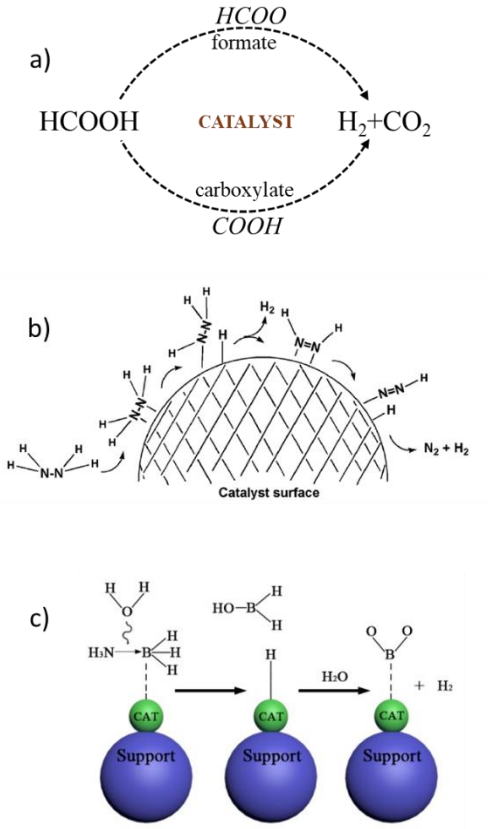

Figure S1. Schematic representation of the mechanisms of (**a**) formic acid, (**b**) hydrazine hydrate (Reprinted with permission from Ref. Singh, S. K.; Xu, Q., Nanocatalysts for hydrogen generation from hydrazine. *Catalysis Science & Technology* **2013,** *3*, 1889-1900. Royal Society of Chemistry) and (**c**) hydrolytic ammonia borane decomposition (Reprinted with permission from Ref. Liu, M.; Zhou, L.; Luo, X.; Wan, C.; Xu, L., Recent Advances in Noble Metal Catalysts for Hydrogen Production from Ammonia Borane. *Catalysts* **2020,** *10*, 788. The Authors).

Table S1. Properties of the considered hydrogen storage materials.

	Formic acid	Hydrazine hydrate	Ammonia borane (solid)
Formula	HCOOH	$N_2H_4{\cdot}H_2O$	NH_3BH_3
Hydrogen content, wt %	4.4	8	19.6 (7.8, when dissolved in H_2O)
By-products in decomposition	CO_2 (CO, H_2O)	N_2 (NH_3, H_2O)	NH_4BO_2 (if performed in water)
Molecular weight, g mol^{-1}	46.03	50.06	30.87
Boiling point, K	373.9	393	-
Melting point, K	281.5	221	377
Density, g cm^{-3}	1.22	1.029	0.78

Table S2. The most active catalysts in the formic acid dehydrogenation (TOF>1000 h^{-1}).

Catalyst	MNP mean size, nm	Temperature, K	Additive	TOF, h^{-1}	Reference
		Monometallic	catalysts		
Au/ZrO_2	1.8	323	triethylamine	1590	1
Au Schiff base modified SiO_2	1.2	323	none	4368	2
Pd/silicalite	1.5	323	Na formate	3027	3
Pd/silicalite	1.7	333	none	5803	4
Pd/C (MSC-30)	2.3	323	Na formate	2623	5
Pd/C	1.4	323	Na formate	4452	6
Pd/graphene (phenylenediamine modified)	1.5	323	Na formate	3810	7
Pd/C (MSC-30)	1.4	333	Na formate	8414	8
Pd/Fe-N-C	1.4	323	Na formate	7361	9
Pd/N-C	3.2	298	none	5530	10
Pd/C doped with K ions	3.6	353	gas-phase reaction	3600	11
		Bimetallic	catalysts		
AuPd/C	2-3	298	Na formate	1120	12
AuPd/N- graphene (amine modified)	2.4	298	Na formate	4446	13
$AuPd/ZrO_2/C/rGO$	2.5	323	Na formate	4500	14
AuPd/rGO	3.9	323	Na formate	4840	15
AuRh/N-C	1.6	333	Na formate	2297	16

Table S3. The most active catalysts in the hydrazine hydrate dehydrogenation (TOF>150 h^{-1}).

Catalyst	MNP mean size, nm	Temperature, K	TOF, h^{-1}	Reference
$Ni\text{-}CeO_2/SiO_2$	2.2	343	219.5	17
PtNi	2.4	298	150	18
PtNi/N-graphene	2.2	303	943	19
$PtNi/ZrO_2/C/graphene$	1.8	323	1920	20
$PtNi/TiO_2/Ti_3C_2T_x$	2.8	323	1220	21
PtNi/N-C	<10	323	1602	22
$PtNi\text{-}CeO_x$/N-graphene	2-3	323	3064	23
IrNi/lanthanum oxycarbonate	2.3	323	1250	24
$PtCo/La(OH)_3$	3.0	323	2400	25
RhNiP/graphene	2.75	323	471	26
PtNiP/rGO	2.0	323	742	27
$NiFe\text{-}Cr_2O_3$	10.8	343	893.5	28
$NiCu/CeO_2$	13.1	323	1450	29

Table S4. The most active catalysts in the ammonia borane dehydrogenation in the presence of water or methanol (TOF>200 min^{-1}).

Catalyst	MNP mean size, nm	Temperature, K	TOF, min^{-1}	Reference
hydrolysis				
Pt/MIL-101	1.8	298	414	30
Ru/graphene	1.9	298	600	31
Pt/CNTs	1.3	303	567	32
Rh/CNTs	2.3	298	760	33
PdRh@PVP	2.5	298	1333	34
RhRu/ZSM-5	0.7	298	1006	35
RuCu/ TiO_2@C-N	5.4	298	626	36
Ni_2Pt/ZIF-8	2.0	293	2222	37
CoNi/MoC	atomically dispersed	298	321.1	38
methanolysis				
Rh/CC3R	1.1	298	215.3	39