



Green synthesis of symmetric aryl ether dinitro compounds in deep eutectic solvents

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A series of symmetric aryl ether dinitro compounds were synthesized by 4Urea/ZnCl₂ as green solvent and catalyst, and halogenated aromatic hydrocarbon derivatives and dihydroxy compounds as raw materials. This method has the advantages of easy operation, simple recrystallization, environmental friendliness and biodegradability. The structures of the products were subjected to ¹H NMR, and the reaction conditions, reaction mechanism and greening advantages of deep eutectic solvents were discussed at the end of the experiments. We learned from the results of the experiments that both the aliphatic dihydroxy compounds and the aromatic dihydroxy compounds could proceed effectively in Ullmann reaction, and the desired products could be obtained in acceptable yields. When using this methodology, symmetric aryl ether dinitro compounds could be synthesized simply at 80°C in 2 h with high yields (66%~94%). Meanwhile, the DES, called deep eutectic solvent which is composed of 4Urea/ZnCl₂, could be recycled at least five times without an obvious decrease in the catalytic activity.

Keywords: Deep eutectic solvent, Ullmann reaction, symmetric aryl ether dinitro compound, fine chemical intermediates.

Introduction

Symmetric aryl ether dinitro compounds are main raw materials for the preparation of aromatic polyimide (PI)^{1,2}. Polyimide has a wide application in aerospace and microelectronics for their many unique properties, such as excellent high-temperature resistance, outstanding mechanical properties, good chemical corrosion resistance, small thermal expansion coefficient and low dielectric constant^{3,4}. However, aromatic polyimides prepared from aromatic tetracarboxylic anhydride and aromatic amine have poor light transmittance resulting from their coloration. It is considered that the main reason for this defect is the formation of a charge transfer complex between the aromatic moieties in these polymers⁵. These problems limit the further applications of these polyimides. Recently, during the synthesis, flexible groups such as trifluoromethyl and ether bonds are introduced into the dinitro structure, which can improve the poor light transmittance and solubility of polyimide. Since then, they are widely used for alignment layers (ALs) of advanced thin film transistor liquid crystal display devices (TFT-LCDs) and photovoltaic materials⁶.

Since it is the important precursor of PI, many relevant research workers are devoted to the optimization of its reaction conditions. At present, there are many methodologies

focused on the synthesis of aromatic ethers processes, including Goldberg reaction, Buchwald and Hartwig coupling⁷, Williamson reaction⁸, Ullmann coupling⁹, etc. Ullmann coupling is one of the most common methods for the synthesis of aromatic ethers. However, some drawbacks may exist in these reactions, such as excessive reaction time, high reaction temperatures, expensive metal catalysts, cumbersome post-processing, low yields and so on. Moreover, highly polar toxic solvents and unrecyclable catalysts are used in them. Therefore, it is still of positive significance to explore the green, efficient and simple methods for the synthesis of symmetric aryl ether dinitro compounds (Table 1).

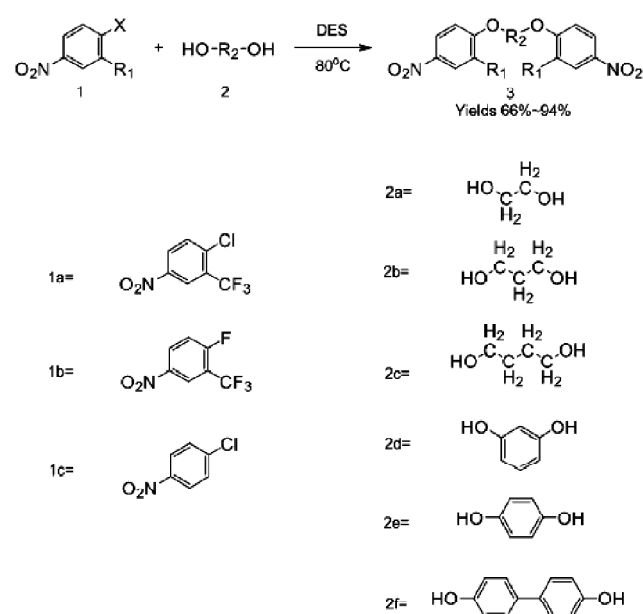
Within the concept of green chemistry, solvents and catalysts constitute the extremely critical part. Replacing hazardous solvents and non-recyclable catalysts with cleaner and more environmentally friendly components is the current means and concepts of green chemical industry in recent years. A hot topic of research for this purpose has been performed, which is about ionic liquids (ILs). However, there are many problems in the use of ILs like complicated synthesis steps, difficulty in purification and use of expensive solvents, which greatly limit their application. Thence, deep eutectic solvents (DESSs), as an alternative to ILs, have been proposed. DESSs were first depicted by Abbot *et al.*¹³, which are

Table 1. Comparison of efficiency of various methods in synthesis of aromatic ether derivatives

Entry	Catalyst	Conditions	Time (h)	Yield(%) ^b	Ref.
1	[NBu ₄][Pd(DMSO)Cl ₃]	K ₃ PO ₄ , DMF, 50–140°C	3–6	12–97	10
2	N,O-Cu@AS-MNPs	K ₂ CO ₃ , DMSO, 120°C	24	15–96	11
3	CuCl	NaOH, NMP, 140–170°C	8	15–64	12
4	DES	NMP, 80°C			Present work

formed from quaternary ammonium salts and amide compounds with low melting point. Not only do DESs have analogous characteristics to those of ILs, such as low vapor pressure, non-flammability and adjustable structure and performance, but they are also reusable, lower cost and biodegradable. Because of these excellent properties, DESs have received extensive attention in the fields of electrochemistry¹⁴, biocatalysis¹⁵, separation and purification¹⁶, nanotechnology¹⁷, etc. More prominently, DESs can even exert strong H-bond interaction and coordination if they are prepared from urea and metal salts.

For the above reasons, in this paper, we reported the Ullmann protocol using 4Urea/ZnCl₂ both as catalyst and solvent to synthesis symmetric aryl ether dinitro groups, which are the precursor of PI. The adopted method and conclusions obtained in this study will be presented in later chapters. The synthetic route is as follows (Fig. 1).

**Fig. 1.** Synthesis route of symmetric aryl ether dinitro compound in deep eutectic solvent.

Results and discussion

At first the reaction of preparing 1,2'-bis(4-nitrotrifluoromethylphenoxy)ethane (**3a**) with 2-chloro-5-nitrobenzotrifluoride (**1a**) and 1,2-ethanediol (**2a**) (model reaction) was examined without using 4Urea/ZnCl₂ (DES) that target products had not been observed after a 12 h's prolonged reaction. DESs such as 4Urea:ZnCl₂ (Table 2, entry 6), Urea:2SnCl₂ (Table 2, entry 7), Chloride:PTSA (Table 2, entry 8), Chloride:glycerol (Table 2, entry 9) Chloride:2Oxalic acid (Table 2, entry 10) and Chloride:2ZnCl₂ (Table 2, entry 11) have intrinsic advantages as green catalysts. Therefore, overture were made to investigate the model reaction with these catalysts. Simultaneously, the catalytic activity of the DES was compared with other catalysts reported for preparing symmetric aryl ether dinitro compounds by Ullmann cou-

Table 2. Effect of different catalytic systems on the reaction^a

Entry	Catalyst	Conditions	Time (h)	Yield (%) ^b
1	–	140°C, NMP	12	0
2	5% Fe ₃ O ₄	K ₂ CO ₃ , DMF, 140°C	17	62
3	5% CuI	KOH, NMP, 200°C	24	68
4	5% CuBr	Cs ₂ CO ₃ , DMSO, 160°C	18	65
5	5% CeO ₂	K ₃ PO ₄ , DMSO, 140°C	16	52
6	5% 4Urea/ZnCl ₂	NMP, 80°C	4	88
7	5% Urea/2SnCl ₂	NMP, 80°C	6	67
8	5% ChCl/PTSA	NMP, 80°C	5	40
9	5% ChCl/glycerol	NMP, 80°C	5	22
10	5% ChCl/Ox	NMP, 80°C	6	0
11	5% ChCl/2ZnCl ₂	NMP, 80°C	6	0

^aReaction conditions: a mixture of 2-chloro-5-nitrobenzotrifluoride (**1a**) and 1,2-ethanediol (**2b**) with catalyst (5 mol%) in solvents was heated at 80°C.

^bYield of isolated product.

pling between aryl halides and dihydroxy compounds. With the DES (4Urea/ZnCl₂), the product **3a** was obtained in a satisfactory yield (Table 2, entry 6).

The yields of the catalyst system using dipolar solvents and metal catalysts were generally low (52%~68%), and the process produces a large amount of alkaline wastewater. With respect to the different catalytic effects attributed to different components of DESs, these results illustrated the important role of 4Urea/ZnCl₂ in the catalytic reaction clearly. During the reaction, DES combined with the reactants through hydrogen bonding, to increase the reactivity and promote this process as a benign media. DES could also be used as an alkaline reagent to capture protons in hydroxyl groups and it could improve the nucleophilicity of oxygen. In addition, DES played the role as a dipolar solvent with the advantage of high polarity, which will make nucleophile more uniformly dispersed. The above results showed that the catalytic amount of 4Urea/ZnCl₂ was the most effective catalyst for this reaction, and finally the ideal product yield was obtained within 4 h.

To obtain the optimum reaction conditions, A series of experiments based on the model reaction have been designed using the amount of 4Urea/ZnCl₂ (5, 10, 20, 30% and solvent-free) in different temperatures (50, 60, 70, 80, 90, 100°C). The results of optimization processes were listed in Table 3. The best yield of the product could be obtained by performing the reaction at 80°C in the presence of 5% DES/NMP of catalyst within 4 h (Table 3, entry 4). We performed the reaction in the presence of a larger amount of the catalyst. Under solvent-free conditions, the product **3** was obtained in 93% yield and the reaction time was shortened to 2

Table 3. Effect of different DES dosage and reaction time on the reaction^a

Entry	Conditions	Temp. (°C)	Time (h)	Yield (%) ^b
1	5% DES/NMP	50	4	36
2	5% DES/NMP	60	4	54
3	5% DES/NMP	70	4	74
4	5% DES/NMP	80	4	88
5	5% DES/NMP	90	4	86
6	5% DES/NMP	100	4	85
7	10% DES/NMP	80	4	88
8	20% DES/NMP	80	4	89
9	30% DES/NMP	80	4	91
10	100% DES (solvent-free)	80	2	93

^aReaction conditions: a mixture of 2-chloro-5-nitrobenzotrifluoride (**1a**) and 1,2-ethanediol (**2b**) with DES was heated at 80°C in 4 h.
^bYield of isolated product.

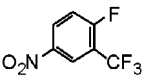
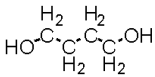
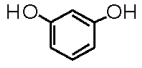
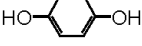
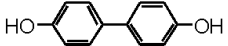
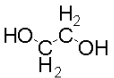
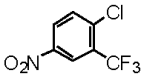
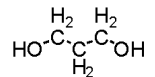
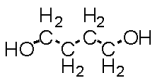
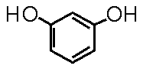

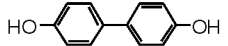
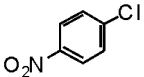
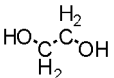
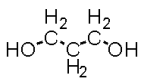
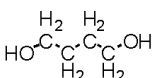
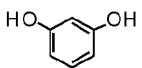

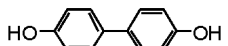
h (Table 3, entry 10), but it can not be the best choice. From the above, the solvent-free reaction did not require the addition of a large polar dipolar solvent, and the post-reaction treatment was simple and convenient.

To examine universality of substrate and show the ability of 4Urea/ZnCl₂ to synthesis various symmetric aryl ether dinitro compounds, different aryl halides were employed with dihydroxy compounds at the optimized conditions. And the results obtained in these reactions were listed in Table 4. These reactions proved that 4Urea/ZnCl₂ could be used as the generalized and valid catalyst for Ullmann protocol syn-

Table 4. Synthesis of symmetrical aromatic dinitro compounds^a

Reactant ₁	Reactant ₂	Conditions	Time (h)	Product	Yield (%) ^b
		DES, 80°C	2	3a	94
		DES, 80°C	2	3b	89

Table-4 (contd.)

		DES, 80°C	3	3c	78
		DES, 80°C	4	3d	92
		DES, 80°C	3	3e	86
		DES, 80°C	2	3f	88
		DES, 80°C	2	3a	93
		DES, 80°C	3	3b	66
		DES, 80°C	2	3c	88
		DES, 80°C	3	3d	92
		DES, 80°C	3.5	3e	78
		DES, 80°C	2	3f	82
		DES, 80°C	3.5	3g	85
		DES, 80°C	3.5	3h	81
		DES, 80°C	2	3i	73
		DES, 80°C	2.5	3j	76
		DES, 80°C	2	3k	78
	DES, 80°C	3	3l	66	

^aReaction conditions: a mixture of an aryl halide (**1**; R₁ = various) and a dihydroxy compound (**2**; R₂ = various) with DES (solvent-free) was heated at 80°C.

^bYield of isolated product.

thesis of symmetric aryl ether dinitro compounds. Both the aliphatic dihydroxy compounds and the aromatic dihydroxy compounds could proceed effectively in the reaction, and the desired product could be obtained in an acceptable yield

in the range of 66–94%. The experimental results showed that the nucleophilicity of substrates, the density of the aromatic ring electron cloud, the steric hindrance and leaving ability of halogen groups were important factors affecting the

yields of reaction. The aliphatic dihydroxy compounds with strong nucleophilic ability were used as the reaction substrates, and the reaction obtained higher product yields than that using aromatic dihydroxy compounds. Although the long carbon chain dihydroxy compounds had higher electron cloud density, they were largely affected by steric hindrance, which resulted in a lower yield of the product than aromatic compounds. For the leaving groups, compared with Cl, F had stronger electronegativity and stronger ability to leave. When a strong electron withdrawing group existed in the *ortho* or *para* position of the aromatic ring, the density of the aromatic ring electrons gets lower, which accelerated the departure rate of halogen groups. If the density of the aromatic ring electron cloud was high (no trifluoromethyl group), the yields would decrease.

According to the above reaction characteristics and reaction product structure, the catalytic reaction mechanism of 4Urea/ZnCl₂ is as shown in Fig. 2. During the reaction, 4Urea/ZnCl₂ mainly activates the reactants through hydrogen bonding, which plays an important role in improving nucleophilic and electrophilic properties. In the aromatic system, the reaction speed increases with the decrease of electron density. The hydrogen bond formed by 4Urea/ZnCl₂ and the reactants is an important factor for raising the potential, enhancing the electrophilicity of the aryl halide and the nucleophilicity of the latter. This process promoted the attack of the dihydroxy compounds on the forming of a resonance stable

structure. Finally, the departure of Cl⁻ is accelerated, which also promoted the formation of the product.

Recycling of the 4Urea/ZnCl₂ catalyst:

To investigate the recyclability of DES, the reaction solution was added to water (50 mL) and shaken vigorously after completing the reaction. The product was extracted from organic phase by using ethyl acetate (50 mL). The lower aqueous phase was evaporated to get pure DES which should be dried at 60°C and could be used repeatedly (the next four times) without further purification. The results were listed in Table 5 and they showed that if the catalyst was not repeatedly used more than 5 times, the catalytic activity would not be significantly lowered and the yield of the product not be significantly lost.

Table 5. Performance of the DES cycle^a

Entry	Cycle	Time (h)	Yield (%) ^b
1	1st run	2	93
2	2nd run	2	93
3	3rd run	2	92
4	4th run	2	91
5	5th run	2	89
6	6th run	2	85

^aReaction conditions: a mixture of 2-chloro-5-nitrobenzotrifluoride (**1**; R₁ = various) and 1,2-ethanediol (**2**; R₂ = various) with DES (solvent-free) was heated in 2 h at 80°C.

^bYield of isolated product.

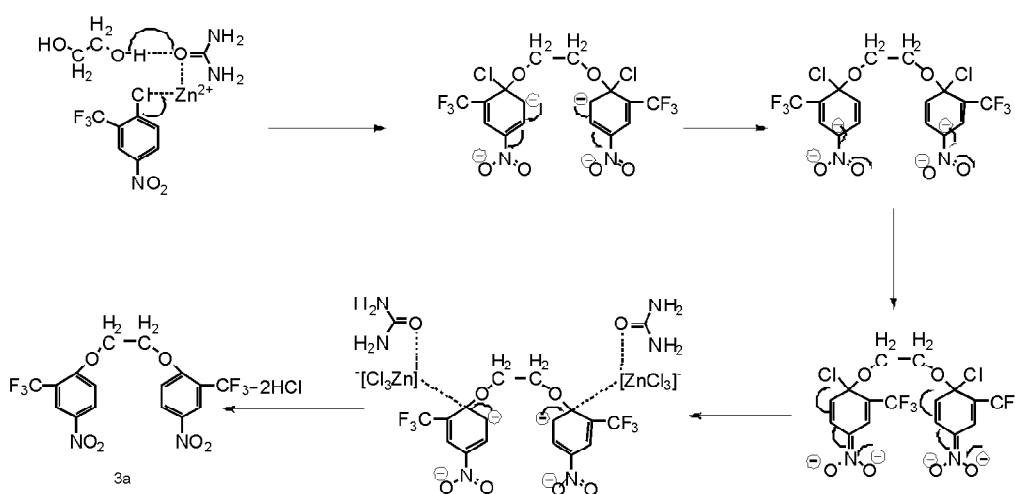


Fig. 2. Possible mechanism of synthesis of symmetric aryl ether dinitro compound by use of the DES catalyst.

Conclusion

In summary, an effective, environmentally friendly and simple procedure has been presented for the synthesis of symmetric aryl ether dinitro compounds using DES (4Urea/ZnCl₂) both as catalyst and solvent. The DES can be recycled at least five times without obviously decreasing in the catalytic activity. This process has the advantages of high atomic economy, simple operation and recyclability.

Experimental

Materials:

¹H NMR (TMS: δ: 0.00 as an internal standard) spectra were recorded on a Bruker 500M spectrometer in DMSO-*d*₆ unless otherwise stated. All samples were analyzed by High Performance Liquid Chromatography P230 (Dalian Elite Corporation). Most of reagents were purchased from Shanghai Aladdin Biochemical Technology Co., Ltd. and were used without further purification unless otherwise specified.

Preparation of catalyst:

The DES was prepared according to the literature¹³. Urea (30 g, 0.5 mol) and ZnCl₂ (17.0 g, 0.125 mol) were heated with stirring at 150°C until a homogeneous colorless liquid was obtained. The DES formed (47 g, 100%) was cooled and used in reactions without any purification.

General procedure for preparation of symmetrical aryl ether dinitromonomer (3a–3l):

A mixture of aryl halide (2 mmol), dihydroxy compound (1 mmol) and 4Urea/ZnCl₂ (5% DES/NMP), as a catalyst, was stirred in a 100 ml round bottom flask with a condenser on the top. The mixture has been stirred for 2–3 h. It was slowly warmed up to 80°C. The progress of the reaction was monitored by HTLC (mobile phase = Methanol:Water = 80:20). After completing the reaction, deionized water (50 mL) were added and the mixture was stirred for 30 min. The crude product was extracted from organic phase by ethyl acetate and recrystallized from ethanol to obtain the pure product. The DES catalyst was recycled and used without further purification.

Selected spectral data:

1,2'-Bis(-nitrotrifluoromethylphenoxy)ethane (**3a**) white solid, (yield: 93%), m.p. 208–210°C, ¹H NMR (500 MHz, DMSO): δ 8.52 (dd, *J* 9.2, 2.5 Hz, 2H), 8.34 (d, *J* 2.3 Hz, 2H), 7.57 (d, *J* 9.3 Hz, 2H), 4.69 (s, 4H).

1,3'-Bis(4-nitrotrifluoromethylphenoxy)propane (**3b**) white solid, (yield: 89%), m.p. 220–225°C, ¹H NMR (500 MHz, DMSO): δ 8.49 (dd, *J* 9.2, 2.6 Hz, 2H), 8.32 (d, *J* 2.2 Hz, 2H), 7.49 (d, *J* 9.3 Hz, 2H), 4.51–4.32 (m, 4H), 2.29 (p, *J* 5.8 Hz, 2H).

1,4'-Bis(4-nitrotrifluoromethylphenoxy)butane (**3c**) white solid, (yield: 78%), m.p. 190–195°C, ¹H NMR (500 MHz, DMSO): δ 8.55 (dd, *J* 9.2, 2.4 Hz, 2H), 8.40 (d, *J* 2.2 Hz, 2H), 7.53 (d, *J* 9.3 Hz, 2H), 4.43 (d, *J* 20.9 Hz, 4H), 2.01 (d, *J* 13.1 Hz, 4H).

1,3'-Bis(4-nitrotrifluoromethylphenoxy)benzene (**3d**) white solid, (yield: 92%), m.p. 109–110°C (Lit.¹⁸ 108–112°C), ¹H NMR (500 MHz, DMSO): δ 8.55 (d, *J* 2.6 Hz, 2H), 8.52 (dd, *J* 9.1, 2.8 Hz, 2H), 7.70 (t, *J* 8.2 Hz, 1H), 7.34 (d, *J* 9.1 Hz, 2H), 7.32–7.28 (m, 1H), 7.26 (dd, *J* 8.2, 2.3 Hz, 2H).

1,4'-Bis(4-nitrotrifluoromethylphenoxy)benzene (**3e**) white solid, (yield: 86%), m.p. 220–221°C (Lit.¹⁹ 219–221°C), ¹H NMR (500 MHz, DMSO): δ 8.48 (s, 2H), 8.43 (d, *J* 9.2 Hz, 2H), 7.35 (s, 4H), 7.21 (d, *J* 9.2 Hz, 2H).

4,4'-Bis(4-nitrotrifluoromethylphenoxy)biphenyl (**3f**) white solid, (yield: 88%), m.p. 198–201°C (Lit.²⁰ 198–202°C), ¹H NMR (500 MHz, DMSO): δ 8.55 (d, *J* 2.6 Hz, 2H), 8.50 (dd, *J* 9.2, 2.6 Hz, 2H), 7.86 (d, *J* 8.6 Hz, 4H), 7.36 (d, *J* 8.6 Hz, 4H), 7.22 (d, *J* 9.2 Hz, 2H).

1,2'-Bis(4-nitrophenoxy)ethane (**3g**) beige solid, (yield: 85%), m.p. 143–145°C (Lit.²¹ 142–147°C), ¹H NMR (500 MHz, DMSO): δ 8.19 (d, *J* 9.2 Hz, 4H), 7.19 (d, *J* 9.2 Hz, 4H), 4.51 (s, 4H).

1,3'-Bis(4-nitrophenoxy)propane (**3h**) beige solid, (yield: 81%), m.p. 131–133°C (Lit.²² 129–132°C), ¹H NMR (500 MHz, DMSO): δ 8.18 (d, *J* 8.3 Hz, 4H), 7.15 (d, *J* 8.5 Hz, 4H), 4.28 (t, *J* 6.1 Hz, 4H), 2.24 (d, *J* 6.0 Hz, 2H).

1,4'-Bis(4-nitrophenoxy)butane (**3i**) yellow solid, (yield: 73%), m.p. 141–142°C (Lit.²³ 141–144°C), ¹H NMR (500 MHz, DMSO): δ 8.21 (d, *J* 9.1 Hz, 4H), 7.15 (d, *J* 9.1 Hz, 4H), 4.21 (s, 2H), 1.93 (s, 2H).

1,3'-Bis(4-nitrophenoxy)benzene (**3j**) yellow solid, (yield: 76%), m.p. 110–112°C (Lit.²⁴ 109–110°C), ¹H NMR (500 MHz, DMSO): δ 8.25 (d, *J* 9.2 Hz, 4H), 7.57 (t, *J* 8.1 Hz, 1H), 7.20 (d, *J* 9.2 Hz, 4H), 7.10 (d, *J* 2.0 Hz, 1H), 7.09–7.06 (m, 2H).

1,4'-Bis(4-nitrophenoxy)benzene (**3k**) yellow solid, (yield: 78%), m.p. 232–234°C (Lit.²⁵ 233–236°C), ¹H NMR (500 MHz,

DMSO): δ 8.24 (t, *J* 11.0 Hz, 4H), 7.30 (s, 4H), 7.18 (d, *J* 9.1 Hz, 4H).

4,4'-Bis(4-nitrophenoxy)biphenyl (**3I**) beige solid, (yield: 66%), m.p. 201–202°C, (Lit.^{26]} 200–202°C), ¹H NMR (500 MHz, DMSO): δ 8.22 (dd, *J* 17.3, 9.1 Hz, 4H), 7.78 (d, *J* 8.5 Hz, 4H), 7.25 (t, *J* 10.6 Hz, 4H), 7.15 (t, *J* 12.3 Hz, 4H).

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