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Stainless-Steel Ball-Milling Method for Hydro-/Deuterogenation using H_2O/D_2O as a Hydrogen/Deuterium Source

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A one-pot continuous-flow method for hydrogen (deuterium) generation and subsequent hydrogenation (deuterogenation) was developed using a stainless-steel (SUS304)-mediated ball-milling approach. SUS304, especially zero-valent Cr and Ni as constituents of the SUS304, and mechanochemical processing played crucial roles in the development of the reactions.

Minimizing the dependence on fossil fuels is currently under the spotlight to avoid global warming[1] through the reduction of greenhouse gases emitted from the combustion these materials. Additionally, the depletion of fossil fuels is of pending concern and alternatives are required. Although hydrogen gas is a sustainable and very promising next-generation energy carrier, many improvements are needed in the distribution, storage, generation and consumption of hydrogen to build a hydrogen economy.[2] While significant research was conducted on the topic over the past decade,[3] there are still many problems to be solved, such as energy efficiency, sustainability, safety, and operability. Therefore, the creation of an innovative, pioneering, and fundamental study with the potential for growth is extremely important.

Recently, we reported a quantitative stainless-steel (SUS304)-mediated gaseous H_2 generation method derived from H_2O that could be achieved using a simple ball-milling setup consisting of a planetary ball-mill machine with a stainless-steel vessel and balls.[7] Full conversion of H_2O in the reaction vessel was achieved by the rotation frequency dependent ball-milling reaction (see the Supporting Information for details) that was especially enabled by Cr metal as a structural component of the SUS304. During the course of the study, we discovered that the hydrogenation of reducible functionalities could smoothly proceed under the ball-milling conditions in the presence of H_2O as a hydrogen source. Although numerous protocols are published that use ball milling for organic chemistry,[8] an uninterrupted one-pot reaction of the SUS304-mediated quantitative production of H_2 from H_2O and subsequent hydrogenation of organic compounds is not reported in the literature. This is regarded as a convenient and direct H_2 transfer approach from H_2O to organic compounds.

Diphenylacetylene (**1**) and H_2O (30 equiv.) were added to a tightly-closed 12 mL SUS304 vessel without gas replacement (under air). The mixture was rotated at 800 rpm using a Fritsch Pulverisette 7 Classic Line Ball Mill (P-7) with SUS304 balls (ca. 5 mm diameter, 50 pieces). The rotation was paused for 1 min every 30 min after which the rotation was reversed; this method was used over the 3 h reaction time to prevent failure of the planetary ball mill. The organic products were determined by ^1H NMR analysis after filtration.

Hydrogenation of the alkyne moiety showed significant progress resulting in the formation of a mixture of *cis* and *trans*-stilbenes (**2** and **3**) and diphenylethane (**4**) in the ratio of 36:8:56 (Table 1, entry 1). The hydrogenation continued for 12 h to give diphenylethane (**4**) as the sole product in 98% isolated yield (entries 2 and 3). The reaction was dramatically accelerated by the addition of 5 mol% of Pd foil (entry 4), whereas 0.1 equivalents of 7,7,8,8-tetracyanoquinodimethane (TCNQ) completely inhibited the hydrogenation (entry 5). It was concluded that the series of reactions involved a single electron-transfer process and a hydrogenation step. Furthermore, the hydrogenation became less efficient with a decrease in the number of SUS304 balls (mechanochemical efficiency, entries 6 and 7), rotation speed (mechanochemical efficiency, entries 8–10), and usage of H_2O (hydrogen source, entries 11 and 12).

In our early study, it was demonstrated that the presence of zero-valent Cr within the constituents of SUS304 is essential to promote H_2 gas generation.[7] We subsequently investigated the metal efficiencies of zero-valent Ni, Fe, and Cr to clarify hydrogenation using H_2O (5 equiv.) as a hydrogen source under ball-milling conditions [ca. 5 mm diameter zirconia (ZrO_2) balls (50 pieces), 80 mL ZrO_2 vessel specially made for the Fritsch Pulverisette Premium Line 7 Ball Mill (PLP-7), 1100 rpm for 30 min]; we previously reported the chemical inactivity of ZrO_2 for the hydrogenation (Table 2, entry 1).[7] No significant effect was observed by the addition of 1 equivalent of zero-valent Fe, Cr, Ni, Mn, or Cr and Ni mixed powders as components of

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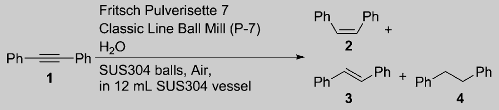
Table 1. Stainless-steel-mediated hydrogenation using H_2O as a reductant under ball-milling conditions.



Entry	H_2O [equiv.]	Number of balls	Rotation [rpm]	*t* [h]	Ratio [%][a]			
					1	2	3	4
1	30	50	800	3	0	36	8	56
2	30	50	800	6	0	5	5	90
3	30	50	800	12	0	0	0	100[b]
4[c]	30	50	800	3	0	0	0	100
5[d]	30	50	800	6	100	0	0	0
6	30	25	800	12	0	0	6	94
7	30	10	800	12	0	11	14	75
8	30	25	650	12	79	16	1	4
9	30	25	500	12	100	0	0	0
10	30	25	250	12	100	0	0	0
11	20	50	800	12	0	9	4	87
12	10	50	800	12	0	16	8	76

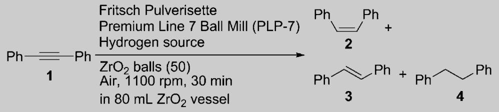
[a] The ratio was determined by ^{1}H NMR spectroscopy. [b] 98 % of **4** was isolated. [c] The reaction was performed in the presence of Pd foil (5.0 mol %). [d] The reaction was performed in the presence of TCNQ (0.1 equiv.).

SUS304 (entries 2, 3, 5, 7, and 8) in a 80 mL ZrO_2 vessel. A substantial amount of hydrogenation could proceed by the coexistence of Cr and Ni powders together with 5 equivalents of H_2O in a 20 mL ZrO_2 vessel (entry 9, ca. 35 % of **1** was consumed), whereas the single application of zero-valent Cr or Ni in a 20 mL ZrO_2 vessel only achieved 5 % hydrogenation (entries 4 and 6). As H_2 gas should be generated in the presence of zero-valent Cr powder (entry 3),[7] the hydrogenation may in this case be catalyzed by Ni powder. Furthermore, a certain level of hydrogen partial pressure may be necessary for hydrogenation (compare entries 8 and 9). As expected, hydrogenation proceeded in the presence of Ni powder under the H_2 gas conditions (entry 11), while Cr or Mn powder did not act as an efficient catalyst (entries 10 and 12). Therefore, the Ni-catalyzed hydrogenation proceeded with the consumption of H_2 gas generated by the Cr-mediated H_2 generation under the ball-milling conditions.

The scope of the reactions under the ball-milling conditions at 800 rpm is summarized in Table 3. Alkyne, alkene, aromatic bromide and iodide, nitro, and azide functionalities were all efficiently reduced under the reaction conditions (entries 1-10), although the phenolic benzyl ether could not be hydrogenated (entry 4). Aromatic ketones were hydrogenated to the corresponding *sec*-benzyl alcohol derivatives (entries 11–13). Consequently, the zero-valent Ni, as a constituent of SUS304, smoothly catalyzed the H_2 fixation reaction under the ball milling conditions.[9]

Deuterium labeled compounds are extremely useful in wide variety of scientific fields including analysis of metabolism, reaction mechanisms, kinetics, heavy drugs, and material sciences.[10] Although gaseous D_2 is useful as a convenient deuteri-

Table 2. Effect of metals and H_2 on the hydrogenation.

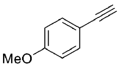
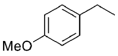
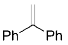
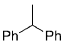
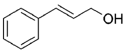
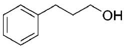
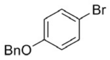
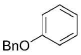
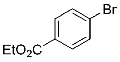
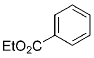
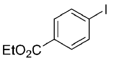
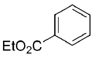
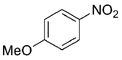
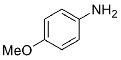
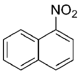
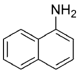
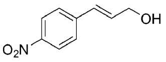
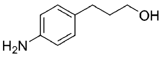
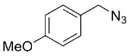
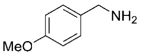
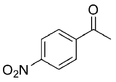
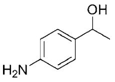
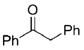
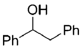
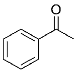
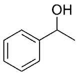


Entry	Hydrogen source	Additive (1 equiv.)	Ratio [%]			
			1	2	3	4
1	H_2O (5 equiv.)	none	100	0	0	0
2	H_2O (5 equiv.)	Fe	99	0	1	0
3	H_2O (5 equiv.)	Cr	91	3	5	1
4[a]	H_2O (5 equiv.)	Cr	95	1	3	1
5	H_2O (5 equiv.)	Ni	96	2	2	0
6[a]	H_2O (5 equiv.)	Ni	95	2	1	1
7	H_2O (5 equiv.)	Mn	95	1	2	2
8	H_2O (5 equiv.)	Cr and Ni[b]	95	2	1	2
9[a]	H_2O (5 equiv.)	Cr and Ni[b]	65	8	16	10
10	H_2 (7.2 equiv.)	Cr	81	11	5	1
11	H_2 (7.2 equiv.)	Ni	0	0	0	100[c]
12	H_2 (7.2 equiv.)	Mn	93	3	3	1

[a] A 20 mL ZrO_2 vessel was used instead of a 80 mL ZrO_2 vessel. [b] Cr and Ni powders were used (1 equiv. each). [c] 63 % of 4 was isolated.

Table 3. Scope of substrates used in the hydrogenation.[a]

$$\text{Substrate} \xrightarrow[\substack{\text{SUS 304 balls (50)}\\ \text{Air, 800 rpm,}\\ \text{in 12 mL SUS304 vessel}}]{\substack{\text{Fritsch Pulverisette 7}\\ \text{Classic Line Ball Mill (P-7)}\\ H_2O\text{ (30 equiv)}}} \text{Product}$$

Entry	Substrate	Product	*t* [h]	Yield [%]
1			12	69
2			6	100
3			6	95
4			12	91[b]
5			6	70
6			6	85
7			12	78
8			12	62
9			6	76
10			6	70
11			6	88
12			6	80
13			6	70

[a] 0.5 mmol of the substrate was reacted with 30 equiv. of H_2O using 50 SUS304 balls in a 12 mL SUS304 vessel. [b] A mixture of the unchanged substrate and the product (9:91) was obtained.

um source, the use of D_2 as a deuterium source is still problematic owing to the cost, flammability, and designation as a strategic material. Therefore, the development of in situ generation methods that retain small amounts of D_2 within the reaction vessel is of interest. Over the course of the present study, we have also discovered that the D_2 gas generated from D_2O could proceed with nearly the same efficiency as the H_2 generation from H_2O using a planetary ball mill in an 80 mL SUS304 vessel equipped with a wireless manometer terminal to observe the real-time internal pressure and temperature (compare Figure S1–S2 in the Supporting Information). The complete conversion (disappearance) of D_2O (271 µL, 15 mmol) into D_2 gas was achieved at 800 rpm in approximately 70 min. Therefore, we further investigated the deuterium labeling (fixation) reactions of substrates possessing reducible functionalities within the molecule. As expected, the deuterogenation efficiently occurred on alkyne, diyne, alkene, aromatic halide, and aromatic carbonyl functionalities with nearly quantitative deuterium efficiencies (Table 4, entries 1–9). Furthermore, the deuteration based upon the H–D exchange reaction also proceeded on the relatively active methylene and methyl groups. Consequently, more than 50% deuterium efficiencies were observed on the both methylenes of 3-phenyl ethyl propionate (entry 5) and the methyl group of 1-phenylethanol even though the deuterium efficiencies were not very high (entry 9).

In conclusion, we have demonstrated a SUS304 ball-milling-mediated continuous H_2 (D_2) generation reaction from H_2O (D_2O) along with the hydrogenation (deuterogenation and H–D exchange) reaction. The SUS304 and mechanochemical processing play crucial roles in the development of the reactions. It was especially revealed that zero-valent Cr as a constituent of SUS304 served for the H_2 (D_2) generation and Ni acted as a catalyst for the hydrogenation (deuterogenation). In all cases of the in situ hydrogenation and deuterogenation, the pure products could be isolated in good to excellent yields after simple workup procedures, which reduces the environmental impact. The present reactions are attractive as an environmentally-benign hydrogenation (deuterogenation) using H_2O (D_2O) as a hydrogen (deuterium) source. The feature of the present reaction is an innovative and widely applicable method. Further applications as a clean energy process for the next generation and research contributions towards a hydrogen economy are ongoing in our group.

Experimental Section

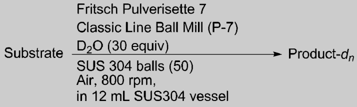
Equipment

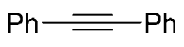
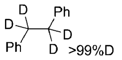
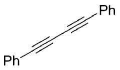
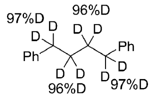
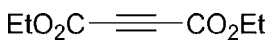
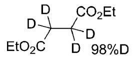
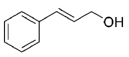
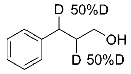
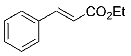
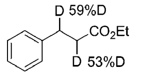
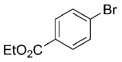
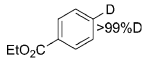
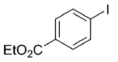
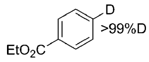
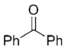
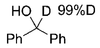
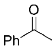
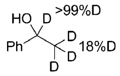
All experiments were performed using a commercially available Pulverisette 7 Classic Line Ball Mill (P-7) or Pulverisette Premium Line 7 Ball Mill (PLP-7) with a SUS304 or ZrO_2 vessel (20 mL or 80 mL) and cover manufactured by Fritsch.

Stainless-steel-mediated hydrogenation using H_2O as a reductant

Diphenylacetylene (89.1 mg, 0.5 mmol), SUS304 balls (ca. 5 mm diameter, adequate pieces), and H_2O (adequate amount) were added to a 12 mL SUS304 vessel. The vessel was closed using a SUS304 cover. The vessel was placed in a planetary ball mill (P-7) and rotated at 250, 500, 650, or 800 rpm for the adequate time, the rotation was stopped once for 1 min after the first 30 min-rotation, then the vessel was rotated again for another 30 min (inverse rotation). After the rotation, MeOH (20 mL) was added to the reaction mixture, and the suspension was filtered through a Celite pad and washed with MeOH (20 mL). The filtrate was concentrated in vacuo to yield the reduced products (**2**, **3**, and **4**). The ratio of products was determined by ^{1}H NMR spectroscopy.

Table 4. Deuteration using D_2O[a]



Entry	Substrate	Product	*t* [h]	Yield [%]
1			12	93
2			6	83
3			6	48
4			12	87
5			6	98
6			9	66
7			12	91
8			9	84
9			6	75

[a] 0.5 mmol of the substrate was reacted with 30 equiv. of D_2O using 50 SUS304 balls in a 12 mL SUS304 vessel.

Effect of metals and H_2 on the hydrogenation

Entries 1–4: Diphenylacetylene (89.1 mg, 0.5 mmol), selected metal powder (0.5 mmol), ZrO_2 balls (ca. 5 mm diameter, 50 pieces), and H_2O (270 µL, 15 mmol) were added to a 80 mL ZrO_2 vessel. The vessel was tightly sealed using a ZrO_2 cover. The vessel was placed in a planetary ball mill (PLP-7) and rotated at 1100 rpm for 30 min. After the rotation, MeOH (20 mL) was added to the reaction mixture, and the suspension was filtered through a Celite pad and washed with MeOH (20 mL). The filtrate was concentrated in vacuo to yield the reduced products (**2**, **3**, and **4**). The ratio of products was determined by ^{1}H NMR spectroscopy.

Entries 5 and 6: Diphenylacetylene (89.1 mg, 0.5 mmol), selected metal powder (0.5 mmol), and ZrO_2 balls (ca. 5 mm diameter, 50 pieces) were added to a 80 mL ZrO_2 vessel. The vessel was tightly sealed using a ZrO_2 cover. The inside gas was replaced by H_2 through the valve attached to cover. The vessel was placed in a planetary ball mill (PLP-7) and rotated at 1100 rpm for 30 min. After the rotation, MeOH (20 mL) was added to the reaction mixture, and the suspension was filtered through a Celite pad and washed with MeOH (20 mL). The filtrate was concentrated in vacuo to yield the reduced products (**2**, **3**, and **4**). The ratio of products was determined by ^{1}H NMR spectroscopy.

Scope of substrates used in the hydrogenation

The selected substrate (0.5 mmol), SUS304 balls (ca. 5 mm diameter, 50 pieces), and H_2O (270 μL, 15 mmol) were added to a 12 mL SUS304 vessel. The vessel was closed using a SUS304 cover. The vessel was placed in a planetary ball mill (P-7) and rotated at 800 rpm for the adequate time. After the rotation, MeOH (20 mL) was added to the reaction mixture, and the suspension was filtered through a Celite pad and washed with MeOH (20 mL). The filtrate was concentrated in vacuo to yield the reduced product. The residue was purified by silica-gel column chromatography, if necessary.

Deuteration using D_2O

The selected substrate (0.5 mmol), SUS304 balls (ca. 5 mm diameter, 50 pieces), and H_2O (270 μL, 15 mmol) were added to a 12 mL SUS304 vessel. The vessel was closed using a SUS304 cover. The vessel was placed in a planetary ball mill (P-7) and rotated at 800 rpm for the adequate time. After the rotation, MeOH (20 mL) was added to the reaction mixture, and the suspension was filtered through a Celite pad and washed with MeOH (20 mL). The filtrate was concentrated in vacuo to yield the reduced product. The residue was purified by silica-gel column chromatography, if necessary.

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Keywords: (heavy) water · ball milling · deuterogenation · hydrogenation · stainless-steel

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