (ATR) 3350, 1749, 1653, 1609, 1578, 1558, 1512, 988, 949, 930, 914, 903, 837, 824, 775, 756, 740, 729, 667 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 2.02 (dd, 1H, J = 8.1, 15.0 Hz), 2.30 (dd, 1H, J = 5.2, 15.0 Hz), 2.53 (d, 3H, J = 4.9 Hz), 2.87 (s, 3H), 3.76 (s, 3H), 3.81 (s, 3H), 5.01 (dd, 1H, J = 5.2, 8.1 Hz), 5.04 (q, 1H, J = 4.9 Hz), 6.78−6.82 (m, 2H), 6.89−6.94 (m, 2H), 7.17−7.22 (m, 2H), 7.53−7.57 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 26.2 (q), 29.5 (q), 36.9 (t), 55.1 (q), 55.2 (q), 62.5 (d), 86.5 (s), 113.2 (d), 113.7 (d), 127.4 (d), 128.2 (d), 131.0 (s), 134.4 (s), 156.6 (s), 159.0 (s), 159.4 (s), 169.8 (s). Anal. Calcd for $C_{21}H_{24}N_2O_5$: C, 65.61; H, 6.29; N, 7.29. Found: C, 65.54; H, 6.30; N, 7.22.

*N-Methyl-2-(3′-methyl-2′-oxo-10,11-dihydrospiro[dibenzo[a,d][7]annulene-5,5′-oxazolidin]-4′-yl)acetamide (**12d**).* Colorless paste (74 mg, 84%); R_f 0.2 (hexanes−ethyl acetate, 1:2); IR (ATR) 3310, 1748, 1647, 1558, 1541, 1483, 999, 908, 772, 725, 667, 664 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 2.13−2.22 (m, 2H), 2.60 (d, 3H, J = 5.0 Hz), 2.79 (s, 3H), 2.90−3.02 (m, 2H), 3.23−3.31 (m, 1H), 3.62−3.71 (m, 1H), 4.71 (t, 2H, J = 6.0 Hz), 5.14 (brs, 1H), 7.10−7.15 (m, 1H), 7.20−7.29 (m, 5H), 7.65−7.68 (m, 1H), 7.81−7.85 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 26.4 (q), 29.9 (q), 31.4 (t), 33.3 (t), 38.4 (t), 66.2 (d), 86.0 (s), 124.0 (d), 126.19 (d), 126.20 (d), 126.4 (d), 128.47 (d), 128.54 (d), 130.6 (d), 131.3 (d), 135.0 (s), 137.7 (s), 137.9 (s), 140.7 (s), 156.4 (s), 169.9 (s); HRMS (ESI, ion trap) calcd for $C_{21}H_{23}N_2O_3$ (M + H$^+$) 351.1709; found 351.1706.

*N-Methyl-2-(3′-methyl-2′-oxospiro[dibenzo[a,d][7]annulene-5,5′-oxazolidin]-4′-yl)acetamide (**12e**).* White solid (74 mg, 85%); R_f 0.25 (ethyl acetate); mp 193−195 °C; IR (ATR) 3316, 1748, 1647, 1558, 1485, 999, 955, 908, 885, 858, 800, 772, 725, 669 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 1.76−1.82 (m, 1H), 1.88 (dd, 1H, J = 3.6, 14.9 Hz), 2.67 (d, 3H, J = 4.7 Hz), 2.70 (s, 3H), 4.34 (dd, 1H, J = 3.6, 8.6 Hz), 5.34 (brs, 1H), 6.99 (d, 1H, J = 11.7 Hz), 7.17 (d, 1H, J = 11.7 Hz), 7.31−7.38 (m, 3H), 7.39−7.46 (m, 3H), 7.80−7.84 (m, 1H), 7.85−7.89 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 26.2 (q), 30.2 (q), 38.2 (t), 62.0 (d), 84.9 (s), 122.9 (d), 124.1 (d), 127.5 (d), 127.6 (d), 128.7 (d), 128.8 (d), 129.0 (d), 129.3 (d), 130.7 (d), 131.5 (s), 131.8 (d), 132.2 (s), 135.1 (s), 139.6 (s), 156.2 (s), 169.8 (s). Anal. Calcd for $C_{21}H_{20}N_2O_3$: C, 72.40; H, 5.79; N, 8.04. Found: C, 72.32; H, 5.78; N, 7.91.

*N-Methyl-2-(3-methyl-2-oxo-5,5-diphenyloxazolidin-4-yl)-propanamide (**12h**).* Colorless paste (49 mg, 58%, *erythro*:*threo* = 70:30 dr); R_f 0.3, 0.35 (acetate); IR (ATR) 3339, 1740, 1647, 1541, 1494, 912, 893, 756, 729, 696, 671, 646, 617, 666 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 0.86 (d, 0.9H, J = 6.9 Hz), 1.09 (d, 2.1H, J = 7.5 Hz), 2.40−2.47 (m, 0.7H), 2.53 (d, 2.1H, J = 4.6 Hz), 2.59 (d, 0.9H, J = 4.6 Hz), 2.67−2.74 (m, 0.3H), 2.88 (s, 0.9H), 2.92 (s, 2.1H), 4.96 (brs, 0.9H), 5.02 (dd, 0.3H, J = 2.9, 5.1 Hz), 5.09 (d, 0.7H, J = 5.7 Hz), 5.28 (brs, 0.3H), 7.18−7.46 (m, 8H), 7.63−7.68 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 11.2 (q), 13.3 (q), 26.48 (q), 26.51 (q), 30.3 (q), 32.5 (q), 40.6 (d), 42.7 (d), 66.0 (d), 67.2 (d), 87.1 (s), 87.7 (s), 125.5 (d), 125.8 (d), 126.8 (d), 127.2 (d), 127.77 (d), 127.81 (d), 128.1 (d), 128.30 (d), 128.34 (d), 128.6 (d), 137.8 (s), 138.3 (s), 143.1 (s), 143.6 (s), 156.8 (s), 157.2 (s), 172.7 (s), 174.1 (s); HRMS (ESI, ion trap) calcd for $C_{20}H_{23}N_2O_3$ (M + H$^+$) 339.1709; found 339.1707.

*2-(5,5-Bis(4-fluorophenyl)-3-methyl-2-oxooxazolidin-4-yl)-N-methylpropanamide (**12i**).* Colorless paste (51 mg, 54%, *erythro*:*threo* = 78:22 dr); R_f 0.15 (hexanes−ethyl acetate, 1:2); IR (ATR) 3339, 1744, 1647, 1601, 1541, 1508, 908, 899, 831, 806, 762, 754, 727, 677, 662 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 0.86 (d, 0.66H, J = 6.9 Hz), 1.11 (d, 2.34H, J = 6.9 Hz), 2.33−2.40 (m, 0.78H), 2.55 (d, 2.34H, J = 4.6 Hz), 2.61 (d, 0.66H, J = 4.9 Hz), 2.68−2.74 (m, 0.22H), 2.87 (s, 0.66H), 2.93 (s, 2.34H), 5.00 (brs, 0,78H), 5.02 (d, 0.78H, J = 6.9 Hz), 5.07 (d, 0.22H, J = 4.8 Hz), 5.29−5.33 (m, 0.22H), 6.94−7.02 (m, 2H), 7.04−7.11 (m, 2H), 7.31−7.42 (m, 2H), 7.61−7.67 (m, 2H); ^{13}C NMR ($CDCl_3$) δ major; 13.8 (q), 26.4 (q), 32.8 (q), 42.8 (d), 66.3 (d), 87.1 (s), 115.0 (d, J_{CCF} = 21.6 Hz), 115.6 (d, J_{CCF} = 21.6 Hz), 127.8 (d, J_{CCCF} = 8.4 Hz), 129.0 (d, J_{CCCF} = 8.4 Hz), 134.0 (s, J_{CCCCF} = 2.7 Hz), 138.7 (s, J_{CCCCF} = 2.7 Hz), 156.9 (s), 162.1 (s, J_{CF} = 249.2 Hz), 162.5 (s, J_{CF} = 248.6 Hz), 173.8 (s), minor; 14.1 (q), 26.5 (q), 30.0 (q), 40.2 (d), 67.0 (d), 86.3 (s), 114.7 (d, J_{CCF} = 21.6 Hz), 115.6 (d, J_{CCF} = 21.6 Hz), 127.3 (d, J_{CCCF} = 8.4 Hz), 129.3 (d, J_{CCCF} = 8.4 Hz), 133.5 (s, J_{CCCCF} = 2.7 Hz), 139.5 (s, J_{CCCCF} = 2.7 Hz), 156.4 (s), 162.0 (s, J_{CF} = 248.6 Hz), 162.5 (s, J_{CF} = 248.6 Hz), 172.5 (s); HRMS (ESI, ion trap) calcd for $C_{20}H_{21}F_2N_2O_3$ (M + H$^+$) 375.1520; found 375.1518.

*2-(5,5-Bis(4-methoxyphenyl)-3-methyl-2-oxooxazolidin-4-yl)-N-methylpropanamide (**12j**).* Colorless paste (54 mg, 54%, *erythro*:*threo* = 45:55 dr); R_f 0.2 (ethyl acetate); IR (ATR) 3321, 1740, 1647, 1609, 1578, 1541, 1510, 989, 899, 827, 789, 756, 727, 677, 669, 662 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 0.83 (d, 1.65H, J = 6.9 Hz), 1.10 (d, 1.35H, J = 6.9 Hz), 2.36−2.43 (m, 0.45H), 2.53 (d, 1.35H, J = 4.6 Hz), 2.62 (d, 1.65H, J = 5.2 Hz), 2.64−2.70 (m, 0.55H), 2.87 (s, 1.65H), 2.93 (s, 1.35H), 3.757 (s, 1.65H), 3.762 (s, 1.35H), 3.79 (s, 3H), 4.87 (d, 0.55H, J = 5.2 Hz), 4.91 (brs, 1H), 4.97 (d, 0.45H, J = 6.3 Hz), 5.45 (brs, 0.55H), 6.76−6.82 (m, 2H), 6.87−6.92 (m, 2H), 7.25−7.34 (m, 2H), 7.52−7.57 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 12.0 (q), 13.4 (q), 26.36 (q), 26.40 (q), 30.6 (q), 32.5 (q), 40.8 (d), 42.8 (d), 55.09 (q), 55.13 (q), 55.16 (q), 55.17 (q), 66.2 (d), 67.5 (d), 87.2 (s), 87.6 (s), 113.0 (d), 113.2 (d), 113.7 (d), 126.9 (d), 127.1 (d), 128.2 (d), 128.7 (d), 130.2 (s), 130.7 (s), 135.4 (s), 135.6 (s), 157.1 (s), 157.3 (s), 158.8 (s), 159.0 (s), 159.29 (s), 159.31 (s), 173.2 (s), 174.2 (s); HRMS (ESI, ion trap) calcd for $C_{22}H_{27}N_2O_5$ (M + H$^+$) 399.1920; found 399.1918.

(R)-2-Fluoro-N-methyl-2-((S*)-3-methyl-2-oxo-5,5-diphenyloxazolidin-4-yl)acetamide (**threo-12k**).* White solid (54 mg, 63%); R_f 0.6 (hexanes−ethyl acetate, 1:5); mp 204−205 °C; IR (ATR) 3358, 1753, 1680, 1545, 1493, 932, 849, 822, 793, 773, 760, 752, 700, 671 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 2.86 (d, 3H, J = 4.7 Hz), 2.87 (s, 3H), 4.59 (d, 1H, J_{HF} = 46.5 Hz), 5.17 (d, 1H, J_{HF} = 27.1 Hz), 6.48 (brs, 1H), 7.27−7.44 (m, 8H), 7.53−7.58 (m, 2H); ^{13}C NMR (125 MHz; $CDCl_3$) δ 25.9 (q), 30.2 (q), 64.8 (d, J_{CCF} = 16.5 Hz), 86.3 (s), 87.6 (d, J_{CF} = 198.2 Hz), 125.7 (d), 126.1 (d), 128.1 (d), 128.4 (d), 128.5 (d), 128.6 (d), 137.9 (s), 142.0 (s), 156.7 (s), 168.1 (s, J_{CF} = 19.2 Hz). Anal. Calcd for $C_{19}H_{19}FN_2O_3$: C, 66.66; H, 5.59; N, 8.18. Found: C, 66.68; H, 5.62; N, 8.11.

(R)-2-((S*)-5,5-Bis(4-fluorophenyl)-3-methyl-2-oxooxazolidin-4-yl)-2-fluoro-N-methylacetamide (**threo-12l**).* White solid (46 mg, 49%); R_f 0.35 (hexanes−ethyl acetate, 1:1); mp 221−222 °C; IR (ATR) 3387, 1763, 1719, 1670, 1636, 1603, 1549, 1508, 1474, 962, 955, 941, 928, 858, 847, 837, 822, 812, 804, 787, 756, 727, 685, 669, 656 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 2.876 (d, 3H, J = 4.4 Hz), 2.879 (s, 3H), 4.53 (d, 1H, J_{HF} = 46.3 Hz), 5.10 (d, 1H, J_{HF} = 26.9 Hz), 6.47 (brs, 1H), 7.02−7.12 (m, 4H), 7.34−7.40 (m, 2H), 7.48−7.54 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 26.1 (q), 30.3 (q), 64.8 (d, J_{CCF} = 16.8 Hz), 85.7 (s), 87.6 (d, J_{CF} = 197.9 Hz), 115.6 (d, J_{CCF} = 22.8 Hz), 115.8 (d, J_{CCF} = 21.6 Hz), 127.8 (d, J_{CCCF} = 8.4 Hz), 128.2 (d, J_{CCCF} = 8.4 Hz), 133.6 (s, J_{CCCCF} = 3.6 Hz), 137.6 (s, J_{CCCCF} = 3.6 Hz), 156.4 (s), 162.4 (s, J_{CF} = 249.5 Hz), 162.7 (s, J_{CF} = 249.5 Hz), 167.8 (s, J_{CCF} = 18.0 Hz). Anal. Calcd for $C_{19}H_{17}F_3N_2O_3$: C, 60.32; H, 4.53; N, 7.40. Found: C, 60.25; H, 4.52; N, 7.22.

(R)-2-((S*)-5,5-Bis(4-methoxyphenyl)-3-methyl-2-oxooxazolidin-4-yl)-2-fluoro-N-methylacetamide (**threo-12m**).* Colorless paste (82 mg, 82%); R_f 0.4 (hexanes−ethyl acetate, 1:5); IR (ATR) 3337, 1751, 1672, 1609, 1582, 1541, 1510, 991, 849, 829, 820, 800, 781, 758, 731, 692, 662 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 2.86 (d, 3H, J = 4.9 Hz), 2.87 (s, 3H), 3.78 (s, 3H), 3.80 (s, 3H), 4.56 (d, 1H, J_{HF} = 46.3 Hz), 5.07 (d, 1H, J_{HF} = 26.9 Hz), 6.47 (brs, 1H), 6.83−6.91 (m, 4H), 7.27−7.33 (m, 2H), 7.41−7.46 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 26.0 (q), 30.2 (q), 55.1 (q), 55.2 (q), 64.9 (d, J_{CCF} = 16.5 Hz), 86.3 (s), 87.8 (d, J_{CF} = 197.9 Hz), 113.6 (d), 113.8 (d), 127.2 (d), 127.6 (d), 130.2 (s), 134.2 (s), 156.9 (s), 159.2 (s), 159.5 (s), 168.2 (s, J_{CCF} = 19.2 Hz); HRMS (ESI, ion trap) calcd for $C_{21}H_{24}FN_2O_5$ (M + H$^+$) 403.1669; found 403.1666.

(R)-2-Fluoro-N-methyl-2-((S*)-3′-methyl-2′-oxo-10,11-dihydrospiro[dibenzo[a,d][7]annulene-5,5′-oxazolidin]-4′-yl)acetamide*

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*(**threo-12n**)*. White solid (76 mg, 82%); R_f 0.25 (hexanes−ethyl acetate, 1:1); mp 228−229 °C; IR (ATR) 3374, 1738, 1682, 1595, 1543, 1483, 793, 779, 760, 752, 741, 706, 665 cm^{-1}; ^1H NMR ($CDCl_3$) δ 2.78 (s, 3H), 2.85 (d, 3H, J = 5.0 Hz), 2.98−3.09 (m, 2H), 3.32−3.40 (m, 1H), 3.49−3.57 (m, 1H), 4.78 (d, 1H, J_{HF} = 46.3 Hz), 4.94 (d, 1H, J_{HF} = 25.2 Hz), 6.57 (brs, 1H), 7.15−7.19 (m, 1H), 7.19−7.30 (m, 5H), 7.62−7.66 (m, 1H), 7.84−7.88 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 25.9 (q), 30.2 (q), 31.8 (t), 32.9 (t), 68.7 (d, J_{CCF} = 16.8 Hz), 85.4 (s), 87.3 (d, J_{CF} = 198.8 Hz), 124.0 (d), 126.37 (d), 126.44 (d), 126.6 (d), 128.6 (d), 128.8 (d), 130.8 (d), 131.0 (d), 134.8 (s), 136.90 (s), 136.92 (s), 140.3 (s), 156.9 (s), 168.2 (s, J_{CCF} = 19.2 Hz). Anal. Calcd for $C_{21}H_{21}FN_2O_3$: C, 68.47; H, 5.75; N, 7.60. Found: C, 68.44; H, 5.76; N, 7.54.

*6-(1-Hydroxy-1,2,3,4-tetrahydronaphthalen-1-yl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (**erythro-13d**)*. Colorless paste (66 mg, 91%); R_f 0.35 (hexanes−ethyl acetate, 1:2); IR (ATR) 3406, 1701, 1643, 1483, 988, 970, 926, 901, 756, 729, 702, 677, 669 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.75−1.85 (m, 3H), 1.97−2.04 (m, 1H), 2.13−2.19 (m, 1H), 2.72 (s, 3H), 2.74−2.81 (m, 3H), 3.09 (s, 3H), 3.18 (dd, 1H, J = 1.0, 16.8 Hz), 3.77 (dd, 1H, J = 1.0, 8.0 Hz), 7.10−7.13 (m, 1H), 7.21−7.26 (m, 2H), 7.56−7.59 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 19.4 (t), 27.0 (q), 29.3 (t), 31.2 (t), 33.3 (t), 37.8 (q), 61.3 (d), 73.9 (s), 126.0 (d), 127.1 (d), 127.8 (d), 129.0 (d), 137.7 (s), 139.0 (s), 153.9 (s), 169.5 (s); HRMS (ESI, ion trap) calcd for $C_{16}H_{21}N_2O_3$ (M + H^+) 289.1552; found 289.1551.

*6-(1-Hydroxy-1,2,3,4-tetrahydronaphthalen-1-yl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (**threo-13d**)*. Colorless paste (47 mg, 65%); R_f 0.2 (hexanes−ethyl acetate, 1:2); IR (ATR) 3420, 1701, 1647, 1485, 984, 972, 924, 756, 727, 679, 669 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.74−1.96 (m, 4H), 2.13−2.19 (m, 1H), 2.63 (s, 1H), 2.67 (s, 1H), 2.74−2.86 (m, 3H), 3.20 (s, 3H), 3.75 (d, 1H, J = 7.7 Hz), 7.09−7.13 (m, 1H), 7.21−7.26 (m, 2H), 7.37−7.41 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 19.4 (t), 27.2 (q), 29.0 (t), 32.3 (t), 33.6 (t), 38.0 (q), 60.8 (d), 75.7 (s), 126.4 (d), 126.7 (d), 128.0 (d), 129.1 (d), 136.9 (s), 138.4 (s), 153.9 (s), 169.6 (s); HRMS (ESI, ion trap) calcd for $C_{16}H_{21}N_2O_3$ (M + H^+) 289.1552; found 289.1549.

(R)-6-((S*)-1-Hydroxy-2,3-dihydro-1H-inden-1-yl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (**erythro-13e**)*. White solid (53 mg, 78%); R_f 0.3 (hexanes−ethyl acetate, 1:2); mp 173−175 °C; IR (ATR) 3424, 1686, 1647, 1508, 1474, 988, 970, 945, 916, 907, 810, 789, 762, 752, 727, 683, 669, 658 cm^{-1}; ^1H NMR ($CDCl_3$) δ 2.03−2.11 (m, 1H), 2.23 (brs, 1H), 2.40−2.46 (m, 1H), 2.70 (s, 3H), 2.76−2.80 (m, 2H), 2.90−3.00 (m, 2H), 3.15 (s, 3H), 3.72 (dd, 1H, J = 3.9, 5.7 Hz), 7.21−7.30 (m, 4H); ^{13}C NMR ($CDCl_3$) δ 26.6 (q), 29.7 (t), 32.0 (t), 38.5 (q), 39.5 (t), 61.2 (d), 86.3 (s), 124.0 (d), 125.3 (d), 126.8 (d), 129.2 (d), 142.8 (s), 143.3 (s), 153.6 (s), 168.2 (s). Anal. Calcd for $C_{15}H_{18}N_2O_3$: C, 65.68; H, 6.61; N, 10.21. Found: C, 65.64; H, 6.59; N, 10.10.

(R)-6-((R*)-1-Hydroxy-2,3-dihydro-1H-inden-1-yl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (**threo-13e**)*. Colorless paste (54 mg, 79%); R_f 0.45 (hexanes−ethyl acetate, 1:5); IR (ATR) 3358, 1701, 1647, 970, 945, 916, 907, 808, 779, 760, 729, 712, 683 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.97−2.05 (m, 1H), 2.29 (brs, 1H), 2.35−2.41 (m, 1H), 2.69 (s, 3H), 2.79−2.87 (m, 2H), 2.91−2.99 (m, 1H), 3.09 (s, 3H), 3.44 (dd, 1H, J = 1.9, 6.4 Hz), 7.20−7.32 (m, 4H); ^{13}C NMR ($CDCl_3$) δ 27.1 (q), 29.0 (t), 32.6 (t), 38.4 (q), 39.2 (t), 59.8 (d), 86.4 (s), 124.1 (d), 125.0 (d), 127.1 (d), 129.0 (d), 142.3 (s), 143.9 (s), 153.8 (s), 169.8 (s); HRMS (ESI, ion trap) calcd for $C_{15}H_{19}N_2O_3$ (M + H^+) 275.1396; found 275.1395.

(5R,6S*)-6-((R*)-1-Hydroxy-1,2,3,4-tetrahydronaphthalen-1-yl)-1,3,5-trimethyldihydropyrimidine-2,4(1H,3H)-dione (**trans-erythro-13f**)*. White solid (31 mg, 41%); R_f 0.3 (hexanes−ethyl acetate, 1:1); mp 205−206 °C; IR (ATR) 3389, 1705, 1651, 1522, 1487, 989, 962, 955, 914, 901, 874, 851, 837, 795, 758, 745, 718, 669, 658 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.30 (d, 3H, J = 7.2 Hz), 1.73−1.83 (m, 3H), 1.95−2.02 (m, 1H), 2.73−2.79 (m, 2H), 2.74 (s, 3H), 3.10 (s, 3H), 3.26 (q, 1H, J = 7.2 Hz), 3.40 (s, 1H), 7.10−7.13 (m, 1H), 7.21−7.28 (m, 2H), 7.56−7.59 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 18.4 (q), 19.5 (t), 27.2 (q), 29.4 (t), 33.7 (t), 35.5 (d), 38.6 (q), 68.9 (d), 74.0 (s), 126.2 (d), 127.2 (d), 128.0 (d), 129.2 (d), 137.8 (s), 139.1 (s), 153.6 (s), 173.5 (s). Anal. Calcd for $C_{17}H_{22}N_2O_3$: C, 67.53; H, 7.33; N, 9.26. Found: C, 67.54; H, 7.33; N, 9.21.

(5R,6S*)-6-((S*)-1-Hydroxy-1,2,3,4-tetrahydronaphthalen-1-yl)-1,3,5-trimethyldihydropyrimidine-2,4(1H,3H)-dione (**trans-threo-13f**)*. Colorless paste (53 mg, 70%); R_f 0.25 (hexanes−ethyl acetate, 1:1); IR (ATR) 1697, 1647, 1485, 978, 951, 916, 878, 858, 799, 758, 727, 669 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.26 (d, 3H, J = 7.5 Hz), 1.74−1.93 (m, 4H), 2.19 (brs, 1H), 2.67−2.86 (m, 3H), 2.70 (s, 3H), 3.18 (s, 3H), 3.41 (s, 1H), 7.09−7.13 (m, 1H), 7.20−7.26 (m, 2H), 7.36−7.40 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 18.2 (q), 19.4 (t), 27.3 (q), 29.1 (t), 33.4 (t), 36.9 (d), 38.8 (q), 68.2 (d), 75.5 (s), 126.1 (d), 126.8 (d), 128.0 (d), 129.2 (d), 137.0 (s), 138.5 (s), 153.5 (s), 173.4 (s); HRMS (ESI, ion trap) calcd for $C_{17}H_{23}N_2O_3$ (M + H^+) 303.1709; found 303.1707.

(5R,6R*)-6-((S*)-1-Hydroxy-1,2,3,4-tetrahydronaphthalen-1-yl)-1,3,5-trimethyldihydropyrimidine-2,4(1H,3H)-dione (**cis-erythro-13f**)*. Colorless paste (60 mg, 80%); R_f 0.35 (hexanes−ethyl acetate, 1:1); IR (ATR) 3444, 1705, 1655, 1477, 989, 897, 874, 829, 812, 789, 772, 754, 737, 714, 692, 669, 652 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.51 (d, 3H, J = 7.5 Hz), 1.61−1.69 (m, 2H), 1.74−1.81 (m, 1H), 1.92−1.99 (m, 1H), 2.34 (s, 3H), 2.62−2.73 (m, 2H), 3.07−3.14 (m, 1H), 3.15 (s, 3H), 3.90 (d, 1H, J = 6.3 Hz), 7.07−7.10 (m, 1H), 7.17−7.28 (m, 2H), 7.61−7.64 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 13.3 (q), 19.4 (t), 27.5 (q), 29.7 (t), 33.4 (t), 37.9 (q), 39.5 (d), 66.6 (d), 74.9 (s), 126.1 (d), 127.6 (d), 128.0 (d), 129.3 (d), 137.9 (s), 140.1 (s), 154.0 (s), 172.5 (s); HRMS (ESI, ion trap) calcd for $C_{17}H_{23}N_2O_3$ (M + H^+) 303.1709; found 303.1706.

(5R,6R*)-6-((R*)-1-Hydroxy-1,2,3,4-tetrahydronaphthalen-1-yl)-1,3,5-trimethyldihydropyrimidine-2,4(1H,3H)-dione (**cis-threo-13f**)*. Colorless paste (56 mg, 74%); R_f 0.45 (hexanes−ethyl acetate, 1:2); IR (ATR) 3422, 1701, 1647, 1483, 976, 935, 916, 897, 841, 772, 752, 727, 690, 669 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.05 (d, 3H, J = 6.2 Hz), 1.72−1.99 (m, 5H), 2.70 (brs, 3H), 2.73−2.85 (m, 2H), 2.96−3.03 (m, 1H), 3.23 (s, 3H), 3.90 (d, 1H, J = 6.0 Hz), 7.07−7.11 (m, 1H), 7.19−7.26 (m, 2H), 7.35−7.38 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 12.9 (q), 19.2 (t), 27.5 (q), 29.3 (t), 34.6 (t), 38.1 (q), 39.0 (d), 65.3 (d), 75.6 (s), 126.4 (d), 126.6 (d), 127.9 (d), 129.1 (d), 136.8 (s), 139.4 (s), 153.9 (s), 172.7 (s); HRMS (ESI, ion trap) calcd for $C_{17}H_{23}N_2O_3$ (M + H^+) 303.1709; found 303.1707.

(5R,6S*)-6-((R*)-1-Hydroxy-2,3-dihydro-1H-inden-1-yl)-1,3,5-trimethyldihydropyrimidine-2,4(1H,3H)-dione (**trans-erythro-13g**)*. Colorless paste (45 mg, 63%); R_f 0.3 (hexanes−ethyl acetate, 1:2); IR (ATR) 3383, 1699, 1647, 955, 920, 905, 793, 772, 756, 729, 667 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.26 (d, 3H, J = 7.2 Hz), 2.03−2.10 (m, 1H), 2.36−2.43 (m, 1H), 2.70 (s, 3H), 2.83−2.98 (m, 4H), 3.13 (s, 3H), 3.36 (d, 1H, J = 1.1 Hz), 7.19−7.28 (m, 4H); ^{13}C NMR ($CDCl_3$) δ 18.0 (q), 26.8 (q), 29.8 (t), 36.2 (d), 39.2 (q), 39.7 (t), 68.5 (d), 86.4 (s), 124.0 (d), 125.4 (d), 126.9 (d), 129.4 (d), 142.9 (s), 143.4 (s), 153.2 (s), 172.3 (s); HRMS (ESI, ion trap) calcd for $C_{16}H_{21}N_2O_3$ (M + H^+) 289.1552; found 289.1551.

(5R,6S*)-6-((S*)-1-Hydroxy-2,3-dihydro-1H-inden-1-yl)-1,3,5-trimethyldihydropyrimidine-2,4(1H,3H)-dione (**trans-threo-13g**)*. White solid (42 mg, 58%); R_f 0.4 (hexanes−ethyl acetate, 1:2); mp 157−158 °C; IR (ATR) 3327, 1694, 1645, 1510, 1472, 993, 966, 918, 903, 876, 851, 820, 795, 754, 723, 669, 652 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.29 (d, 3H, J = 7.4 Hz), 1.96−2.04 (m, 1H), 2.20 (brs, 1H), 2.34−2.40 (m, 1H), 2.71 (s, 3H), 2.79−2.87 (m, 1H), 2.89−2.98 (m, 2H), 3.07 (s, 1H), 3.08 (s, 3H), 7.22−7.31 (m, 4H); ^{13}C NMR ($CDCl_3$) δ 17.7 (q), 27.2 (q), 29.1 (t), 37.3 (d), 39.2 (q), 39.4 (t), 67.1 (d), 86.3 (s), 124.0 (d), 125.1 (d), 127.2 (d), 129.1 (d), 142.4 (s), 143.9 (s), 153.4 (s), 173.6 (s). Anal. Calcd for $C_{16}H_{20}N_2O_3$: C, 66.65; H, 6.99; N, 9.72. Found: C, 66.68; H, 7.00; N, 9.62.

(5R,6R*)-6-((S*)-1-Hydroxy-2,3-dihydro-1H-inden-1-yl)-1,3,5-trimethyldihydropyrimidine-2,4(1H,3H)-dione (**cis-erythro-13g**)*. Colorless paste (49 mg, 68%); R_f 0.25 (hexanes−ethyl acetate, 1:1); IR (ATR) 3422, 1701, 1647, 1479, 988, 955, 916, 895, 841, 808, 783, 756, 729, 708, 669 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.52 (d, 3H, J = 7.4 Hz), 1.87−1.96 (m, 1H), 2.01 (brs, 1H), 2.37−2.43 (m, 1H), 2.39 (s, 3H), 2.57−2.65 (m, 1H), 2.87 (dd, 1H, J = 8.8, 16.5 Hz), 3.06−3.13 (m, 1H), 3.09 (s, 3H), 3.75 (d, 1H, J = 6.1 Hz), 7.21−7.31 (m, 3H), 7.38−7.41 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 12.8 (q), 27.5 (q), 29.9 (t), 38.0

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(t), 38.2 (q), 39.7 (d), 65.8 (d), 86.0 (s), 123.7 (d), 125.4 (d), 126.7 (d), 129.1 (d), 142.7 (s), 145.9 (s), 153.5 (s), 172.6 (s); HRMS (ESI, ion trap) calcd for $C_{16}H_{21}N_2O_3$ (M + H$^+$) 289.1552; found 289.1551

(5R,6R*)-6-((R*)-1-Hydroxy-2,3-dihydro-1H-inden-1-yl)-1,3,5-trimethyldihydropyrimidine-2,4(1H,3H)-dione* (***cis-threo-13g***). White solid (44 mg, 61%); R_f 0.2 (hexanes−ethyl acetate, 1:1); mp 107−109 °C; IR (ATR) 3406, 3221, 1690, 1645, 1489, 1474, 991, 962, 897, 831, 772, 754, 726, 669 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 1.49 (d, 3H, J = 7.0 Hz), 1.89 (brs, 1H), 1.91−1.98 (m, 1H), 2.26 (s, 3H), 2.55−2.62 (m, 1H), 2.82−2.91 (m, 1H), 2.94−3.05 (m, 2H), 3.21 (s, 3H), 3.37 (d, 1H, J = 5.6 Hz), 7.14−7.18 (m, 1H), 7.22−7.32 (m, 3H); ^{13}C NMR ($CDCl_3$) δ 13.3 (q), 27.5 (q), 29.2 (t), 37.1 (q), 39.1 (d), 41.6 (t), 63.9 (d), 86.1 (s), 124.4 (d), 124.7 (d), 127.4 (d), 128.8 (d), 142.1 (s), 145.1 (s), 153.9 (s), 172.8 (s); HRMS (ESI, ion trap) calcd for $C_{16}H_{21}N_2O_3$ (M + H$^+$) 289.1552; found 289.1551.

(5R,6S*)-5-Fluoro-6-((S*)-1-hydroxy-1,2,3,4-tetrahydronaphthalen-1-yl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione* (***cis-erythro-13h***). White solid (41 mg, 54%); R_f 0.3 (hexanes−ethyl acetate, 1:1); mp 190−192 °C; IR (ATR) 3352, 1713, 1645, 1514, 1487, 991, 947, 920, 878, 841, 818, 797, 773, 756, 718, 669 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 1.74−1.81 (m, 2H), 1.87−1.93 (m, 1H), 2.10−2.17 (m, 1H), 2.42 (d, 1H, J = 7.5 Hz), 2.71−2.77 (m, 2H), 2.80 (s, 3H), 3.70 (s, 3H), 4.22 (d, 1H, J = 6.9 Hz), 5.38 (dd, 1H, J = 6.9 Hz, J_{HF} = 47.0 Hz), 7.09−7.13 (m, 1H), 7.21−7.27 (m, 2H), 7.54−7.59 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 19.2 (t), 27.4 (q), 29.3 (t), 35.1 (t), 38.7 (q), 64.6 (d, J_{CCF} = 18.0 Hz), 74.5 (s), 83.9 (d, J_{CF} = 195.5 Hz), 126.0 (d), 127.6 (d), 128.1 (d), 129.0 (d), 137.9 (s), 138.0 (s), 152.6 (s), 165.9 (s, J_{CCF} = 20.7 Hz). Anal. Calcd for $C_{16}H_{19}FN_2O_3$: C, 62.73; H, 6.25; N, 9.14. Found: C, 62.76; H, 6.24; N, 9.09.

(5R,6S*)-5-Fluoro-6-((R*)-1-hydroxy-1,2,3,4-tetrahydronaphthalen-1-yl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione* (***cis-threo-13h***). Colorless paste (18 mg, 23%); R_f 0.3 (hexanes−ethyl acetate, 1:1); IR (ATR) 3480, 1713, 1667, 1506, 1483, 916, 866, 849, 785, 750, 721, 669 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 1.69−1.77 (m, 1H), 1.84−1.95 (m, 1H), 2.03−2.10 (m, 2H), 2.15 (s, 3H), 2.53−2.59 (m, 1H), 2.85−2.92 (m, 2H), 3.28 (s, 3H), 3.93 (d, 1H, J = 6.6 Hz), 5.35 (dd, 1H, J = 6.6 Hz, J_{HF} = 47.0 Hz), 7.08−7.12 (m, 1H), 7.21−7.28 (m, 2H), 7.30−7.34 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 19.5 (t), 27.5 (q), 27.9 (t), 35.4 (t), 37.0 (q), 62.5 (d, J_{CCF} = 19.2 Hz), 75.1 (s, J_{CCCF} = 2.4 Hz), 84.1 (d, J_{CF} = 195.5 Hz), 126.6 (d), 126.7 (s), 130.1 (d), 128.1 (d), 128.7 (d), 136.1 (s), 138.5 (s), 152.8 (s), 167.3 (s, J_{CCF} = 20.4 Hz); HRMS (ESI, ion trap) calcd for $C_{16}H_{20}FN_2O_3$ (M + H$^+$) 307.1458; found 307.1456.

(5R,6S*)-5-Fluoro-6-((S*)-1-hydroxy-2,3-dihydro-1H-inden-1-yl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione* (***cis-erythro-13i***). Colorless paste (51 mg, 70%); R_f 0.25 (hexanes−ethyl acetate, 1:1); IR (ATR) 3323, 1742, 1676, 1543, 1508, 1474, 924, 845, 826, 785, 777, 764, 729, 710, 683, 669 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 1.97−2.06 (m, 1H), 2.29 (brs, 1H), 2.64 (s, 3H), 2.63−2.69 (m, 1H), 2.84−2.91 (m, 1H), 2.99−3.07 (m, 1H), 3.28 (s, 3H), 4.13 (d, 1H, J = 7.0 Hz), 5.26 (dd, 1H, J = 7.0 Hz, J_{HF} = 47.1 Hz), 7.20−7.31 (m, 4H); ^{13}C NMR ($CDCl_3$) δ 27.0 (q), 29.4 (t), 39.2 (q), 40.7 (t), 63.8 (d, J_{CCF} = 20.7 Hz), 83.5 (d, J_{CF} = 196.7 Hz), 86.3 (s, J_{CCCF} = 2.4 Hz), 124.2 (d), 125.5 (d), 126.7 (d), 129.5 (d), 142.5 (s), 144.1 (s), 152.5 (s), 164.9 (s, J_{CCF} = 21.6 Hz); HRMS (ESI, ion trap) calcd for $C_{15}H_{18}FN_2O_3$ (M + H$^+$) 293.1301; found 293.1300.

(5R,6S*)-5-Fluoro-6-((R*)-1-hydroxy-2,3-dihydro-1H-inden-1-yl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione* (***cis-threo-13i***). Colorless paste (47 mg, 64%); R_f 0.3 (hexanes−ethyl acetate, 1:1); IR (ATR) 3401, 1719, 1655, 1474, 914, 864, 847, 754, 725 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 1.96−2.05 (m, 1H), 2.26 (brs, 1H), 2.30 (s, 3H), 2.75−2.91 (m, 2H), 2.95−3.02 (m, 1H), 3.23 (s, 3H), 3.66 (d, 1H, J = 6.9 Hz), 5.35 (dd, 1H, J = 6.9 Hz, J_{HF} = 47.0 Hz), 7.22−7.34 (m, 4H); ^{13}C NMR ($CDCl_3$) δ 27.5 (q), 29.1 (t), 37.8 (q), 40.6 (t), 61.6 (d, J_{CCF} = 20.4 Hz), 83.2 (d, J_{CF} = 195.5 Hz), 85.1 (s), 124.7 (d), 124.9 (d), 127.4 (d), 129.1 (d), 142.3 (s), 143.9 (s), 152.8 (s), 167.2 (s, J_{CCF} = 20.4 Hz); HRMS (ESI, ion trap) calcd for $C_{15}H_{18}FN_2O_3$ (M + H$^+$) 293.1301; found 293.1299.

N-Methyl-2-((1R,4′S*)-3′-methyl-2′-oxo-3,4-dihydro-2H-spiro[naphthalene-1,5′-oxazolidin]-4′-yl)propanamide* (***erythro-14f***). Colorless paste (38 mg, 50%, 68:32 dr); R_f 0.25 (ethyl acetate); IR (ATR) 3350, 1734, 1665, 1558, 982, 922, 903, 880, 835, 810, 772, 727, 667, 652 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 0.46 (d, 0.96H, J = 6.6 Hz), 1.17 (d, 2.04H, J = 7.0 Hz), 1.89−2.20 (m, 6H), 2.25−2.32 (m, 0.68H), 2.28 (d, 2.04H, J = 5.0 Hz), 2.40−2.47 (m, 0.32H), 2.72 (d, 0.96H, J = 4.9 Hz), 2.76−2.93 (m, 2H), 2.94 (s, 0.96H), 3.09 (s, 2.04H), 3.71 (d, 0.32H, J = 9.9 Hz), 4.03 (d, 0.68H, J = 8.7 Hz), 4.25 (brs, 0.68H), 6.22 (brs, 0.32H), 7.09−7.14 (m, 1H), 7.18−7.28 (m, 1H), 7.51−7.55 (m, 0.68H), 7.57−7.60 (m, 0.32H); ^{13}C NMR ($CDCl_3$) δ major: 15.9 (q), 17.4 (t), 26.3 (q), 27.6 (t), 34.2 (q), 34.8 (t), 44.2 (d), 67.4 (d), 83.4 (s), 125.2 (d), 126.9 (d), 128.3 (d), 129.0 (d), 133.6 (s), 139.0 (s), 158.3 (s), 173.7 (s), minor: 14.3 (q), 18.5 (t), 26.3 (q), 28.3 (t), 31.9 (q), 35.9 (t), 42.5 (d), 69.4 (d), 82.4 (s), 125.7 (d), 127.2 (d), 128.5 (d), 128.9 (d), 133.4 (s), 137.9 (s), 158.1 (s), 174.2 (s); HRMS (ESI, ion trap) calcd for $C_{17}H_{23}N_2O_3$ (M + H$^+$) 303.1709; found 303.1706.

N-Methyl-2-((1R,4′R*)-3′-methyl-2′-oxo-2,3-dihydrospiro[indene-1,5′-oxazolidin]-4′-yl)propanamide* (***erythro-14g***). Colorless paste (11 mg, 15%, 55:45 dr); R_f 0.35 (ethyl acetate); IR (ATR) 3321, 1732, 1647, 1545, 1476, 908, 762, 727, 683, 669, 652 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 0.55 (d, 1.65H, J = 6.9 Hz), 1.23 (d, 1.35H, J = 6.9 Hz), 2.25−2.31 (m, 0.45H), 2.33 (d, 1.35H, J = 4.6 Hz), 2.35−2.55 (m, 2.55H), 2.72 (d, 1.65H, J = 4.6 Hz), 2.84−3.11 (m, 2H), 2.93 (s, 1.65H), 3.08 (s, 1.35H), 3.95 (d, 0.55H, J = 9.2 Hz), 4.15 (d, 0.45H, J = 7.5 Hz), 4.64 (brs, 0.45H), 6.10 (brs, 0.55H), 7.21−7.35 (m, 3H), 7.36−7.39 (m, 0.45H), 7.43−7.47 (m, 0.55H); ^{13}C NMR ($CDCl_3$) δ 14.3 (q), 15.4 (q), 26.2 (q), 26.4 (q), 28.4 (t), 28.6 (t), 31.3 (q), 33.4 (q), 40.1 (t), 41.9 (t), 42.4 (d), 43.8 (d), 66.8 (d), 67.7 (d), 91.2 (s), 91.6 (s), 124.4 (d), 124.6 (d), 125.17 (d), 125.24 (d), 126.3 (d), 126.8 (d), 129.4 (d), 129.7 (d), 139.0 (s), 139.1 (s), 144.0 (s), 144.7 (s), 158.3 (s), 158.6 (s), 173.6 (s), 174.1 (s); HRMS (ESI, ion trap) calcd for $C_{16}H_{21}N_2O_3$ (M + H$^+$) 289.1552; found 289.1551.

(2R)-2-Fluoro-N-methyl-2-((4′S*)-3′-methyl-2′-oxo-3,4-dihydro-2H-spiro[naphthalene-1,5′-oxazolidin]-4′-yl)acetamide* (***14h***). Colorless paste (54 mg, 70%, 60:40 dr); R_f 0.4 (hexanes−ethyl acetate, 1:5); ^{1}H NMR ($CDCl_3$) δ 1.86−2.18 (m, 4H), 2.21−2.27 (m, 0.6H), 2.33−2.41 (m, 0.4H), 2.78−2.93 (m, 6.8H), 2.96 (s, 1.2H), 4.27 (d, 0.6H, J_{HF} = 46.0 Hz), 4.43 (d, 0.6H, J_{HF} = 25.9 Hz), 4.59 (d, 0.6H, J_{HF} = 28.5 Hz), 5.16 (d, 0.4H, J_{HF} = 46.8 Hz), 6.68 (brs, 0.6H), 6.95 (brs, 0.4H), 7.11−7.16 (m, 1H), 7.20−7.32 (m, 2.4H), 7.54−7.59 (m, 0.6H); ^{13}C NMR ($CDCl_3$) δ major: 18.2 (t), 25.8 (q), 27.8 (t), 30.1 (q), 34.6 (t), 66.0 (d, J_{CCF} = 17.4 Hz), 82.3 (s), 87.4 (d, J_{CF} = 197.3 Hz), 125.8 (d), 126.8 (d), 128.7 (d), 128.8 (d), 132.4 (s), 137.7 (s), 157.6 (s), 168.1 (s, J_{CCF} = 20.4 Hz), minor: 19.2 (t), 26.0 (q), 28.9 (t), 30.0 (q), 30.4 (t), 66.3 (d, J_{CCF} = 16.8 Hz), 79.7 (s), 87.3 (d, J_{CF} = 199.1 Hz), 124.7 (d), 127.0 (d), 128.8 (d), 129.3 (d), 136.7 (s), 137.2 (s), 157.4 (s), 167.7 (s, J_{CCF} = 19.8 Hz); HRMS (ESI, ion trap) calcd for $C_{16}H_{20}FN_2O_3$ (M + H$^+$) 307.1458; found 307.1456.

2-Fluoro-N-methyl-2-((1R,4′S*)-3′-methyl-2′-oxo-2,3-dihydrospiro[indene-1,5′-oxazolidin]-4′-yl)acetamide* (***erythro-14i***). Colorless paste (61 mg, 83%); R_f 0.3 (hexanes−ethyl acetate, 1:2); IR (ATR) 3325, 1740, 1670, 1543, 1476, 987, 847, 826, 791, 760, 727, 706, 683, 669, 658 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 2.38−2.45 (m, 1H), 2.50−2.56 (m, 1H), 2.85 (d, 3H, J = 5.0 Hz), 2.93 (s, 3H), 2.95−3.00 (m, 2H), 4.31 (d, 1H, J_{HF} = 28.9 Hz), 4.40 (d, 1H, J_{HF} = 46.1 Hz), 6.56 (brs, 1H), 7.25−7.31 (m, 2H), 7.32−7.37 (m, 1H), 7.49−7.52 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 26.0 (q), 28.3 (t), 30.2 (q), 41.1 (t), 65.0 (d, J_{CCF} = 18.0 Hz), 88.1 (d, J_{CF} = 197.3 Hz), 90.7 (s), 124.8 (d), 125.2 (d), 127.1 (d), 129.9 (d), 137.5 (s), 143.7 (s), 158.0 (s), 167.9 (s, J_{CCF} = 19.2 Hz); HRMS (ESI, ion trap) calcd for $C_{15}H_{18}FN_2O_3$ (M + H$^+$) 293.1301; found 293.1300.

2-Fluoro-N-methyl-2-((1R,4′R*)-3′-methyl-2′-oxo-2,3-dihydrospiro[indene-1,5′-oxazolidin]-4′-yl)acetamide* (***threo-14i***). White solid (50 mg, 68%); R_f 0.3 (hexanes−ethyl acetate, 1:2); mp 198−200 °C; IR (ATR) 3366, 1749, 1732, 1668, 1558, 1541, 976, 964, 922, 847, 820, 758, 723, 700, 679, 669 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 2.37−2.45 (m, 1H), 2.56−2.63 (m, 1H), 2.93 (d, 3H, J = 5.0 Hz), 2.94−2.98 (m, 1H), 2.99 (s, 3H), 3.16−3.24 (m, 1H), 4.60 (d, 1H, J_{HF} = 30.3 Hz), 5.11 (d, 1H, J_{HF} = 47.3 Hz), 6.70 (brs, 1H), 7.24−7.37 (m, 4H); ^{13}C NMR ($CDCl_3$) δ 26.0 (q), 29.6 (t), 30.2 (q), 33.7 (t), 64.3 (d, J_{CCF} = 18.0 Hz), 88.2 (d, J_{CF} = 199.1 Hz), 89.5 (s), 121.8 (d), 125.1 (d), 127.5 (d), 129.9 (d), 142.3 (s), 143.3 (s), 157.2 (s), 167.5 (s, J_{CCF} = 19.2 Hz);

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HRMS (ESI, ion trap) calcd for $C_{15}H_{18}FN_2O_3$ (M + H^+) 293.1301; found 293.1299.

1,3-Dimethyl-1-((3R,4R*)-4-methyl-5-oxo-2,2-diphenyltetrahydrofuran-3-yl)urea (**trans-15h**).* Colorless paste (41 mg, 49%); R_f 0.2 (hexanes−ethyl acetate, 1:1); IR (ATR) 3366, 1771, 1630, 1533, 1489, 986, 908, 764, 729, 700, 664 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.12 (d, 3H, J = 7.5 Hz), 2.14 (s, 3H), 2.69−2.75 (m, 1H), 2.83 (d, 3H, J = 4.6 Hz), 4.25 (brs, 1H), 6.09 (brs, 1H), 7.19−7.30 (m, 4H), 7.33−7.38 (m, 2H), 7.45−7.50 (m, 2H), 7.75−7.79 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 15.3 (q), 27.7 (q), 29.5 (q), 39.5 (d), 64.2 (d), 91.3 (s), 125.2 (d), 125.5 (d), 127.4 (d), 127.9 (d), 128.0 (d), 128.6 (d), 140.2 (s), 143.8 (s), 158.8 (s), 177.6 (s); HRMS (ESI, ion trap) calcd for $C_{20}H_{22}N_2O_3$ (M + H^+) 339.1709; found 339.1707.

1-((3R,4R*)-2,2-Bis(4-fluorophenyl)-4-methyl-5-oxotetrahydrofuran-3-yl)-1,3-dimethylurea (**trans-15i**).* White solid (22 mg, 24%); R_f 0.3 (hexanes−ethyl acetate, 1:2); mp 238−240 °C; IR (ATR) 3306, 1771, 1626, 1601, 1549, 1506, 1489, 989, 966, 951, 935, 868, 833, 808, 768, 727, 692, 679, 669 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.14 (d, 3H, J = 7.7 Hz), 2.17 (s, 3H), 2.69−2.76 (m, 1H), 2.84 (d, 3H, J = 4.6 Hz), 4.29 (brs, 1H), 6.01 (brs, 1H), 6.94−7.00 (m, 2H), 7.01−7.07 (m, 2H), 7.39−7.44 (m, 2H), 7.72−7.77 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 15.5 (q), 27.8 (q), 29.7 (q), 39.4 (d), 64.4 (d), 90.7 (s), 115.1 (d, J_{CCF} = 21.6 Hz), 115.7 (d, J_{CCF} = 21.6 Hz), 127.1 (d, J_{CCCF} = 8.4 Hz), 127.6 (d, J_{CCCF} = 7.8 Hz), 136.1 (s, J_{CCCCF} = 3.6 Hz), 139.8 (s, J_{CCCCF} = 3.0 Hz), 158.8 (s), 162.0 (s, J_{CF} = 247.7 Hz), 162.3 (s, J_{CF} = 247.4 Hz), 177.1 (s). Anal. Calcd for $C_{20}H_{20}F_2N_2O_3$: C, 64.16; H, 5.38; N, 7.48. Found: C, 64.09; H, 5.40; N, 7.38.

1-((3R,4R*)-2,2-Bis(4-methoxyphenyl)-4-methyl-5-oxotetrahydrofuran-3-yl)-1,3-dimethylurea (**trans-15j**).* White solid (15 mg, 15%); R_f 0.25 (hexanes−ethyl acetate, 1:2); mp 232−234 °C; IR (ATR) 3345, 1773, 1622, 1609, 1549, 1506, 1485, 989, 941, 926, 816, 795, 768, 729, 677, 667 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.15 (d, 3H, J = 7.5 Hz), 2.15 (s, 3H), 2.67−2.74 (m, 1H), 2.83 (d, 3H, J = 4.6 Hz), 3.76 (s, 3H), 3.77 (s, 3H), 4.23−4.27 (m, 1H), 5.98 (brs, 1H), 6.77−6.82 (m, 2H), 6.84−6.89 (m, 2H), 7.30−7.34 (m, 2H), 7.62−7.67 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 15.2 (q), 27.8 (q), 29.7 (q), 39.3 (d), 55.17 (q), 55.20 (q), 64.0 (d), 91.1 (s), 113.4 (d), 113.9 (d), 126.6 (d), 127.0 (d), 132.8 (s), 136.2 (s), 158.8 (s), 158.9 (s), 159.0 (s), 177.7 (s). Anal. Calcd for $C_{22}H_{26}N_2O_5$: C, 66.32; H, 6.58; N, 7.03. Found: C, 66.35; H, 6.57; N, 6.99.

Methyl (R)-2-((R*)-3-Methyl-2-(methylimino)-5,5-diphenyloxazolidin-4-yl)propanoate (**16h**).* Colorless paste (79 mg, 90%); R_f 0.2 (ethyl acetate−ethanol, 10:1); IR (ATR) 1701, 1522, 1491, 964, 939, 883, 808, 760, 750, 733, 698, 664 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.01 (d, 3H, J = 7.0 Hz), 2.59−2.66 (m, 1H), 2.96 (s, 3H), 3.11 (s, 3H), 3.53 (s, 3H), 4.97 (d, 1H, J = 4.7 Hz), 7.25−7.42 (m, 8H), 7.54−7.59 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 11.6 (q), 32.9 (q), 33.1 (q), 41.4 (d), 52.1 (q), 67.4 (d), 89.6 (s), 125.6 (d), 126.6 (d), 128.0 (d), 128.2 (d), 128.3 (d), 128.7 (d), 138.4 (s), 143.1 (s), 154.5 (s), 175.0 (s); HRMS (ESI, ion trap) calcd for $C_{21}H_{25}N_2O_3$ (M + H^+) 353.1865; found 353.1862.

Ethyl (R)-2-((R*)-3-Methyl-2-(methylimino)-5,5-diphenyloxazolidin-4-yl)propanoate (**16h′**).* Colorless paste (82%, 90%); R_f 0.25 (ethyl acetate−ethanol, 5:1); IR (ATR) 1701, 1528, 966, 922, 860, 760, 752, 727, 698, 665 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.01 (d, 3H, J = 6.9 Hz), 1.17 (t, 3H, J = 7.2 Hz), 2.58−2.65 (m, 1H), 3.06 (brs, 3H), 3.13 (s, 3H), 3.86−3.94 (m, 1H), 4.00−4.08 (m, 1H), 5.03 (brs, 1H), 7.28−7.43 (m, 8H), 7.53−7.58 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 11.5 (q), 13.8 (q), 31.7 (q), 33.6 (q), 41.2 (d), 61.2 (t), 67.7 (d), 125.4 (d), 126.5 (d), 128.2 (d), 128.3 (d), 128.7 (d), 128.8 (d), 137.5 (s), 142.2 (s), 155.7 (s), 174.0 (s); HRMS (ESI, ion trap) calcd for $C_{22}H_{26}N_2O_3$ (M + H^+) 367.2022; found 367.2019.

Methyl (R)-2-((R*)-5,5-Bis(4-fluorophenyl)-3-methyl-2-(methylimino)oxazolidin-4-yl)propanoate (**16i**).* Colorless paste (87 mg, 90%); R_f 0.2 (ethyl acetate−ethanol, 10:1); IR (ATR) 1703, 1603, 1508, 989, 966, 887, 835, 804, 758, 727, 704, 652 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.03 (d, 3H, J = 7.3 Hz), 2.52−2.59 (m, 1H), 2.90 (s, 3H), 3.07 (s, 3H), 3.51 (s, 3H), 4.81 (d, 1H, J = 5.6 Hz), 6.98−7.04 (m, 2H), 7.05−7.12 (m, 2H), 7.27−7.35 (m, 2H), 7.51−7.58 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 12.0 (q), 32.8 (q), 33.4 (q), 41.5 (d), 52.0 (q), 67.5 (q), 88.9 (s), 115.2 (d, J_{CCF} = 21.6 Hz), 115.6 (d, J_{CCF} = 21.6 Hz), 127.5 (d, J_{CCCF} = 8.4 Hz), 128.7 (d, J_{CCCF} = 8.4 Hz), 134.0 (s, J_{CCCCF} = 3.6 Hz), 138.7 (s, J_{CCCCF} = 2.4 Hz), 153.9 (s), 162.2 (s, J_{CF} = 248.3 Hz), 162.5 (s, J_{CF} = 248.0 Hz); HRMS (ESI, ion trap) calcd for $C_{21}H_{23}F_2N_2O_3$ (M + H^+) 389.1677; found 389.1675.

Methyl (R)-2-((R*)-5,5-Bis(4-methoxyphenyl)-3-methyl-2-(methylimino)oxazolidin-4-yl)propanoate (**16j**).* Colorless paste (92 mg, 89%); R_f 0.3 (ethyl acetate−ethanol, 1:1); IR (ATR) 1697, 1609, 1582, 1508, 986, 964, 827, 772, 729, 712, 667, 652 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.00 (d, 3H, J = 7.0 Hz), 2.21 (brs, 3H), 2.55−2.61 (m, 1H), 2.84 (s, 3H), 3.05 (s, 3H), 3.51 (s, 3H), 3.78 (s, 3H), 3.80 (s, 3H), 4.76 (d, 1H, J = 5.3 Hz), 6.80−6.85 (m, 2H), 6.86−6.91 (m, 2H), 7.21−7.27 (m, 2H), 7.44−7.49 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 11.9 (q), 33.1 (q), 33.2 (q), 41.6 (d), 52.0 (q), 55.2 (q), 55.3 (q), 67.5 (d), 88.9 (s), 113.4 (d), 113.8 (d), 127.0 (d), 128.1 (d), 131.0 (s), 135.7 (s), 154.4 (s), 159.0 (s), 159.3 (s), 175.2 (s); HRMS (ESI, ion trap) calcd for $C_{23}H_{29}N_2O_5$ (M + H^+) 413.2076; found 413.2074.

Isomerization of *cis*-Adducts to *trans*-Adducts. A mixture of ***cis*-3h** (103 mg, 0.25 mmol) and DMAP (10 mg) was heated under a nitrogen atmosphere for 24 h. After cooling to ambient temperature, the mixture was purified by column chromatography on silica gel (hexanes−EtOAc) to give ***trans*-3h** (69 mg) in 67% yield.

(5R,6S*)-6-(Diphenyl((trimethylsilyl)oxy)methyl)-1,3,5-trimethyldihydropyrimidine-2,4(1H,3H)-dione (**trans-3h**).* White solid (69 mg, 67%); R_f 0.4 (hexanes−ethyl acetate, 2:1); mp 148−149 °C; IR (ATR) 1703, 1659, 1508, 1481, 995, 953, 922, 891, 868, 835, 789, 779, 752, 746, 718, 708, 660 cm^{-1}; ^1H NMR ($CDCl_3$) δ −0.25 (s, 9H), 1.33 (d, 3H, J = 7.5 Hz), 2.42 (s, 3H), 2.91 (q, 1H, J = 7.5 Hz), 3.22 (s, 3H), 4.09 (s, 1H), 7.27−7.39 (m, 10H); ^{13}C NMR ($CDCl_3$) δ 1.5 (q), 18.4 (q), 26.8 (q), 37.3 (d), 40.5 (q), 69.6 (d), 84.5 (s), 127.5 (d), 128.1 (d), 128.4 (d), 128.6 (d), 128.7 (d), 139.8 (s), 140.5 (s), 153.0 (s), 171.9 (s). Anal. Calcd for $C_{23}H_{30}N_2O_3Si$: C, 67.28; H, 7.37; N, 6.82. Found: C, 67.39; H, 7.42; N, 6.75.

(5R,6S*)-6-(Bis(4-fluorophenyl)((trimethylsilyl)oxy)methyl)-1,3,5-trimethyldihydropyrimidine-2,4(1H,3H)-dione (**trans-3i**).* Colorless paste (70 mg, 63%); R_f 0.5 (hexanes−ethyl acetate, 2:1); IR (ATR) 1705, 1659, 1603, 1506, 1487, 999, 934, 920, 897, 874, 839, 822, 812, 752, 731, 689, 675 cm^{-1}; ^1H NMR ($CDCl_3$) δ −0.23 (s, 9H), 1.34 (d, 3H, J = 7.5 Hz), 2.50 (s, 3H), 2.84 (q, 1H, J = 7.5 Hz), 3.22 (s, 3H), 4.03 (s, 1H), 7.01−7.06 (m, 2H), 7.08−7.13 (m, 2H), 7.29−7.37 (m, 4H); ^{13}C NMR ($CDCl_3$) δ 1.5 (q), 18.4 (q), 26.8 (q), 37.2 (d), 40.6 (q), 69.7 (d), 83.7 (s), 114.5 (d, J_{CCF} = 21.6 Hz), 115.5 (d, J_{CCF} = 21.6 Hz), 130.4 (d, J_{CCCF} = 7.8 Hz), 130.5 (d, J_{CCCF} = 7.8 Hz), 135.4 (s, J_{CCCCF} = 2.4 Hz), 136.2 (s, J_{CCCCF} = 3.0 Hz), 152.8 (s), 162.3 (s, J_{CF} = 248.3 Hz), 162.5 (s, J_{CF} = 250.7 Hz), 171.6 (s); HRMS (ESI, ion trap) calcd for $C_{23}H_{29}F_2N_2O_3Si$ (M + H^+) 447.1916; found 447.1914.

(5R,6S*)-6-(Bis(4-methoxyphenyl)((trimethylsilyl)oxy)methyl)-1,3,5-trimethyldihydropyrimidine-2,4(1H,3H)-dione (**trans-3j**).* Colorless paste (88 mg, 75%); R_f 0.6 (hexanes−ethyl acetate, 1:1); IR (ATR) 1703, 1661, 1609, 1580, 1508, 1485, 999, 934, 895, 876, 837, 806, 768, 752, 727, 679 cm^{-1}; ^1H NMR ($CDCl_3$) δ −0.24 (s, 9H), 1.32 (d, 3H, J = 7.5 Hz), 2.48 (s, 3H), 2.87 (q, 1H, J = 7.5 Hz), 3.22 (s, 3H), 3.81 (s, 3H), 3.84 (s, 3H), 4.02 (s, 1H), 6.82−6.92 (m, 4H), 7.23−7.32 (m, 4H); ^{13}C NMR ($CDCl_3$) δ 1.5 (q), 18.4 (q), 26.9 (q), 37.2 (d), 40.5 (q), 55.18 (q), 55.24 (q), 69.9 (d), 83.8 (s), 112.8 (d), 113.6 (d), 129.9 (d), 130.0 (d), 131.8 (s), 132.6 (s), 153.0 (s), 159.1 (s), 159.5 (s), 171.9 (s); HRMS (ESI, ion trap) calcd for $C_{25}H_{35}N_2O_5Si$ (M + H^+) 471.2315; found 471.2312.

(5R,6R*)-6-(Diphenyl((trimethylsilyl)oxy)methyl)-5-fluoro-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (**trans-3k**).* White solid (73 mg, 70%); R_f 0.4 (hexanes−ethyl acetate, 5:1); mp 163−165 °C; IR (ATR) 1715, 1670, 1487, 978, 955, 907, 876, 839, 789, 772, 750, 716, 704, 652 cm^{-1}; ^1H NMR ($CDCl_3$) δ −0.20 (s, 9H), 2.60 (s, 3H), 3.04 (s, 3H), 4.61 (d, 1H, J_{HF} = 24.7 Hz), 5.15 (d, 1H, J_{HF} = 45.8 Hz), 7.34−7.44 (m, 10H); ^{13}C NMR ($CDCl_3$) δ 1.5 (q), 27.1 (q), 39.5 (q), 68.3 (d, J_{CCF} = 19.2 Hz), 82.7 (s, J_{CCCF} = 10.8 Hz), 85.0 (d, J_{CF} = 175.1 Hz), 128.0 (d), 128.3 (d), 128.46 (d), 128.47 (d), 128.7 (d), 129.0 (d), 139.2 (s), 139.7 (s), 152.1 (s), 163.1 (s, J_{CCF} = 20.4 Hz). Anal. Calcd for $C_{22}H_{27}FN_2O_3Si$: C, 63.74; H, 6.57; N, 6.76. Found: C, 63.78; H, 6.60; N, 6.67.

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(5R,6R*)-6-(Bis(4-methoxyphenyl)((trimethylsilyl)oxy)methyl)-5-fluoro-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (**trans-3m**).* Colorless paste (97 mg, 82%); R_f 0.65 (hexanes−ethyl acetate, 2:1); IR (ATR) 1717, 1670, 1609, 1578, 1508, 1481, 999, 976, 951, 908, 878, 839, 804, 781, 748, 729, 685, 669 cm^{-1}; ^1H NMR ($CDCl_3$) δ 0.19 (s, 9H), 2.65 (s, 3H), 3.04 (s, 3H), 3.82 (s, 3H), 3.84 (s, 3H), 4.53 (d, 1H, J_{HF} = 24.2 Hz), 5.12 (d, 1H, J_{HF} = 46.1 Hz), 6.85−6.93 (m, 4H), 7.26−7.43 (m, 4H); ^{13}C NMR ($CDCl_3$) δ 1.6 (q), 27.1 (q), 39.4 (q), 55.27 (q), 55.29 (q), 68.7 (d, J_{CCF} = 19.2 Hz), 81.9 (s, J_{CCCF} = 10.8 Hz), 85.1 (d, J_{CF} = 175.1 Hz), 113.2 (d), 113.6 (d), 129.7 (d), 129.8 (d), 131.3 (s), 131.8 (s), 152.1 (s), 159.5 (s), 159.7 (s), 163.2 (s, J_{CCF} = 20.1 Hz); HRMS (ESI, ion trap) calcd for $C_{24}H_{32}FN_2O_5Si$ (M + H^+) 475.2065; found 475.2063.

(5R,6S*)-6-(3,4-Dihydronaphthalen-1-yl)-1,3,5-trimethyldihydropyrimidine-2,4(1H,3H)-dione (**trans-8f**).* Colorless paste (59 mg, 83%); R_f 0.35 (hexanes−ethyl acetate, 2:1); IR (ATR) 1744, 1707, 1661, 1599, 1477, 949, 920, 905, 876, 833, 804, 791, 758, 733, 691, 673, 664 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.41 (d, 3H, J = 7.3 Hz), 2.23−2.35 (m, 2H), 2.66−2.76 (m, 2H), 2.90−2.96 (m, 1H), 3.08 (s, 3H), 3.21 (s, 3H), 4.21 (brs, 1H), 5.76 (t, 1H, J = 4.0 Hz), 7.00−7.04 (m, 1H), 7.17−7.23 (m, 3H); ^{13}C NMR ($CDCl_3$) δ 16.8 (q), 22.7 (t), 27.6 (q), 27.8 (t), 35.4 (q), 40.1 (d), 61.4 (d), 121.3 (d), 125.4 (d), 126.5 (d), 127.5 (d), 128.3 (d), 131.78 (s), 131.83 (s), 137.3 (s), 153.8 (s), 171.9 (s); HRMS (ESI, ion trap) calcd for $C_{17}H_{21}N_2O_2$ (M + H^+) 285.1603; found 285.1602.

(5R,6R*)-6-(3,4-Dihydronaphthalen-1-yl)-5-fluoro-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (**trans-8h**).* Colorless paste (42 mg, 58%); R_f 0.55 (hexanes−ethyl acetate, 2:1); IR (ATR) 1721, 1670, 1474, 968, 918, 806, 795, 760, 733, 687, 675 cm^{-1}; ^1H NMR ($CDCl_3$) δ 2.23−2.37 (m, 2H), 2.66−2.78 (m, 2H), 3.11 (s, 3H), 3.27 (s, 3H), 4.80−4.86 (m, 1H), 4.96 (dd, 1H, J = 2.5 Hz, J_{HF} = 47.0 Hz), 5.83−5.86 (m, 1H), 7.17−7.29 (m, 4H); ^{13}C NMR ($CDCl_3$) δ 22.8 (t), 27.5 (t), 27.8 (q), 35.2 (q), 60.0 (d, J_{CCF} = 22.8 Hz), 84.9 (d, J_{CF} = 185.9 Hz), 120.9 (d), 126.9 (d), 127.0 (s, J_{CCCF} = 9.6 Hz), 128.0 (d), 128.1 (d), 128.5 (d), 131.1 (s), 137.0 (s), 152.5 (s), 163.3 (s, J_{CCF} = 21.6 Hz); HRMS (ESI, ion trap) calcd for $C_{16}H_{17}FN_2O_2$ (M + H^+) 289.1352; found 289.1351.

(5R,6S*)-1,3,5-Trimethyl-6-(naphthalen-1-yl)dihydropyrimidine-2,4(1H,3H)-dione (**trans-10f**).* Colorless paste (52 mg, 74%); R_f 0.35 (hexanes−ethyl acetate, 2:1); IR (ATR) 1707, 1661, 1599, 1510, 1479, 999, 908, 797, 789, 775, 758, 727, 664 cm^{-1}; ^1H NMR ($CDCl_3$) δ 1.55 (d, 3H, J = 7.5 Hz), 3.09 (s, 3H), 3.12−3.17 (m, 1H), 3.26 (s, 3H), 4.95 (d, 1H, J = 2.3 Hz), 7.08 (d, 1H, J = 7.5 Hz), 7.42 (t, 1H, J = 8.0 Hz), 7.51−7.60 (m, 2H), 7.75 (d, 1H, J = 8.6 Hz), 7.82 (d, 1H, J = 8.6 Hz), 7.90−7.93 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 17.4 (q), 27.7 (q), 35.6 (q), 42.5 (d), 61.5 (d), 121.6 (d), 122.0 (d), 125.3 (d), 126.0 (d), 126.8 (d), 129.1 (d), 129.5 (d), 130.1 (s), 132.5 (s), 134.3 (s), 154.0 (s), 171.6 (s); HRMS (ESI, ion trap) calcd for $C_{17}H_{19}N_2O_2$ (M + H^+) 283.1447; found 283.1445.

*1,3-Dimethyl-6-(naphthalen-1-yl)pyrimidine-2,4(1H,3H)-dione (**17h**).* Colorless paste (59 mg, 88%); R_f 0.35 (hexanes−ethyl acetate, 2:1); IR (ATR) 1701, 1647, 1616, 1508, 1474, 995, 939, 916, 866, 826, 804, 779, 760, 725, 694, 675 cm^{-1}; ^1H NMR ($CDCl_3$) δ 3.04 (s, 3H), 3.49 (s, 3H), 5.84 (s, 1H), 7.42−7.45 (m, 1H), 7.54−7.61 (m, 3H), 7.62−7.67 (m, 1H), 7.92−7.97 (m, 1H), 7.98−8.01 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 28.1 (q), 33.6 (q), 103.4 (d), 124.2 (d), 125.2 (d), 126.4 (d), 126.9 (d), 127.7 (d), 128.7 (d), 130.1 (s), 130.5 (d), 130.6 (s), 133.2 (s), 152.5 (s), 153.4 (s), 162.5 (s); HRMS (ESI, ion trap) calcd for $C_{16}H_{15}N_2O_2$ (M + H^+) 267.1134; found 267.1133.

(5R,6R*)-5-Fluoro-1,3-dimethyl-6-(naphthalen-1-yl)dihydropyrimidine-2,4(1H,3H)-dione (**trans-10h**).* Colorless paste (54 mg, 95%); R_f 0.55 (hexanes−ethyl acetate, 2:1); IR (ATR) 1721, 1670, 1599, 1508, 1476, 970, 910, 868, 797, 789, 772, 750, 729, 687 cm^{-1}; ^1H NMR ($CDCl_3$) δ 3.14 (s, 3H), 3.31 (s, 3H), 5.18 (dd, 1H, J = 2.6 Hz, J_{HF} = 47.1 Hz), 5.54 (dd, 1H, J = 2.6 Hz, J_{HF} = 16.5 Hz), 7.09−7.13 (m, 1H), 7.43−7.48 (m, 1H), 7.56−7.61 (m, 1H), 7.63−7.67 (m, 1H), 7.86−7.90 (m, 1H), 7.91−7.96 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 27.9 (q), 35.3 (q), 60.2 (d, J_{CCF} = 22.8 Hz), 86.0 (d, J_{CF} = 187.1 Hz), 121.0 (d), 123.2 (d), 125.4 (d), 126.38 (s), 126.44 (d), 127.5 (d), 129.7 (d), 130.2 (d and s), 134.2 (s), 152.7 (d), 163.2 (s, J_{CCF} = 21.6 Hz); HRMS (ESI, ion trap) calcd for $C_{16}H_{16}FN_2O_2$ (M + H^+) 287.1196; found 287.1195.

(5R,6R*)-5-Fluoro-6-(hydroxydiphenylmethyl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (**trans-11k**).* White solid (81 mg, 95%); R_f 0.4 (hexanes−ethyl acetate, 2:1); mp 233−235 °C; IR (ATR) 3329, 1717. 1659, 1491, 986, 976, 899, 827, 799, 772, 752, 739, 696, 660 cm^{-1}; ^1H NMR ($CDCl_3$) δ 2.47 (s, 3H), 2.88 (brs, 1H), 3.11 (s, 3H), 4.44 (d, 1H, J_{HF} = 20.3 Hz), 5.08 (d, 1H, J_{HF} = 46.5 Hz), 7.30−7.46 (m, 10H); ^{13}C NMR ($CDCl_3$) δ 27.1 (q), 38.0 (q), 66.0 (d, J_{CCF} = 18.6 Hz), 78.4 (s, J_{CCCF} = 10.8 Hz), 84.9 (d, J_{CF} = 176.3 Hz), 124.9 (d), 125.7 (d), 126.5 (d), 126.6 (d), 127.0 (d), 127.4 (d), 142.7 (s), 143.0 (s), 152.7 (s), 162.8 (s, J_{CF} = 20.4 Hz). Anal. Calcd for $C_{19}H_{19}FN_2O_3$: C,66.66; H, 5.59; N, 8.18. Found: C, 66.60; H, 5.59; N, 8.12.

(5R,6R*)-5-Fluoro-6-(hydroxybis(4-methoxyphenyl)methyl)-1,3-dimethyldihydropyrimidine-2,4(1H,3H)-dione (**trans-11m**).* Colorless paste (92 mg, 92%); R_f 0.5 (hexanes−ethyl acetate, 1:1); IR (ATR) 3422, 1717, 1655, 1607, 1582, 1508, 1485, 976, 908, 831, 804, 777, 754, 727, 681 cm^{-1}; ^1H NMR ($CDCl_3$) δ 2.55 (s, 3H), 2.57 (brs, 1H), 3.09 (s, 3H), 3.80 (s, 3H), 3.83 (s, 3H), 4.34 (d, 1H, J_{HF} = 20.6 Hz), 5.08 (d, 1H, J_{HF} = 46.4 Hz), 6.84−6.89 (m, 2H), 6.92−6.96 (m, 2H), 7.23−7.28 (m, 2H), 7.31−7.36 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 27.4 (q), 38.4 (q), 55.2 (q), 55.3 (q), 67.5 (d, J_{CCF} = 19.2 Hz), 79.1 (s, J_{CCCF} = 10.8 Hz), 85.5 (d, J_{CF} = 176.3 Hz), 113.8 (d), 114.1 (d), 127.2 (d), 127.9 (d), 134.1 (s), 134.2 (s), 152.8 (s), 159.3 (s), 159.4 (s), 164.4 (s, J_{CCF} = 19.2 Hz); HRMS (ESI, ion trap) calcd for $C_{21}H_{23}FN_2O_5$ (M + H^+) 403.1669; found 403.1668.

(R)-N-Methyl-2-((S*)-3-methyl-2-oxo-5,5-diphenyloxazolidin-4-yl)propanamide (**threo-12h**).* White solid (29 mg, 34%); R_f 0.3 (ethyl acetate); mp 215 °C; IR (ATR) 3300, 1738, 1663, 1560, 1493, 995, 941, 916, 901, 835, 756, 700, 683, 664 cm^{-1}; ^1H NMR ($CDCl_3$) δ 0.84 (d, 3H, J = 6.9 Hz), 2.60 (d, 3H, J = 4.6 Hz), 2.68−2.75 (m, 1H), 2.88 (s, 3H), 4.99 (d, 1H, J = 5.4 Hz), 5.41 (brs, 1H), 7.19−7.34 (m, 4H), 7.36−7.41 (m, 2H), 7.42−7.47 (m, 2H), 7.64−7.69 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 11.4 (q), 26.5 (q), 30.4 (q), 40.7 (d), 67.3 (d), 87.2 (s), 125.6 (d), 127.3 (d), 127.8 (d), 127.9 (d), 128.3 (d), 128.6 (d), 137.8 (s), 143.6 (s), 156.9 (s), 172.8 (s). Anal. Calcd for $C_{20}H_{22}N_2O_3$: C, 70.99; H, 6.55; N, 8.28. Found: C, 70.95; H, 6.52; N, 8.18.

(R)-2-((S*)-5,5-Bis(4-fluorophenyl)-3-methyl-2-oxooxazolidin-4-yl)-N-methylpropanamide (**threo-12i**).* White solid (23 mg, 25%); R_f 0.5 (ethyl acetate); mp 231−232 °C; IR (ATR) 3316, 1744, 1665, 1655, 1603, 1566, 1508, 995, 947, 903, 849, 843, 826, 806, 773, 754, 694, 677 cm^{-1}; ^1H NMR ($CDCl_3$) δ 0.83 (d, 3H, J = 7.3 Hz), 2.63 (d, 3H, J = 4.9 Hz), 2.69−2.76 (m, 1H), 2.88 (s, 3H), 5.03 (d, 1H, J = 5.2 Hz), 5.50 (brs, 1H), 6.93−6.99 (m, 2H), 7.04−7.10 (m, 2H), 7.38−7.43 (m, 2H), 7.61−7.66 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 10.5 (q), 26.6 (q), 30.1 (q), 40.3 (d), 67.0 (d), 86.4 (s), 114.7 (d, J_{CCF} = 21.6 Hz), 115.6 (d, J_{CCF} = 21.6 Hz), 127.4 (d, J_{CCCF} = 8.4 Hz), 129.4 (d, J_{CCCF} = 8.4 Hz), 133.6 (s, J_{CCCCF} = 2.7 Hz), 139.5 (s, J_{CCCCF} = 2.7 Hz), 156.5 (s), 162.1 (s, J_{CF} = 248.3 Hz), 162.5 (s, J_{CF} = 248.0 Hz), 172.6 (s). Anal. Calcd for $C_{20}H_{20}F_2N_2O_3$: C, 64.16; H, 5.38; N, 7.48. Found: C, 64.02; H, 5.42; N, 7.39.

(R)-2-((S*)-5,5-Bis(4-methoxyphenyl)-3-methyl-2-oxooxazolidin-4-yl)-N-methylpropanamide (**threo-12j**).* Colorless paste (28 mg, 28%); R_f 0.35 (ethyl acetate); IR (ATR) 3321, 1742, 1649, 1609, 1580, 1541, 1508, 989, 905, 826, 789, 773, 756, 727, 677 cm^{-1}; ^1H NMR ($CDCl_3$) δ 0.81 (d, 3H, J = 7.0 Hz), 2.63 (d, 3H, J = 4.6 Hz), 2.65−2.71 (m, 1H), 2.88 (s, 3H), 3.76 (s, 3H), 3.80 (s, 3H), 4.85 (d, 1H, J = 5.9 Hz), 5.58 (brs, 1H), 6.76−6.81 (m, 2H), 6.87−6.92 (m, 2H), 7.29−7.34 (m, 2H), 7.51−7.56(m, 2H); ^{13}C NMR ($CDCl_3$) δ 11.8 (q), 26.5 (q), 30.6 (q), 40.9 (d), 55.2 (q), 55.3 (q), 67.5 (d), 87.2 (s), 113.0 (d), 113.8 (d), 126.9 (d), 128.8 (d), 130.3 (s), 135.7 (s), 157.1 (s), 159.0 (s), 159.4 (s), 173.1 (s); HRMS (ESI, ion trap) calcd for $C_{22}H_{27}N_2O_5$ (M + H^+) 399.1920; found 399.1918.

(R)-2-Fluoro-N-methyl-2-((R*)-3-methyl-2-oxo-5,5-diphenyloxazolidin-4-yl)acetamide (**erythro-12k**).* White solid (74 mg, 87%); R_f 0.45 (hexanes−ethyl acetate, 1:5); mp 203−204 °C; IR (ATR) 3312, 1775, 1763, 1717, 1672, 1655, 1551, 1483, 951, 910, 899, 851, 775, 768, 752, 719, 700, 675, 658 cm^{-1}; ^1H NMR ($CDCl_3$) δ 2.34 (d, 3H, J = 5.2 Hz), 2.97 (s, 3H), 5.07 (dd, 1H, J = 2.2 Hz, J_{HF} = 28.2 Hz), 5.15 (dd, 1H, J = 2.2 Hz, J_{HF} = 11.5 Hz), 5.21 (brs, 1H), 7.19−7.29 (m, 3H),

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7.33−7.46 (m, 5H), 7.63−7.66 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 26.2 (q), 28.9 (q), 65.8 (d, J_{CCF} = 16.8 Hz), 85.8 (s), 86.6 (d, J_{CF} = 201.5 Hz), 125.7 (d), 127.5 (d), 128.0 (d), 128.3 (d), 128.6 (d), 136.6 (s), 142.5 (s), 156.5 (s), 166.8 (s, J_{CCF} = 16.8 Hz). Anal. Calcd for $C_{19}H_{19}FN_2O_3$: C,66.66; H, 5.59; N, 8.18. Found: C, 66.65; H, 5.61; N, 8.13.

(R)-2-((R*)-5,5-Bis(4-methoxyphenyl)-3-methyl-2-oxooxazolidin-4-yl)-2-fluoro-N-methylacetamide (**erythro-12m**).* White solid (93 mg, 92%); R_f 0.5 (ethyl acetate); mp 197−199 °C; IR (ATR) 3545, 3368, 1732, 1674, 1609, 1557, 1510, 999, 964, 955, 928, 897, 853, 824, 799, 770, 758, 729, 694, 667 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 2.39 (d, 3H, J = 5.0 Hz), 2.96 (s, 3H), 3.76 (s, 3H), 3.80 (s, 3H), 5.02 (dd, 1H, J = 2.0 Hz, J_{HF} = 6.2 Hz), 5.10 (dd, 1H, J = 2.0 Hz, J_{HF} = 22.5 Hz), 5.33 (brs, 1H), 6.73−6.79 (m, 2H), 6.88−6.93 (m, 2H), 7.24−7.33 (m, 2H), 7.50−7.55 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 26.1 (q), 29.0 (q), 55.2 (q), 55.3 (q), 66.1 (d, J_{CCF} = 16.8 Hz), 85.6 (s), 86.7 (d, J = 201.5 Hz), 112.7 (d), 113.0 (d), 127.1 (d), 129.2 (s), 129.8 (d), 134.8 (s), 156.6 (s), 159.1 (s), 159.5 (s), 166.9 (s, J_{CCF} = 16.8 Hz). Anal. Calcd for $C_{21}H_{23}FN_2O_5$: C,62.68; H, 5.76; N, 6.96. Found: C, 62.77; H, 5.79; N, 6.85.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.joc.6b00670.

X-ray crystallographic data of **3g** (CIF)
X-ray crystallographic data of ***trans*-3h** (CIF)
X-ray crystallographic data of **8e** (CIF)
X-ray crystallographic data of ***cis*-8h** (CIF)
X-ray crystallographic data of ***cis*-8i** (CIF)
X-ray crystallographic data of **11b** (CIF)
X-ray crystallographic data of **11d** (CIF)
X-ray crystallographic data of ***trans*-11h** (CIF)
X-ray crystallographic data of ***cis*-11i** (CIF)
X-ray crystallographic data of ***cis*-11j′** (CIF)
X-ray crystallographic data of ***trans*-11j** (CIF)
X-ray crystallographic data of ***cis*-11n** (CIF)
X-ray crystallographic data of **12b** (CIF)
X-ray crystallographic data of **12c** (CIF)
X-ray crystallographic data of **12e** (CIF)
X-ray crystallographic data of ***threo*-12h** (CIF)
X-ray crystallographic data of ***threo*-12k** (CIF)
X-ray crystallographic data of ***erythro*-12k** (CIF)
X-ray crystallographic data of ***threo*-12l** (CIF)
X-ray crystallographic data of ***threo*-12n** (CIF)
X-ray crystallographic data of ***erythro*-13e** (CIF)
X-ray crystallographic data of ***trans-erythro*-13f** (CIF)
X-ray crystallographic data of ***cis-threo*-13g** (CIF)
X-ray crystallographic data of ***trans-threo*-13g** (CIF)
X-ray crystallographic data of ***cis-erythro*-13h** (CIF)
X-ray crystallographic data of ***trans*-15i** (CIF)
X-ray crystallographic data of ***trans*-15j** (CIF)
^{1}H and ^{13}C NMR spectra of new compounds, X-ray crystallographic data (ORTEP) of **3g**, ***trans*-3h**, **8e**, ***cis*-8h**, ***cis*-8i**, **11b**, **11d**, ***trans*-11h**, ***cis*-11i**, ***cis*-11j′**, ***trans*-11j**, ***cis*-11n**, **12b**, **12c**, **12e**, ***threo*-12h**, ***threo*-12k**, ***erythro*-12k**, ***threo*-12l**, ***threo*-12n**, ***erythro*-13e**, ***trans-erythro*-13f**, ***cis-threo*-13g**, ***trans-threo*-13g**, ***cis-erythro*-13h**, ***trans*-15i**, and ***trans*-15j**, and DFT calculation data (PDF)

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Notes

The authors declare no competing financial interest.

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