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Solvent-less Oxidation of Aromatic Alcohols Using CrO_3/Al_2O_3 under Ultrasonic Irradiation

Maulidan Firdaus [a,*], Triana Kusumaningsih [a], Ichsan Arifagama [a],
Zuhdi Alqowamul Amin [a]

[a] Department of Chemistry, Faculty of Mathematics and Natural Sciences, Universitas Sebelas Maret, Surakarta, Indonesia

*Corresponding author: maulidan@mipa.uns.ac.id

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Abstract

Alcohol oxidation plays an essential contribution to the chemical industry. Innovative green techniques, such as ultrasound irradiation, could be economically remarkable by enhancing reaction yield. In this research, the design and improvement of a new green ultrasound-assisted oxidation of alcohols procedure using CrO_3 supported by Al_2O_3 with the addition of a small amount of *t*- butanol were reported. The oxidation of alcohols was also done without ultrasound irradiation to study the sonochemical effect. Based on FTIR and GC/MS analyses, the alcohols were effectively oxidized into their corresponding aldehydes in satisfactory yields (74–93%). The yield of the obtained aldehydes was increased by applying the ultrasonic irradiation technique, and no over-oxidation products were found. Overall, the innovative procedure offers several benefits, such as being easy to use, environmentally friendly, capable of improving yields, and having shorter oxidation times.

1. Introduction

Oxidizing alcohols to aldehydes is the essential reaction in organic and industrial chemistry [1, 2]. It is well-known that aldehydes are crucial intermediates for preparing fragrances, perfumes, drugs, dyes, and a wide range of fine chemicals [3, 4]. Conversion of alcohol to aldehyde by oxidation is generally performed either in heterogeneous or homogeneous systems. The success of the transformation is highly influenced by reaction conditions, the mole ratio of reactants, temperature, reaction time, and oxidizing agents [5]. This reaction is usually achieved by employing numerous stoichiometric oxidants, including chromium-based reagents, manganese salts, or mild oxidants such as Dess-Martin periodinane, in a reasonably excellent yield [6]. Chromium salt has been widely used among these oxidants because this reagent is a strong oxidizing agent [7, 8]. Unfortunately, these reagents have several drawbacks, such as high toxicity and environmental pollution [9].

The utilization of toxic Cr(VI) in the oxidation of alcohols cause severe ecological problem associated with the utilization of enormous volumes of halogen-based or aromatic-based solvents that involves a considerable supply chain effect. Furthermore, heavy metals and harmful solvent waste treatments are overpriced and must be appropriately treated. The possibility of contamination from the harmful chromium-based oxidants prevents further use of the reaction products for the proper synthesis of medicines, cosmetics, or food additives [8, 10]. Attempts have been made to find an appropriate technique for the oxidation of alcohol using Cr(VI) oxidants through simple modifications with various solid supports such as silica gel, anion exchange resins, zeolites, alumina (Al_2O_3), or zirconium dioxide [8, 9, 11]. Using Cr(VI) impregnated with solid supports could provide an attractive alternative in terms of selectivity and processing convenience but also accelerate the reactions and reduce harmful levels by absorbing hazardous by-products [10, 11].

When designing a new method for alcohol oxidations, developing effective and environmentally friendly procedures according to the principles of green chemistry must be attentively well-thought-out [12, 13]. Thus, one approach to applying green chemistry principles is by carrying out the oxidation either under solvent-free conditions or utilizing a small quantity of solvent, known as solvent-less oxidation. For sustainability concerns and safety awareness, it would be essential to reduce the removal of harmful by-products and decrease the practice of using flammable organic solvents. Alcohol oxidation under solvent-less conditions can be performed by numerous methods such as ultrasound irradiation, conventional stirrer or shaker, microwave-assisted reaction, and grinding [14]. The oxidation of alcohols using CrO_3 in a supporting solid by grinding with an agate mortar and pestle has been reported [8, 11]. Although this method is efficient and effective, it has several limitations, such as a small aggregate of samples might possibly be lost, which results in poor yields due to the difficulty of recovering the sample; since the grinding is performed by hand, it is difficult to quantify the required energy. Depending on the applied power, strong and weak grinding might affect the reaction time and the achieved product [15].

On the other hand, as an environmentally benign technique, ultrasound-assisted oxidation of alcohols has attracted great attention from organic chemists since this procedure is more appropriate, fast, and simple to perform, resulting in higher yields and reduced reaction times [16]. The use of sonochemistry in the oxidation of alcohols has been reported to only mention recent examples: under ultrasonic irradiation, iron sulfate and hydrogen peroxide increased the yield of benzyl alcohol oxidation by +45% compared to the conventional conditions [2], and ultrasonic-assisted oxidation procedure by employing hybrid nanostructure of multiwall carbon nanotubes and titanium dioxide has been reported in converting alcohol into corresponding aldehydes in high yield and shorter reaction time [16], and sonoxidation ZnO coating in a microflow based photo reactor has been effectively described in the selective transformation of benzyl alcohol into benzaldehyde [17].

Although ultrasonic-assisted oxidation with various oxidants has been widely reported, based on a literature survey, the use of CrO_3/Al_2O_3 coupled with sonochemistry for the oxidation of alcohol has not been reported. In connection with our research endeavors using an environmentally benign protocol, herein we report rapid and efficient ultrasonic-assisted oxidation of alcohols with CrO_3 supported on alumina under solvent-less conditions.

2. Materials and Methods

2.1. Materials

Commercial high purity products purchased from E- Merck were used in this study, such as aluminum oxide (Al_2O_3), chromium trioxide (CrO_3), *t*-butanol, n-hexane, ethyl acetate, CH_2Cl_2, anhydrous Na_2SO_4, $KMnO_4$, benzyl alcohol, furfuryl alcohol, *p*-anisyl alcohol, and silica gel.

2.2. Instrumentation

Ultrasound-assisted oxidation of alcohols was conducted using an ultrasonic device from Branson 1510 ultrasonic cleaner operational at 40 kHz of power 90 Watt in constant frequency. Column chromatography was performed on SiO_2 using hexane:ethyl acetate = 7:3 (v/v) as the mobile phase. Silica gel 60 GF_{254} TLC-plate from Merck was used as thin layer chromatography (TLC). The spot on the TLC plate was visualized using permanganate solution. Fourier Transform-Infrared (FTIR) analysis was done using a Shimadzu FTIR Prestige 21 spectrophotometer at r.t. in the range of 4000–500 cm^{-1}. GC/MS analysis (EI) was performed on a Shimadzu QP2010S spectrometer using FID over 30 m RTX-5MS as the column. The m/z value was scanned from 50 to 650 at a rate of 1.00 $scan.s^{-1}$. The column oven temperature was set to 70°C, and the injector column temperature was programmed at 300°C. Helium was employed as the carrier gas, and the measurement was performed in a split–split mode.

2.3. General procedure for alcohol oxidation

2.3.1. Solvent-less oxidation alcohols using the stirring technique

A mixture of 1 g (10 mmol) fine powder CrO_3 and 10 g (98 mmol) Al_2O_3 was ground using a mortar and pestle until smooth for approximately 1 minute. Corresponding alcohol (5 mmol) and 0.5 mL *t*-butanol were added to the mixture. The mixture was then magnetically stirred at r.t. The progress of the reaction was detected by TLC using n-hexane:ethyl acetate = 9:1 (v/v) as the mobile phase until no reactant spot was detected. The reaction mixture was then added with 10 mL of distilled water and extracted using 2 × 20 mL of dichloromethane. The organic layer was dried with anhydrous Na_2SO_4 and decanted for further evaporation. Purification of the product was then performed using column chromatography. The resulting aldehyde was further characterized using FTIR and GC/MS.

2.3.2. Ultrasound-assisted oxidation of alcohols

A mixture of 1 g (10 mmol) CrO_3 and 10 g (98 mmol) Al_2O_3 was ground until smooth using a mortar and pestle. The CrO_3/Al_2O_3 mixture was put into a vial, and 5 mmol of corresponding alcohol and 0.5 mL *t*-butanol were added. The mixture was then sonicated at ambient temperature until TLC showed complete disappearance of the alcohol. Distilled water (10 mL) was added and extracted using 2 × 20 mL dichloromethane. The organic layer was then collected, dried over anhydrous Na_2SO_4, and evaporated. Product purification using column chromatography was then performed. The resulting aldehyde was then further characterized using FTIR and GC/MS.

Products:

Benzaldehyde; clear liquid. $FTIR_{\nu max}$ (neat) cm^{-1}: 3066 (C_{sp^2}–H), 1599, 1582, 1495, 1453, 745, and 708 (aromatic), 2851 and 2733 (C–H aldehyde), 1687 (C=O). GC/MS electron ionization (m/z): 106 (M^+), 77, 51.

Furan-2-carbaldehyde; light yellow liquid. $FTIR_{\nu max}$ (neat) cm^{-1}: 3132 (C_{sp^2}–H), 2850 and 2811 (C–H aldehyde), 1672 (C=O), 1020 (C–O). GC/MS electron ionization (m/z): 96 (M^+), 95 (M-1), 67, 50. The FTIR and GC/MS data are in accordance with the reported reference by Firdaus *et al.* [8].

p-anisaldehyde; clear liquid. FTIR$_{\nu max}$ (neat) cm^{-1}: 3000–3010 (C$_{sp^2}$–H), 2840 and 2740 (C–H aldehyde), 1681 (C=O), 1600, 1577, 1511, 833 (aromatic), 1030 (C–O). GC/MS electron ionization (m/z): 136 (M$^+$), 135 (M-1), 107, 92, 77, 65, 51, 39. The spectral data is in line with the previous publication [8].

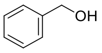
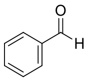
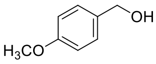
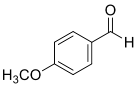
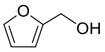
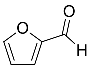
3. Results and Discussion

In this study, several alcohols, such as benzyl alcohol, *p*-anisyl alcohol (4-methoxybenzyl alcohol), and furfuryl alcohol (furan-2-yl)methanol, were chosen as starting materials for the solvent-less oxidation alcohol. CrO_3 was selected as the oxidant since this reagent is a powerful oxidizing reagent capable of oxidizing the alcohol into desired aldehydes. Even though chromium oxide can oxidize alcohols into the corresponding aldehydes in a reasonable yield, the utilization of this oxidant in a large-scale condition is impracticable since it usually requires a long-time reaction and generates hazardous polluting materials. Therefore, to reduce the toxic Cr(VI), Al_2O_3 was added as the solid support. Attaching chromium trioxide to the Al_2O_3 supporting solid materials might prevent the aldehyde contamination and avoid the formation of carboxylic acid due to over oxidation [11]. The Al_2O_3 solid support might act as a transporter to permit CrO_3 to be well embedded, possibly by absorbing and binding the side product of chromic acid, thus decreasing the toxicity level of chromium species [3, 8].

Intending to study the sonochemical effect, we perform the alcohol oxidations under the stirring technique compared to ultrasonic irradiation. Initially, under solvent-free conditions, benzyl alcohol was chosen as starting alcohol for the oxidation using CrO_3/Al_2O_3 at room temperature. During this study, both techniques were checked by TLC using hexane:ethyl acetate = 7:3 (v/v) as the eluent to monitor the progress of the oxidation. In the case of ultrasound-assisted oxidation of benzyl alcohol, TLC analysis showed complete disappearance of the alcohol after 2 hours of reaction time. This result was relatively faster than the stirring technique, which required a reaction time of 3 hours. Regrettably, under solvent-free conditions, the oxidation only afforded benzaldehyde in 60% yield using ultrasonication and 53% yield under the stirring technique.

The oxidations were then conducted under solvent-free conditions to discover an appropriate condition and shorten the reaction time. It was described that adding a small amount of *t*-butanol might increase the reaction yield and shorter the oxidation time [8, 11]. Taking this information, we started to study the oxidation of alcohols using CrO_3/Al_2O_3 under stirring and ultrasonic irradiation in a small quantity of *t*-butanol. In the case of benzyl alcohol oxidation (entry 1, Table 1), the ultrasound-assisted method was completed in 15 minutes, which was faster than the stirring technique of 20 minutes. It was much quicker than without the addition of *t*-butanol. In terms of yield, the ultrasonic irradiation methodology gave a better yield of 92% than the stirring technique, which was 84%.

Table 1. Solvent-less oxidation of alcohols using CrO_3/Al_2O_3

Entry	Reactant	Product	Stirring Technique		Ultrasound Irradiation	
			Time (min.)	Yield (%)	Time (min.)	Yield (%)
1	 Benzyl alcohol	 Benzaldehyde	20	84	15	92
2	 *p*-anisyl alcohol	 *p*-anisaldehyde	25	74	15	90
3	 Furfuryl alcohol	 furfuraldehyde	20	83	15	93

Numerous alcohol oxidations can be performed in greater yields, reducing reaction time in milder conditions *via* ultrasound technique. A reason for accelerating the oxidation using the ultrasonic technique can be described through acoustic cavitation and cage phenomenon that execute the extreme temperature and pressure in just a few seconds, generating a distinctive atmosphere or high energy micro-reactions which assist molecular fragmentation in the collapsing cavities [18]. Nucleation, bubble evolution, and implosive breakdown are the three consecutive processes of the phenomenon known as acoustic cavitation. Ultrasonic irradiation propagates in a liquid that aggravates compression and dilatation of the reaction. Once the compressed medium attains a pressure under the saturated vapor pressure, a micro-bubble of vapors is generated. Subsequent compression and decompression phases create its evolution until its extents a critical size and break down [2]. Thus, this technique increases the reaction rate compared to the conventional procedure, which usually requires longer reaction times and expensive chemicals [16, 19, 20].

Characterization of the resulting solvent-less oxidation of benzyl alcohols using CrO_3/Al_2O_3 was done using FTIR spectrophotometry. The results obtained from the FTIR analysis (Figure 1) showed the appearance of a strong absorption at 1687 cm^{-1} together with the absorption at 2851 cm^{-1} and 2733 cm^{-1}, which indicated the occurrence of the aldehyde group. In addition, the results of the FTIR analysis did not show a broad peak at the absorption around 3700–3000 cm^{-1}, which is typical of alcohol compounds. Meanwhile, the presence of an aromatic group was indicated by the appearance of absorption at 3066 cm^{-1}, indicating the existence of aromatic C-H absorption. It was strengthened again by the presence of absorption peaks which were quite sharp in the area of 1599, 1582, 1495, and 1453 cm^{-1}, which showed the presence of the aromatic C=C. The appearance of sharp peaks at 745 and 708 cm^{-1} indicated the aromatic monosubstitution's presence. The results obtained are very supportive of the functional groups possessed by benzaldehyde.Further analysis of the ultrasonic-assisted oxidation of benzyl alcohol product was performed by a GC/MS spectrometer. Based on GC analysis, one chromatogram peak (Figure 2) with a retention time of 4.865 minutes was obtained in which the MS spectrum (Figure 3) had three main fragmentations at m/z 106, 77, and 51 with 77 as the base peak. The MS displayed the M^+ at m/z = 106, agreeing with benzaldehyde's molecular weight. This confirmed the success of the oxidation. Also, this mass spectrum is similar to the standard mass spectra of benzaldehyde, with a similarity index (SI) of 98. The identification of the benzaldehyde was made based on the documented MS matched with the standard MS from the Wiley7 and National Institute of Standards and Technology (NIST) library delivered by the GC/MS software system [21]. Acquired from the FTIR and GC/MS analyses, the solvent-less ultrasonic-assisted oxidation of benzyl alcohol CrO_3/Al_2O_3 produced benzaldehyde in 92% yield.

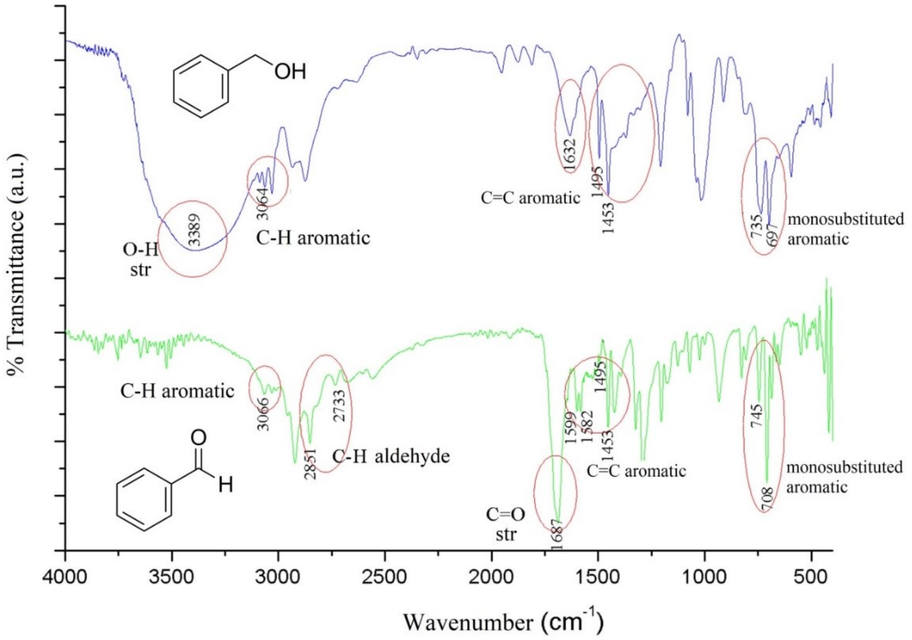


Figure 1. FTIR Spectra of benzyl alcohol (top) and benzaldehyde (bottom)

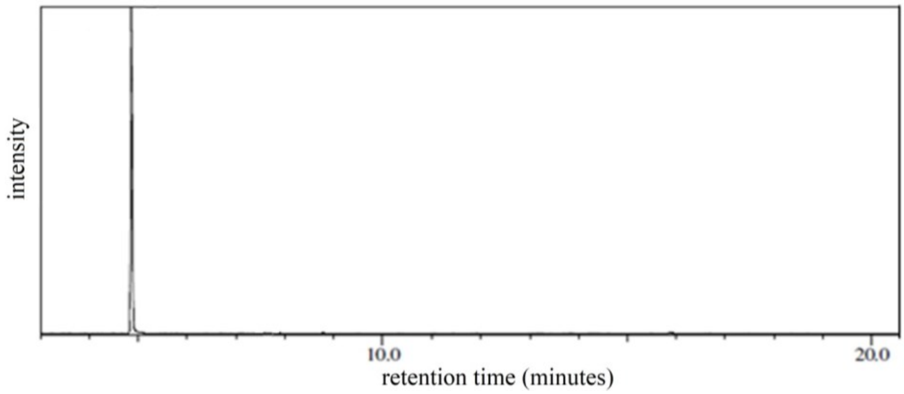
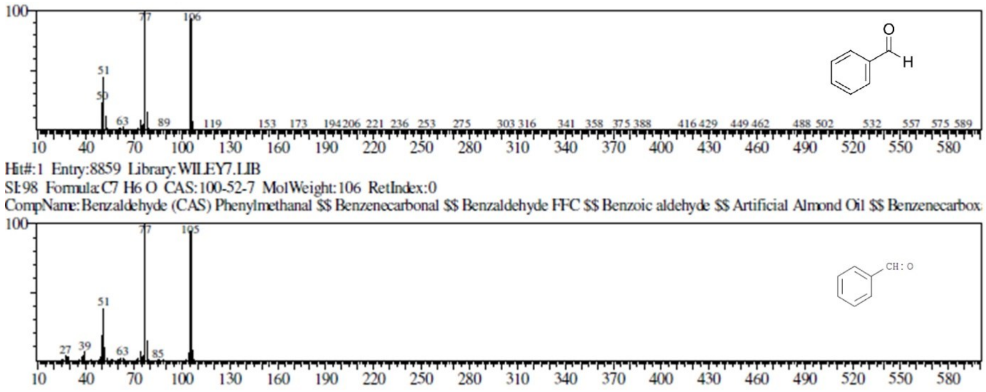


Figure 2. GC Chromatogram of the resulting ultrasonic-assisted oxidation of benzyl alcohol



Hit#:1 Entry:8859 Library:WILEY7.LIB
SI:98 Formula:C7 H6 O CAS:100-52-7 MolWeight:106 RetIndex:0
CompName:Benzaldehyde (CAS) Phenylmethanal $$ Benzenecarbonal $$ Benzaldehyde FFC $$ Benzoic aldehyde $$ Artificial Almond Oil $$ Benzenecarbox

Figure 3. MS spectra of the resulting ultrasonic-assisted benzyl alcohol oxidation (top) and benzaldehyde standard (bottom)

The oxidation of other alcohols was then investigated using the same ultrasonic-assisted oxidation method. Renewable alcohols such as *p*-anisyl alcohol and furfuryl alcohol were chosen because these alcohols could be potentially converted into other valued compounds. In this case, the oxidations generated the corresponding aldehydes in high yields (Table 1, entries 2 and 3). It is worth noting that ultrasonic-assisted oxidation of alcohols using CrO_3/Al_2O_3 is simple to perform and incredibly effective in converting the alcohols to aldehydes in reasonably excellent yield. This methodology can be regarded as ecologically benign processes according to the principles of green chemistry such as harmless *t*-butanol in solvent-less conditions was used, maximizing energy saving because the oxidation was done at ambient temperature and pressure, the oxidation proceeded in a quite fast accomplishment, and toxic waste and side product was minimized.

Effectiveness in resource exploitation can also be accomplished in terms of substituting fossil-based starting materials. It is worth mentioning that all alcohols utilized in this study are 100% renewable. Benzyl alcohol is a natural constituent produced by various plants, especially collecting in comestible fruits and tea leaves, along with in the volatile components of the Cananga tree (*Cananga odorata*), Jasmine (*Jasminum officinale*), hyacinths (*Hyacinthus orientalis*) [22] and the *de novo* biosynthesis of renewable glucose [23]. Furfuryl alcohol, a product obtained from agricultural waste such as corncob and sugar cane bagasse, is sustainable material [24]. *p*-anisyl alcohol is also renewable and can be derived from an anise plant [25]. This ultrasonic-assisted oxidation method is expected to be feasible in substituting the traditional oxidation methodology that still utilizes enormous amounts of solvent in relatively high energy consumption.

4. Conclusion

In this study, the development of simple, effective, relatively fast, and environmentally friendly oxidation of alcohols into aldehydes using CrO_3/Al_2O_3 has been carried out. Three renewable alcohols—benzyl alcohol, *p*-anisyl alcohol, and furfuryl alcohol—were successfully oxidized in satisfactory yields into their corresponding aldehydes. The results indicated that ultrasonic-assisted oxidation alcohols gave better yield and shorter reaction times than stirring techniques. Moreover, it was found that a small amount of *t*-butanol significantly increased both the reaction yield and shorter oxidation time. FTIR and GC/MS analyses confirmed the formation of the resulting aldehydes, and no carboxylic acids were detected.

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