Poll Res. 41 (2) : 718-726 (2022)

ISSN 0257–8050
DOI No.: http://doi.org/10.53550/PR.2022.v41i02.045

A REVIEW OF MODERN TECHNOLOGIES FOR REDUCTION OF DIOXINS AND FURANS FROM FLUE GAS EMISSIONS OF BIOMEDICAL WASTE INCINERATORS IN INDIAN PERSPECTIVE

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(Received 12 January, 2022; Accepted 24 February, 2022)

ABSTRACT

The present paper discusses the mechanism of formation of dioxins and furans and the technologies used to mitigate the formation of such chemical compounds during the combustion process. By reviewing the findings of several investigations, being conducted by several researchers for the last 30 years, it has been found that the formation of Dioxins and Furans can be considerably reduced by using the chemical compounds which function as catalysts. Such catalysts are vanadium pentaoxide V_2O_5, Tungsten Trioxide WO_3, and Titanium dioxide TIO_2. Besides, there are some sulphur compounds which inhibit the formation of dioxins and furans. Such compounds are Sulphur dioxide SO2 and ammonium sulphate $(NH_4)^2$ SO_4. There are some Thiourea chemicals which play an important role in inhibiting the formation of dioxins and furans. Such chemical compounds are Ethanolamine C_2H_7NO, Urea CH_4N_2O, Mono ethanolamine ($H_2NCH_2CH_2OH$) etc. But the greatest drawback in the aforesaid technologies is that, they require heavy capital cost as well as the high operating cost. As a solution to the problems, some German companies have developed the techniques for recovering HCl, Nacl and SO_2 which are marketable and thus, can make the process cost- effective. Another solution is to adopt a good combustion practice by modifying the design of combustion chambers (primary and secondary) as per guidelines issued by CPCB, India (2017). Lastly it may be considered to switch over to the latest alternative technology of Plasma Pyrolysis which provides a complete solution to the problem. The objective of the present study is to help the stakeholders in the process of making decisions and opting for the latest technologies for a pollution free environment.

KEY WORDS : Incineration, Dioxins, Furans, Flue gas, Pollution

INTRODUCTION

Incineration has become the most popular technology for disposal of bio-medical waste round the globe. However there are several problems associated with it which have made this technology the most controversial and disputed. The controversies and disputes have arisen due to the flue gas emissions which contain the harmful gases, such as oxides of nitrogen, sulphur, some trace elements i.e. nickel and mercury and the most toxic chemicals e.g. PCDDs and PCDFs. These compounds are the serious threat to the environment as well as to the human bodies, (Kaivosoza *et al.*, 2012). The super toxicity of PCDDs and PCDFs has attracted the attention of the whole world and as a result, Stockholm convention was called for discussing and restricting the use and production of all POPs including the PCDD and PCDF. The treaty was signed on May 23, 2001. India is also a signatory to the treaty and thus India is under the obligation to restrict the production of

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such harmful Chemicals. Intensive researches have been conducted round the globe for developing techniques for mitigating the generation of these toxic chemicals in the process of waste incineration. Several techniques have been developed for reduction of such toxic chemical compounds. Some modern researchers like Dvorak *et al.* (2010) Vermeulen *et al.* (2014) and Liu *et al.* (2015) have proved by their experimental studies that the reduction in the generation of such chemicals can be done by using the selective catalytic reduction as well as selective catalytic oxidation processes. Some other researchers like Xie *et al.* (2000) and Ke *et al.* (2010) have proved by their experiments that the production of PCDD and PCDF can be greatly reduced by injection of sulphur compounds. Some other researcher like Lin *et al.* (2015) have proved that reduction of PCDD and PCDF can be performed by using the nitrogen compounds (Thiourea). On the other hand there are many researchers like hung *et al.* (2014), Lu *et al.* (2013) and Fujimori *et al.* (2010) who have found by their experiments that the reduction of PCDD/F can be achieved by Good combustion practices and by using scrubber and bag filter in the end of pipe treatment system. The aforesaid technologies for reduction of PCDD/F by using catalysts and inhibitors have been studied by various researchers and found to be effective in reducing the PCDD/F up to 99% but the greatest drawback is that, such technologies require heavy capital cost as well as the operating costs. In order to find a cost-effective solution some German companies have succeeded in developing a recovery process of HCL (Hydrochloric Acid), Nacl and SO2 which are marketable. By adopting these techniques the problem of high cost may be solved. (Hartenstein HU-1993).

CPCB India, 2017 has issued guidelines for improving the design of the combustion chamber and also for optimizing the operation procedure. Although the change over to the new improved design would require some extra investment, yet it would be the better solution. CPCB India, 2017, has also issued guidelines for installation and operation of the latest innovative technology of "plasma pyrolysis" which is capable of reducing the emissions of all pollutants including the PCDD and PCDF far below the legal emission limits. Many researchers have been conducting their investigations to find the pathways to formation of Dioxins and furans and evolving various technologies to mitigate the formation of such toxic substances. Muhammad Zubair and Amina Adrees (2019) have described the process of formation of dioxins and furans. According to the authors these compounds are formed by two mechanisms. One mechanism is the formation from precursors and the other mechanism is the formation through the de-Novo synthesis (which means the formation of complex molecules from the simple molecules) Mukherjee *et al.* (2015) have studied the pathways to the formation of dioxins and furans and presented the process of De-Novo Synthesis for formation of PCDD/PCDF. Chang *et al.* (2011) have presented the report on the hazardous effect of PCDD/F on health of human beings as well as animals. Choi and Lee (2007) have presented the report on concentration of PCDD/PCDF at the inlet and outlet of wet scrubber in incinerator of Korea. Dvorak *et al.* (2010) have presented the report on the investigations on the technique of removal of Dioxins and furans for protecting the environment. Fujimori *et al.* (2010) have presented the result of the investigations on the reduction of POPs (Persistent Organic Pollutants). Hatanaka *et al.* (2004) have discussed the role of copper chloride in the formation of dioxins and furans. Hung *et al.* (2014) have presented the result of reduction process by using the catalysts. Husinger *et al.* (2007) have discussed the several processes for reduction in formation of Dioxins and Furans. Kaivosoja *et al.* (2012) have discussed the role of catalysts in the reduction of PCDD/F. Ke *et al.* (2010) have discussed the inhibition of de-novo synthesis in formation of dioxins and furans. Korell *et al.* (2009) have presented the report on the simultaneous removal of mercury, dioxins and furans & fine particles from flue gas. Lin *et al.* (2015) have discussed the unintentional generation of dioxins and furans, during the process of waste incineration. Lu *et al.* (2013) have discussed the use of activated carbon for treatment of flue gases municipal solid waste incinerator. Vermeulen *et al.* (2014) have observed that urea can efficiently decompose the dioxins reducing it to about 90%. Wu *et al.* (2012) have discussed the reduction of PCDD/F by adding sulphur compounds.

MATERIALS AND METHODS

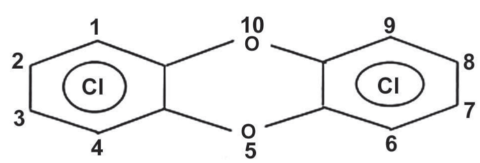
The present study report is based on the review of the scientific literature on formation as well as reduction of dioxins and furans available globally up till now. The study has been mainly focused on the formation of such pollutants during the combustion process in waste incinerators and also on the process adopted for mitigating the generation of Dioxins and furans. Sufficient literature has been reviewed which exhibit that by improving the design of the combustion chamber for ensuring the complete combustion the formation of PCDD/F can be mitigated. (Lu *et al.* (2013)]. The guidelines and suggestions issued by CPCB India (2017) have also been reviewed where improving the design for ensuring complete combustion has been suggested which would mitigate the pollutants including PCDD/F.CPCB India (2017) has also issued guidelines for switching over to the latest alternative technology of plasma pyrolysis which have been reviewed (www.cpcb.nic.in).

Description of PCDDs and PCDFs

Dioxins and Furans are the names of two groups of persistent organic pollutants (POPs). In literature dioxin is expressed in short form as PCDD and the furan is expressed as PCDF. They are jointly expressed as PCDD/PCDF and in further short form they are expressed as PCDD/F. All of these compounds are made up of hydrogen, carbon, chlorine and oxygen atoms. These are differentiated from one another on the basis of number of atoms of Chlorine as well as the positions of these chlorine atoms in the molecular structure.

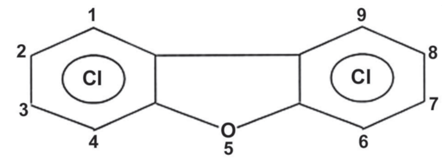
The molecular structure of PCDDs is presented below –

1) The above molecular structure shows that dioxins have oxygenated tri-cyclic molecules.



The molecular structure of PCDDs & PCDFs

2) Two benzene rings are bound by Oxygen atoms at 5th and 10th position.
3) The numerical figures show the positions where



(Adapted from www.cpcb.nic.in)

the chlorine atoms may be substituted in different combinations.
4) The number of chlorine atoms varies from 1 to 8.

The structure of furans is given below –

1) In Furans one oxygen atom at the 5th position binds two benzene rings to become a tricycle molecule.
2) The number of chlorine atoms may vary from 1 to 8.
3) The numerical figures show the positions where the chlorine atoms may be substituted in different combinations.

THE INFRASTRUCTURE FACILITIES FOR MEASUREMENT OF PCDD/F IN INDIA

The dioxins and furans have been successfully characterized and analyzed by using the following instruments.

(1) Infra-red spectroscopy (IR Spectroscopy).
(2) Ultraviolet spectroscopy (UV-Spectroscopy).
(3) High Resolution Gas Chromatography (HRGC).
(4) Nuclear Magnetic Resonance (NMR) techniques.
(5) High Resolution mass spectroscopy (HRMS).

CPCB has observed that the infrastructure facilities for measurement of PCDDs and PCDFs are too little in the country. (www.cpcb.nic.in, Parivesh Newsletter, Dec., 2004)

When the same number of chlorine atoms exist in dioxins and furans they are termed as homologue group. The following table shows the number of isomers of dioxins and furans.

The above chart shows that there are 210 different structures (75+135) and they are called congeners. (Cpcb.nic.in Parivesh News Letter, Dec., 2004).

Unintentional Generation

Dioxins and Furans are not created for any specific

Table 1. Isomers of Dioxins (PCDD) and Furan (PCDF)

Number of Chlorine Atoms →	1	2	3	4	5	6	7	8		
Number of Dioxins Isomers →	2	10	14	22	14	10	2	1	**Total**	**75**
Number of Furans Isomers →	4	16	28	38	28	16	4	1	**Total**	**135**

purpose; however these are generated unintentionally during the process of combustion.

All the aforesaid congeners of dioxins and furans are toxic but the magnitude of toxicity widely varies. Such variations are evaluated by toxicity equivalency factors.

The Toxicity Equivalency Factors

There are several systems to measure the toxicity, such as international toxic equivalency for dioxins and furans which are represented as I-TEQ. WHO has also evolved a system represented as WHO-TEQ which is also used all over the world? The following table shows the measurement of I-TEF.

Process of generation of Dioxins and Furans

PCDD/F is formed during the process of combustion. These toxic chemicals generate due to the disintegration of organic material caused by high temperature together with the chlorinated compounds and transition metals. It has been found that such chemicals are formed when the temperature is between the ranges of 200 ^{o}C to 400 ^{o}C.
(www.cpcb.delhi.nic.in)

The mechanism of generation of Dioxins and Furans

The First Mechanism

Some researchers like Tosine *et al.* (1985) have conducted analysis of the raw waste before their incineration and found that some trace amounts of the PCDD/F were existed in the components of the raw materials. When such raw wastes are fed into the incineration, then, due to incomplete combustion, the PCDD/F is emitted.

The Second Mechanism

The second mechanism is that the PCDD/F is formed from the precursor compounds. The precursor compounds are chlorophenol (C6H5ClO), Chlorobenzene (C6H5Cl) or polycyclic aromatic hydrocarbons, such as, Benzo (A) Fluorene (C17H12), Benzo (A) Pyrene (C20H12) etc. and the chlorine donor compounds such as PVC (C2H3Cl) n, and hydrogen chloride. The interaction between the aforesaid compounds is promoted by a transition metal catalyst.

(Dickson and Karasek (1987)

The Third Mechanism

The third mechanism is de-novo synthesis. Which means "a new" or "from the beginning". It means that complex molecules are formed from simple molecules by the process of synthesis. It functions as a promoter in the generation of PCDD/F by oxidation of carbon particulates. The process is catalyzed by the presence of transition metal. This process is supposed to happen, not in the primary chamber but in the post-combustion chamber, where the temperature is cooled down up to the level favorable of generation of PCDD/F. The most appropriate temperature for this process is the range from 500 ^{o}C to 200 ^{o}C.

Stieglitz *et al.* (1987) have found that the temperature of 3000C have been found to be most favorable for the de novo process.

Toxicity of Congeners of Dioxins	I-TEF	Toxicity of Congeners of Furans	I-TEF
Congeners having 4 chlorine atoms at positions 2, 3, 7, 8	1.0	Congeners having 4 chlorine atoms at positions 2, 3, 7, 8	0.1
Congeners having 5 chlorine atoms at positions 1, 2, 3, 7, 8	0.5	Congeners having 5 chlorine atoms	
		at positions 1, 2, 3, 7, 8	0.05
		at positions 2, 3, 4, 7, 8	0.5
Congeners having 6 chlorine atoms		Congeners having 6 chlorine atoms	
at positions 1, 2, 3, 7, 8, 9	0.1	at positions 2, 3, 4, 6, 7, 8	0.1
at positions 1, 2, 3, 6, 7, 8	0.1	at positions 1, 2, 3, 7, 8, 9	0.1
at positions 1, 2, 3, 4, 7, 8	0.1	at positions 1, 2, 3, 6, 7, 8	0.1
		at positions 1, 2, 3, 4, 7, 8	0.1
Congeners having 7 chlorine atoms at positions 1, 2, 3, 4, 6, 7, 8	0.01	Congeners having 7 chlorine atoms	
		at positions 1, 2, 3, 4, 7, 8, 9	0.01
		at positions 1, 2, 3, 4, 6, 7, 8	0.01
Congeners having 8 chlorine atoms	0.001	Congeners having 8 chlorine atoms	0.001

NATO-CCMS, 1998(30)

The Process of Reduction of PCDD/PCDF

The process of reduction of Dioxins and Furans starts from the preparation of raw waste (feed stock) and continue, covering the processes in primary combustion chamber, post combustion chamber, air pollution control devices and also covering the guidelines issued by CPCB (India) for reducing all pollutants including PCDD/F.

In this direction the role of catalysts, inhibitors and all other steps to be taken thereafter, may be described stepwise:

(1) Role of Catalysts in reduction of Dioxins and Furans

(i) Lu *et al.* (2013) have reported that the generation of PCDD/F may be controlled by using NH_3-SCR Catalyst.

The catalyst accelerates the speed of reaction and prevents the formation of PCDD/F.

(ii) The other chemical compounds used as catalysts are Vanadium Pentoxide V_2O_5 and tungsten trioxide WO_3 and Titanium dioxide TIO_2. These catalysts have been found to effectively decompose the dioxins and furans without any adverse effect on the operating condition of the combustion chamber (Chang *et al.,* 2009).

The reactions result in the formation of non-toxic substances like H_2O, CO_2 and HCl and the generation of dioxins and furans is reduced to a greater extent (Dvorak *et al.,* 2010).

(2) Role of inhibitor chemicals in reduction of dioxins and furans.

(i) Wu *et al.* (2012) have studied and found that sulfur compounds SO_2 and ammonium sulphate $(NH_4)^2SO_4$ prevent the generation of PCDD/Fs.

(ii) Samaras *et al.* (2000) have reported that organic sulfur has been proved to be active inhibitors and the reduction capability is as high as 98%.

(3) Role of Thiourea in reduction of PCDD/F.

(i) Lin *et al.* (2015)have found that some chemical compounds that contain nitrogen, such as, Ethanolamine C_2H_7NO, Mono-ethanolamine $H_2NCH_2CH_2OH$, urea CH_4N_2O, Ammonia NH_3, Triethanolamine $C_4H_{15}NO_3$ and Di-methylamine $(CH_3)^2NH$ are very suitable inhibitors.

(4) Role of good combustion practice.

(i) Weber *et al.* (2002) Suggested that the generation of Dioxins and Furans can be controlled by adopting good combustion practice which means that the complete combustion must be accomplished in the combustion chamber. The author has suggested modifying the design of the equipment's in such a way that the complete combustion may be achieved.

(ii) Hung *et al.* (2014) have studied the optimum condition for complete combustion and found that the 3 Ts parameters (Temperature, residence time and Turbulence) should be maintained at the optimum level. Accordingly for achieving complete combustion the appropriate level should be:

1. **Temperature –** A range of temperature between 900 °C and 1000 °C.
2. **Residence Time -**The residence time should be more than 1 second.
3. **Turbulence –** The appropriate level of turbulence should be in terms of Reynolds number, the turbulence should be greater than the Reynolds number 50,000, which is preferred to create sufficient turbulence in the combustion chamber.

(5) Role of High Temperature of 1100 °C and fast cooling in secondary chamber:

(i) In the secondary chamber the temperature should be raised to 1100 °C for a minimum residence time of two seconds, so that the partly burned materials eg. Soot, smoke and volatiles in flue gas including PCDD/F may be combusted completely and also be decomposed and destroyed (CPCB India, 2017 www.cpcb.nic.in).

ii) Rapid Cooling – In the final stage the post combustion temperature should be cooled down rapidly, so that, the temperature makes come down below 200 °C. The formation of PCDD/F is mitigated at the temperature below 200° C (Korell *et al.,* 2009).

(6) Guidelines issued by CPCB India for reduction of pollutants including PCDD/F

CPCB India (2017) had issued guidelines for biomedical waste incinerators in India. In the guidelines there are suggestions which are technically sound for modifying the design for ensuring complete combustion, which would greatly reduce the emissions of pollutants including the PCDD/F. The guidelines are aimed at improving the design of the primary and secondary chambers and for ensuring the complete combustion. Some of the important design criteria to be adopted for improving the performances are as follows:

(i) The ratio between the volume of the waste (feed-stock) and the volume of the combustion chamber

The ratio between the volume of the waste (feed-stock) and the volume of the combustion chamber should be in the proportion of 1 to 5 i.e. the volume of the combustion chamber should be five times more than the volume of the waste.

(ii) Rounded corners from the inside

The corners in the sides and the top portion of the combustion chamber should be rounded from the inside, so that, the formation of dead zones, are cold pockets may be avoided.

(iii) The measurement devices to be fitted in the combustion chamber

For the purpose of the measurement of the rate of flow, the supply of air and the pressure gauges etc. should be suitably attached in the primary and the secondary chamber.

(iv) The air pressure in the combustion chamber to be kept low in comparison to the ambient air

In order to prevent the possibility of any leakage of gases from the combustion chamber, the air pressure inside the combustion chamber should be maintained at lower level in comparison to the pressure of the ambient air of the room.

(v) The inner wall of the combustion chamber should be insulated with heat- resistant fire bricks

The inner side of the combustion chamber should be insulated with heat-resistant fire bricks/refractory system which may be able to withstand the temperature of at least 1000 ^{o}C

(vi) The insulation of the inner wall in the secondary chamber

The inner wall of the secondary chamber should be insulated with refractory system/fire bricks which may sustain the minimum temperature of 1200 ^{o}C.

(vii) The Provision of separate burners for both chambers

In order to achieve the required temperature in both chambers, there should be two burners, one for the primary and the other for the secondary chamber, so that, the desired temperature may be obtained.

(viii) The direction of the flame should be pointed towards the center

The flame arising from the burner in the primary chamber should be pointed towards the direction of the center where the waste is to be combusted.

(ix) The direction of the flame in the secondary chamber

The flame arising from the burner in the secondary chamber should be in the appropriate direction, so that the flue gases containing volatiles and soot etc. may be completely combusted.

(x) Provision of View Ports

There should be provision for view ports for observing the visual condition of the combustion process and adjust the combustion process accordingly maintaining the parameters of temperature, time and turbulence. The view port should be equipped with glass which may withstand the high temperature. There should be a metal covering for the glass. The view port should be of projected type.

(xi) To be equipped with computer recording devices:

The incinerator should be equipped with computer recording system, so that, all relevant data e.g. Date, time, batch number, temperature in both the chambers and also at the stack exit point may be recorded.

(xii) Maintaining suitable high water pressure

A suitable high water pressure must be maintained for rapid cooling of the flue gas. It is necessary that the temperature of the flue gas should be cooled down to below 200 ^{o}C, so that the process of De Novo synthesis may be avoided for preventing the formation of PCDD/F during the process of cooling down.

(xiii) Attaching the provision of Air Pollution Control Device in the Incinerator

The devices for pollution control (APCD) must be attached with the waste incineration system which should be able to comply with the prescribed standard of emission limits of all air pollutants including PCDD/F. CPCB India (2017) has advised to use the adsorption system by using activated carbon etc. and subsequently using fabric filter so that the emissions may be completely controlled.

(xiv) The Provision of Alarm System

The incinerator system should be equipped with an alarm system which may alert in cases of any type of failure in the system, so that remedial measures may be taken immediately.

With the aforesaid improvements in the design and operations, it can be confidently expected that due to complete combustion of the waste the emissions of all pollutants including PCDD/F would be mitigated and would be able to meet the prescribed limits.

(7) Changing over to the new alternative technology of Plasma Pyrolysis for reduction of PCDD/F

The technology of plasma pyrolysis is an innovative technology. In this technology high temperature is produced by using plasma torch and then the reactions take place and wastes are fully destroyed. This technology is well demonstrated and well proven. This technology is being used in most of the developed countries. In India this technology has been developed by FCIPT Ahmedabad who has transferred the technology to Ahmedabad based M/s Bhagwati Pyrotech Limited. The manufacturers have proved and claimed that by using this technology they can meet the emission limits of all pollutants including PCDD/F, set by US-EPA and the CPCB (India).

(Nema *et al.* (2002)].

(8) Reduction of Dioxins of Furans through the Air Pollution Control Devices:

The following steps are to be taken for reducing pollutants including dioxins and furans:

(i) Injection of Adsorbent

In this method the powdered activated carbon is injected. The activated carbon has been proved to be a very good adsorbent. The activated carbon adsorbs the PCDD/F while they are on their way for moving out of the stack. Here in this process the PCDD/F together with other particulate matters of the flue gas are separated through the bag filters.

(ii) Activated Carbon Reactor (A.C.R.)

Activated carbon reactor is another solution for reducing dioxins and furans. In this method the granular (HOC) functions as adsorbent. This method is more efficient because the size of the particles of HOC is comparatively larger.

(iii) Entrained Flow Reactor

The entrained flow reaction is the process for the pre-cleaning system which consists of dry or semi-dry or wet scrubbing system. With this system the PCDD/F together with some other pollutants such as SOx, HCl, HF and Hg are removed to value below the legal emission limits. In this process the adsorbent is usually applied. The systems works with following provisions.

- The system of injection of adsorbent.
- The provision of fabric filters for removing the adsorbent which has been used.
- Provision of a system of recirculation of adsorbent.
- Provision of a system for storing the used adsorbent.

RESULTS AND DISCUSSION

The aforesaid studies show that the generation of PCDD/F can be reduced by using several types of catalysts and the inhibiters. These processes require heavy investment. In order to lessen the burden of high investment some German scientists have succeeded in developing the technology to recover some marketable items from the waste incineration processes such as HCl, NaCl, and $CuCl_2$ which may give some economic relief. But there are two problems with these technologies which make them unsuitable for developing countries like India; firstly these technologies require heavy capital investment and secondly these processes are highly complicated which would increase even the operating cost and even a minor defect in any part of the machinery may disrupt the working system resulting in the shut-down of the plants. In this background the other options such as “Good Combustion practice”. CPCB India (2017) has issued guidelines and suggestions for improvement in the design of the combustion chamber and several improvements in the operative system which should be adopted. It also seems to be necessary that in due course of time the incineration system should switch over to Plasma Pyrolysis Technology for perfect solution.

CONCLUSION

- The design of the incineration plant should be modified in such a way that the complete combustion of the combustible waste may be ensured by maintaining a proper adjustment in temperature, turbulence and time.
- Proper segregation of the waste must be done because PVC contains chlorine which generates the Dioxins and Furans.
- The waste should be dried to minimize the

moisture. Higher percentage of moisture generates more Dioxins and Furans.

- The supply of air should be optimized to ensure complete combustion of the waste.
- The temperature must be raised to $1100\ ^{o}C$ for at least 2 seconds in the post combustion chamber for destroying the dioxins and furans (www.cpcb.nic.in.dioxins and furans).

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