Electronic Supplementary Information

A facile synthesis methodology for preparation of Ag-Ni - Reduced Graphene Oxide: A magnetically separable versatile nanocatalyst for multiple organic reactions and Density Functional Study of its electronic structures

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Synthesis of the materials

Synthesis of graphene oxide (GO)

Graphene oxide was synthesized by using the modified Hummers method. In this method, 1 g of graphite powder and 0.6 g of NaNO₃ were mixed with 35 ml of H₂SO₄ at 0 °C. The mixture was stirred for 6 h, and then 3.8 g of KMnO₄ was added. The temperature was maintained at 35 °C for 8 h for complete oxidation process. After that, 60 ml of deionized water was added slowly and maintained the temperature at 98 °C for 1 h. Then 2 ml of 30% H₂O₂ was added and stirred for 0.5 h. The mixture was centrifuged, and then washed with 10% HCl, followed by distilled water. The yellowish brown precipitate of graphene oxide was obtained and dried at 60 °C.

Synthesis Methodology of (Ag₀·₂₇Ni₀·₇₃)₃₇RGO₆₃

The synthesis of (Ag₀·₂₇Ni₀·₇₃)₃₇RGO₆₃ which is composed of 10 wt % Ag, 27 wt% Ni, and 63 wt% RGO, was conducted in two steps. In the first step, graphene oxide (GO) was prepared by using the modified Hummers method. In the next step, a mixture of (21.13 ml) ethylene glycol and (0.469 mg) PVP (molar ratio 2:1) was prepared and 157.5 mg of GO was dispersed in this mixture. In this dispersion, 39.36 mg of AgNO₃ and 334.46 mg of Ni(NO₃)₆H₂O were added. After addition of metal nitrate salts, the mixture was stirred till salts were dissolved. In this mixture, NaOH (0.469 mg) pellets was added with stirring till the pH of the mixture became ~10, and then N₂H₄ (2.81 ml) was added dropwise keeping metal ion: N₂H₄ molar ratio of 1:40. After complete addition of N₂H₄, the reaction mixture was refluxed at 85 °C for 15 min. Then it was cooled to room temperature and precipitate thus formed was magnetically separated from the reaction mixture by using magnet externally. The collected precipitate was washed with distilled water until the pH of the washing reaches ~7. Finally, the precipitate was washed with acetone,
and then dried at 60 °C for 10 h. The formation and purity of the (Ag$_{0.27}$Ni$_{0.73}$)$_{37}$RGO$_{63}$ was confirmed using XRD.

**Table S1.** Codes of the catalyst and the corresponding compositions (Ag$_x$Ni$_{(1-x)}$)$_y$RGO$_{(100-y)}$.

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Characterization and Instrumentation

Room temperature powder X-ray diffraction (XRD) patterns of the synthesized materials were recorded using a powder X-ray diffractometer (Mini Flex II, Rigaku, Japan) with Cu Kα (λ = 0.15405 nm) radiation at a scanning speed of 3° min⁻¹. Fourier Transform Infrared spectra (FT-IR) were recorded in KBr by using spectrophotometer (IR Affinity-1, Shimadzu, Japan). Thermogravimetric analysis (TGA) was carried out using DTA-60 (Shimadzu, Japan). Field Emission Scanning Electron Microscope (FESEM) images of samples were obtained using Quanta 250 FEG (FEI). Energy dispersive X-ray spectra of the synthesized materials were recorded using an EDAX ELEMENT electron microscope. Raman spectra were recorded on a Renishaw Via Raman microscope with a 633 nm laser excitation. Room temperature magnetization with respect to an external magnetic field was measured for the synthesized catalysts using a Vibrating Sample Magnetometer (VSM) (EV5, ADE Technology, USA). Liquid Chromatography-Mass Spectrometer (LC-MS) full scan spectra were recorded on Agilent LC-MS for the product obtained from A3 coupling and ‘Click reaction’. ¹H NMR (Nuclear Magnetic Resonance) spectra were recorded on a BRUKER 400 ULTRA SHIELD PLUS (400 MHz) instrument using deuterated solvent. The gas chromatograph was carried out using (Shimadzu GC-2014) equipped with a capillary column (30 M × 0.25 mm × 0.25 mm) and a FID detector. Differential Scanning Calorimetric (DSC) analysis was carried out using DSC-60 (Shimadzu, Japan) to determine the melting point of the products obtained from Click reaction.
Computational details

In case of Ag and Ni unit cell (space group Fm-3m (225)) a Monkhorst-Pack mesh\(^2\) of \(k\)-points \(8 \times 8 \times 8\) is used, to sample the Brillouin zone for geometry optimization and for calculating the density of states.

Ag and Ni slab was constructed using Ag and Ni unit cell. The crystal was cleaved along (111) plane with four layer slab and vacuum space 10 Å along z direction. Here, \(4 \times 4 \times 1\) \(k\) point grids were used for optimization of structure and density of states calculations, respectively.

The initial superlattice structure of graphene was constructed using a \(2 \times 2 \times 1\) super cell with 8 atoms and 15 Å vacuum space at z-axis and optimized using \(4 \times 4 \times 1\) Monkhorst-Pack \(k\) point grid\(^2, 3\).

The Ag-Ni interface was constructed with Ni (111) slab placed 3 Å above the Ag (111) slab and 15 Å vacuum space at z-axis. Here, \(4 \times 4 \times 1\) \(k\) point grids were used for optimization of structure and density of states calculations, respectively. The Ag-Ni-graphene superlattice was constructed with Ag-Ni interface placed 3 Å above the monolayer graphene and 15 Å vacuum space at z-axis. Here, \(4 \times 4 \times 1\) \(k\) point grids were used for optimization of structure and density of states calculations respectively. The sizes of the unit cells of the systems simulated are listed in Table S1.

Table S2. The sizes of the unit cells of simulated systems.

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<tr>
<td>Ni unit cell</td>
<td>(a = b = c = 3.52) Å, (\alpha = \beta = \gamma = 90^\circ)</td>
</tr>
<tr>
<td>Graphene</td>
<td>(a = b = 4.9) Å, (c = 31.1) Å, (\alpha = \beta = 90^\circ, \gamma = 120^\circ)</td>
</tr>
<tr>
<td>Ag-slab</td>
<td>(a = b = 2.89) Å, (c = 18.8) Å, (\alpha = \beta = 90^\circ, \gamma = 120^\circ)</td>
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</table>
Ni-slab  \[ a = b = 2.49 \text{ Å} ; \ c = 21.1 \text{ Å} \quad \alpha = \beta = 90^\circ, \ \gamma = 120^\circ \]

Ag-Ni  \[ a = b = 4.99 \text{ Å} ; \ c = 29.5 \text{ Å} \quad \alpha = \beta = 90^\circ, \ \gamma = 120^\circ \]

Ag-Ni-graphene  \[ a = b = 8.26 \text{ Å} ; \ c = 25.3 \text{ Å} \quad \alpha = \beta = 90^\circ, \ \gamma = 120^\circ \]

Strain on interface  \[ \varepsilon_{11} = 1.46 \% , \ \varepsilon_{12} = 1.49 \% \]

Mean Absolute Strain = 3.66 \%

Details of the input files for geometric optimization of the Ni unit cell, Ag unit cell, graphene superlattice, Ni (111) slab, Ag (111) slab, Ag-Ni Interface, and Ag-Ni-graphene superlattice

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**Graphene Superlattice**

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    0.000000000  0.000000000  3.071492989
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    C   12.01070  C.pbe-n-rrkjus_psl.1.0.0.UPF
    Ni   58.69340  Ni.pbe-n-rrkjus_psl.1.0.0.UPF
ATOMIC_POSITIONS crystal
    Ag  0.482591405  0.095672042  0.224743330
    Ag  0.465961102  0.062649592  0.334183136
    Ag  0.831479001  0.246635872  0.276415281
    Ag  0.236669756  0.213712724  0.22111112
    Ag  0.588497593  0.400035102  0.162630853
    Ag  0.591309921  0.406810990  0.277998346
    Ni  0.580536144  0.664930848  0.22265145
    Ni  0.752406341  0.756524723  0.303370193
    Ni  0.416089494  0.588398347  0.302843962
    Ni  0.228387741  0.509982114  0.223294427
    Ni  0.582722583  0.892401734  0.27525982
    Ni  0.067298847  0.658065284  0.194879935
    Ni  0.399651617  0.792505391  0.185373272
    Ni  0.243059066  0.749696117  0.273926238
    C   0.704727396  0.619903418  0.049289043
    C   0.610324761  0.710564696  0.048857847
    C   0.325862383  0.425816647  0.048880519
    C   0.616529369  0.420547169  0.049633459
    C   0.415994403  0.331492447  0.049210889
    C   0.411109831  0.624209457  0.049273478
    C   0.317922411  0.719576935  0.050339693
    C   0.710288943  0.324958605  0.050288225
K_POINTS automatic
    4 4 1  0  0  0
**Fig. S1.** Room temperature wide angle powder XRD pattern of (a) Ni$_{10}$RGO$_{90}$, (b) Ni$_{20}$RGO$_{80}$, (c) Ni$_{40}$RGO$_{60}$, (d) Ag$_{0.05}$Ni$_{0.95}$, (e) Ag$_{0.15}$Ni$_{0.85}$, (f) Ag$_{0.25}$Ni$_{0.75}$, and (g) Ag$_{0.27}$Ni$_{0.73}$.

**Fig. S2.** Room temperature wide angle powder XRD pattern of (a) (Ag$_{0.05}$Ni$_{0.95}$)$_{90}$RGO$_{10}$, (b) (Ag$_{0.05}$Ni$_{0.95}$)$_{70}$RGO$_{30}$, (c) (Ag$_{0.05}$Ni$_{0.95}$)$_{60}$RGO$_{40}$, (d) (Ag$_{0.05}$Ni$_{0.95}$)$_{50}$RGO$_{50}$, (e) (Ag$_{0.05}$Ni$_{0.95}$)$_{40}$RGO$_{60}$, (f) (Ag$_{0.15}$Ni$_{0.85}$)$_{33.5}$RGO$_{66.5}$, (g) (Ag$_{0.37}$Ni$_{0.63}$)$_{40.5}$RGO$_{59.5}$, and (h) (Ag$_{0.45}$Ni$_{0.55}$)$_{44}$RGO$_{56}$. 
Fig. S3. FT-IR spectra of (a) GO, (b) RGO, and (c) (Ag\textsubscript{0.27}Ni\textsubscript{0.73})\textsubscript{37}RGO\textsubscript{63} nanocomposite.

Fig. S4. Raman spectra of (a) GO, (b) RGO, and (c) (Ag\textsubscript{0.27}Ni\textsubscript{0.73})\textsubscript{37}RGO\textsubscript{63} nanocomposite.
**Fig. S5.** STEM image of synthesized (Ag$_{0.27}$Ni$_{0.73}$)$_{37}$RGO$_{63}$ nanocomposite.

**Fig. S6.** (a) FESEM micrographs and (b-d) elemental color mapping of synthesized Ag$_{0.50}$Ni$_{0.50}$ nanocomposite.
Fig. S7. (a) FESEM micrographs and (b-e) elemental color mapping of synthesized \((\text{Ag}_{0.27}\text{Ni}_{0.73})_{37}\text{RGO}_{63}\) nanocomposite.

Fig. S8. EDS spectra of synthesized \(\text{Ag}_{0.50}\text{Ni}_{0.50}\) nanocomposite
**Fig. S9.** EDS spectra of synthesized (Ag_{0.27}Ni_{0.73})_{37}RGO_{63} nanocomposite.

**Fig. S10.** The initial structure of (a) graphene superlattice, (b) Ni unitcell, (c) Ag unitcell (d) Ni (111) slab, (e) Ag (111) slab, (f) Ag-Ni Interface, and (d) Ag-Ni-graphene superlattice.
Fig. S11. The optimized structure of (a) graphene superlattice, (b) Ni unitcell, (c) Ag unitcell (d) Ni (111) slab, (e) Ag (111) slab, (f) Ag-Ni Interface, and (d) Ag-Ni-graphene superlattice.

Fig. S12. The band structure and density of states of Ni (111) slab spin up.
Fig. S13. The band structure and density of states of Ni (111) slab spin down.

Fig. S14. The band structure and density of states of Ag (111) slab.
**Fig. S15.** The band structure and density of states of Ag-Ni interface spin up.

**Fig. S16.** The band structure and density of states of Ag-Ni interface spin down.
Fig. S17. The band structure and density of states of graphene superlattice.

Fig. S18. The band structure and density of states of Ag-Ni-graphene superlattice spin up.
Fig. S19. The band structure and density of states of Ag-Ni-graphene superlattice spin down.

Fig. S20. (a) Electrochemical impedance spectra of Pure Ni, Pure Ag, Ag$_{0.50}$Ni$_{0.50}$, (Ag$_{0.27}$Ni$_{0.73}$)$_{37}$RGO$_{63}$ and RGO (b) Inset shows the high frequency region of the impedance spectra and equivalent circuit used for fitting the Nyquist plots.
**Fig. S21.** Time dependent UV-Vis spectral changes of the reaction mixture of 4-NP catalyzed by (a) $\text{Ag}_{0.15}\text{Ni}_{0.85}$, (b) $\text{Ag}_{0.25}\text{Ni}_{0.75}$, (c) $\text{Ag}_{0.27}\text{Ni}_{0.73}$, (d) $(\text{Ag}_{0.05}\text{Ni}_{0.95})_{90}\text{RGO}_{10}$, (e) $(\text{Ag}_{0.05}\text{Ni}_{0.95})_{70}\text{RGO}_{30}$, (f) $(\text{Ag}_{0.05}\text{Ni}_{0.95})_{70}\text{RGO}_{30}$, (g) $(\text{Ag}_{0.05}\text{Ni}_{0.95})_{40}\text{RGO}_{60}$, (h) $(\text{Ag}_{0.15}\text{Ni}_{0.85})_{33.5}\text{RGO}_{66.5}$, and (i) $(\text{Ag}_{0.27}\text{Ni}_{0.73})_{37}\text{RGO}_{63}$.

**Table S3.** Comparison of catalytic efficiency of different reported catalysts for the reduction reaction of 4-NP in presence of NaBH$_4$. 
<table>
<thead>
<tr>
<th>Catalyst</th>
<th>Rate Constant ( (K_{\text{app}}) )</th>
<th>Reference</th>
</tr>
</thead>
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<td>Au nanoparticle</td>
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<td>[4]</td>
</tr>
<tr>
<td>Ag nanoparticle</td>
<td>4.06\times10^{-3} \text{s}^{-1}</td>
<td>[4]</td>
</tr>
<tr>
<td>Cu nanoparticle</td>
<td>1.5\times10^{-3} \text{s}^{-1}</td>
<td>[5]</td>
</tr>
<tr>
<td>AgNP-PG-5K</td>
<td>5.5\times10^{-3}\text{s}^{-1}</td>
<td>[6]</td>
</tr>
<tr>
<td>AgNP@PGMA-SH</td>
<td>3.94\times10^{-3}\text{s}^{-1}</td>
<td>[7]</td>
</tr>
<tr>
<td>Ag nanoparticles</td>
<td>1.26\times10^{-3}\text{s}^{-1}</td>
<td>[8]</td>
</tr>
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<td>Ag NPs with PAA stabilizer</td>
<td>15.46\times10^{-3}\text{s}^{-1}</td>
<td>[9]</td>
</tr>
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<td>Ni Nps</td>
<td>5.45\times10^{-3}\text{s}^{-1}</td>
<td>[10]</td>
</tr>
<tr>
<td>Ag monometallic</td>
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<td>[11]</td>
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<tr>
<td>Ni monometallic</td>
<td>3.7\times10^{-3}\text{s}^{-1}</td>
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</tr>
<tr>
<td>Ni NPs</td>
<td>2.7\times10^{-3}\text{s}^{-1}</td>
<td>[12]</td>
</tr>
<tr>
<td>RANEY Ni</td>
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<td>[12]</td>
</tr>
<tr>
<td>Ni NPs</td>
<td>1.4\times10^{-3}\text{s}^{-1}</td>
<td>[13]</td>
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<td>Ni-NPs composite brushes</td>
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<td>Pd-Ag</td>
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<td>Au-Ag</td>
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<tr>
<td>Bare Ag_{50}Ni_{50}</td>
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<td>Ag_{0.6}Ni_{0.4}</td>
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<td>[19]</td>
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<td>Ni/Ag</td>
<td>2.16\times10^{-3}\text{s}^{-1}</td>
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<td>Ni-Ag bimetallic</td>
<td>5.6\times10^{-3}\text{s}^{-1}</td>
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<td>Ni-RGO</td>
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<td>Ni-CNF</td>
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<td>Ag-NP/C composite</td>
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<tr>
<td>Ag-KCC</td>
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<tr>
<td>Catalyst</td>
<td>$k$ (s$^{-1}$)</td>
<td>Reference</td>
</tr>
<tr>
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<td>-----------------</td>
<td>-----------</td>
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<tr>
<td>Ag/PAN</td>
<td>$0.20 \times 10^{-3}$</td>
<td>[27]</td>
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<tr>
<td>PANI/Ag</td>
<td>$0.256 \times 10^{-3}$</td>
<td>[28]</td>
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<tr>
<td>RGONS/Ag-NSs</td>
<td>$0.535 \times 10^{-3}$</td>
<td>[29]</td>
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<tr>
<td>Ag/CNF</td>
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<td>Ni-Ag@RGO</td>
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<td>Ag-Au/rGO</td>
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<td>[30]</td>
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<td>CuO$_{0.05}$–rGO</td>
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<td>RGO-Ni$<em>{25}$Co$</em>{75}$</td>
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<td>[32]</td>
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<td>RGO-ZnNi$_{5}$-2</td>
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<td>[33]</td>
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<td>Ag$<em>{50}$Ni$</em>{50}$/RGO</td>
<td>$48.4 \times 10^{-3}$</td>
<td>[18]</td>
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<td>Au$<em>{1}$/Cu$</em>{3}$/rGO</td>
<td>$96 \times 10^{-3}$</td>
<td>[34]</td>
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<td>Fe$_3$O$_4$/graphene/Pt</td>
<td>$20.0 \times 10^{-3}$</td>
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<td>Fe$_3$O$_4$/graphene/Pd</td>
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<td>[35]</td>
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<td>Au/graphene hydrogel</td>
<td>$3.17 \times 10^{-3}$</td>
<td>[36]</td>
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<td>Ag@Fe$_3$O$_4$@C Core shell</td>
<td>$17.1 \times 10^{-3}$</td>
<td>[37]</td>
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<td>PtNi nanosnowflakes/RGO</td>
<td>$2.17 \times 10^{-3}$</td>
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<td>Ag@SBA-15</td>
<td>$1.7 \times 10^{-3}$</td>
<td>[39]</td>
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<td>2.5Ru@SBA-15</td>
<td>$13.5 \times 10^{-3}$</td>
<td>[40]</td>
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<tr>
<td>Pure Ni</td>
<td>$2.46 \times 10^{-3}$</td>
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<td>Ni$<em>{40}$RGO$</em>{60}$</td>
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<td>Ag$<em>{0.05}$Ni$</em>{0.95}$</td>
<td>$1.50 \times 10^{-3}$</td>
<td>This work</td>
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<tr>
<td>$(\text{Ag}<em>{0.05}\text{Ni}</em>{0.95})<em>{60}$ RGO$</em>{40}$</td>
<td>$19.60 \times 10^{-3}$</td>
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### A3 coupling reaction

**Table S4.** Catalytic performance of the as-prepared catalysts for synthesis of N,N-diethyl-3-phenylprop-2-yn-1-amine.
<table>
<thead>
<tr>
<th>Catalyst</th>
<th>Yield (%)</th>
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<tbody>
<tr>
<td>Pure Ni</td>
<td>71</td>
</tr>
<tr>
<td>Pure Ag</td>
<td>79</td>
</tr>
<tr>
<td>Ni&lt;sub&gt;10&lt;/sub&gt;RGO&lt;sub&gt;90&lt;/sub&gt;</td>
<td>43</td>
</tr>
<tr>
<td>Ni&lt;sub&gt;20&lt;/sub&gt;RGO&lt;sub&gt;80&lt;/sub&gt;</td>
<td>67</td>
</tr>
<tr>
<td>Ni&lt;sub&gt;30&lt;/sub&gt;RGO&lt;sub&gt;70&lt;/sub&gt;</td>
<td>80</td>
</tr>
<tr>
<td>Ag&lt;sub&gt;0.50&lt;/sub&gt;Ni&lt;sub&gt;0.50&lt;/sub&gt;</td>
<td>85</td>
</tr>
<tr>
<td>(Ag&lt;sub&gt;0.15&lt;/sub&gt;Ni&lt;sub&gt;0.85&lt;/sub&gt;)&lt;sub&gt;33.5&lt;/sub&gt;RGO&lt;sub&gt;66.5&lt;/sub&gt;</td>
<td>90</td>
</tr>
<tr>
<td>(Ag&lt;sub&gt;0.27&lt;/sub&gt;Ni&lt;sub&gt;0.73&lt;/sub&gt;)&lt;sub&gt;37&lt;/sub&gt;RGO&lt;sub&gt;63&lt;/sub&gt;</td>
<td>95</td>
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<tr>
<td>(Ag&lt;sub&gt;0.37&lt;/sub&gt;Ni&lt;sub&gt;0.63&lt;/sub&gt;)&lt;sub&gt;40.5&lt;/sub&gt;RGO&lt;sub&gt;59.5&lt;/sub&gt;</td>
<td>95</td>
</tr>
<tr>
<td>(Ag&lt;sub&gt;0.45&lt;/sub&gt;Ni&lt;sub&gt;0.55&lt;/sub&gt;)&lt;sub&gt;44&lt;/sub&gt;RGO&lt;sub&gt;56&lt;/sub&gt;</td>
<td>96</td>
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</table>

<sup>a</sup>Reaction condition: Paraformaldehyde (2mmol), Diethylamine (2.4mmol), Phenylacetylene (3 mmol), Catalyst amount 50 mg, Acetonitrile 10 ml, Reaction temperature 100 °C, reaction time 12 h.

Table S5. Synthesis of N,N-diethyl-3-phenylprop-2-yn-1-amine under various reaction conditions in the presence of (Ag<sub>0.27</sub>Ni<sub>0.73</sub>)<sub>37</sub>RGO<sub>63</sub> <sup>a</sup>.
<table>
<thead>
<tr>
<th>Catalyst</th>
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<th>Time</th>
<th>% of Yield</th>
<th>Reference</th>
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<td>Ag/diatomite</td>
<td>Toluene</td>
<td>24 h</td>
<td>82</td>
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<td>Ag₂O/alumina</td>
<td>water</td>
<td>2 h</td>
<td>92</td>
<td>[42]</td>
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<tr>
<td>Ag/SBA-15-6</td>
<td>Glycol</td>
<td>30 min</td>
<td>95</td>
<td>[43]</td>
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<tr>
<td>H-Fe₂O₅@DA/Ag</td>
<td>-</td>
<td>40 min</td>
<td>90</td>
<td>[44]</td>
</tr>
<tr>
<td>Au/ZrO₂</td>
<td>Dioxane</td>
<td>6 h</td>
<td>90</td>
<td>[45]</td>
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<tr>
<td>(Ag₀.２₇Ni₀.₇₃)₃₇RGO₆₃</td>
<td>Acetonitrile</td>
<td>12 h</td>
<td>95</td>
<td>Present work</td>
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</table>

**Styrene Epoxidation reaction**

**Table S7.** Effect of catalyst composition on the percentage of styrene conversion and percentage of selectivity of styrene oxide and benzaldehyde formation.

<table>
<thead>
<tr>
<th>Catalyst</th>
<th>Conversion (%)</th>
<th>Styrene Oxide (%)</th>
<th>PhCHO (%)</th>
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<tr>
<td>Pure Ni</td>
<td>69</td>
<td>85</td>
<td>5</td>
</tr>
<tr>
<td>Pure Ag</td>
<td>76</td>
<td>88</td>
<td>4</td>
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</table>
Fig. S22. Gas chromatography analysis of styrene epoxidation reaction with (a) Pure Ni, (b) Ni$_{30}$RGO$_{70}$, (c) Pure Ag, (d) Ag$_{0.50}$Ni$_{0.50}$, (e) (Ag$_{0.15}$Ni$_{0.85}$)$_{33.5}$RGO$_{66.5}$, (f) (Ag$_{0.27}$Ni$_{0.73}$)$_{37}$RGO$_{63}$, (g) (Ag$_{0.37}$Ni$_{0.63}$)$_{40.5}$RGO$_{59.5}$, and (h) (Ag$_{0.45}$Ni$_{0.55}$)$_{44}$RGO$_{56}$.

**Reaction condition**: Styrene (5 mmol), TBHP (12.5 mmol), Acetonitrile (4 ml), Reaction time = 10 h, Reaction temperature = 100 °C, catalyst amount = 25 mg
**Fig. S23.** Gas chromatography analysis of progress of (Ag_{0.27}Ni_{0.73})_{37}RGO_{63} catalyzed styrene epoxidation reaction with time.

**Table S8.** Epoxidation of styrene under various reaction condition in the presence of the (Ag_{0.27}Ni_{0.73})_{37}RGO_{63}.

<table>
<thead>
<tr>
<th>Reaction Temperature (°C)</th>
<th>Reaction Time (hours)</th>
<th>Catalyst Amount (mg)</th>
<th>Styrene : TBHP molar ratio</th>
<th>Conversion (%)</th>
<th>Selectivity</th>
</tr>
</thead>
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<tr>
<td></td>
<td></td>
<td></td>
<td></td>
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<td>Styrene Oxide (%)</td>
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<tr>
<td>80°C</td>
<td>10</td>
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<td>1:2</td>
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<td>57</td>
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<td>100°C</td>
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<td>25</td>
<td>1:2</td>
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<td>120°C</td>
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<td>1:2</td>
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<td>100°C</td>
<td>10</td>
<td>10</td>
<td>1:2</td>
<td>65</td>
<td>69</td>
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<tr>
<td>100°C</td>
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<td>50</td>
<td>1:2</td>
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<td>74</td>
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<td>1:1</td>
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<td>49</td>
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<td>100°C</td>
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<td>1:2.5</td>
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<tr>
<td>100°C</td>
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<td>1:3.0</td>
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<td>25</td>
<td>1:2.0</td>
<td>50</td>
<td>94</td>
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Fig. S24. Change of conversion and product selectivity with time catalyzed by (Ag$_{0.27}$Ni$_{0.73}$)$_{37}$RGO$_{63}$ for epoxidation of styrene. Reaction condition: 5 mmol of Styrene, 12.5 mmol of TBHP were stirred in 4 ml Acetonitrile for 12 h at 100 °C using 25 mg catalyst.

Table S9. Comparison of the catalytic efficiency of (Ag$_{0.27}$Ni$_{0.73}$)$_{37}$RGO$_{63}$ with different reported catalysts for epoxidation reaction of styrene.
<table>
<thead>
<tr>
<th>Catalyst</th>
<th>Solvent</th>
<th>Time (h)</th>
<th>Styrene Conversion/Yield (%)</th>
<th>Selectivity of Styrene oxide formation</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ag-Fe₃O₄</td>
<td>Toluene</td>
<td>13</td>
<td>100</td>
<td>84</td>
<td>[46]</td>
</tr>
<tr>
<td>Ag-Ni₀.₈₁Fe₂.₁₉O₄</td>
<td>Toluene</td>
<td>5</td>
<td>69.8</td>
<td>84.8</td>
<td>[47]</td>
</tr>
<tr>
<td>Ag-γ-ZrP</td>
<td>Acetonitrile</td>
<td>8</td>
<td>44.7</td>
<td>92.9</td>
<td>[48]</td>
</tr>
<tr>
<td>AgNps/CNFs(1/10)</td>
<td>Isopropanol</td>
<td>8</td>
<td>43.4</td>
<td>38.9</td>
<td>[49]</td>
</tr>
<tr>
<td>Ag CNFs</td>
<td>Acetonitrile</td>
<td>6</td>
<td>61.4</td>
<td>81.5</td>
<td>[50]</td>
</tr>
<tr>
<td>Ag–Cu/Cu₂O CNFs</td>
<td>Acetonitrile</td>
<td>6</td>
<td>99</td>
<td>41.9</td>
<td>[50]</td>
</tr>
<tr>
<td>Ag/SBA-15</td>
<td>Acetonitrile</td>
<td>9</td>
<td>77.7</td>
<td>73.7</td>
<td>[51]</td>
</tr>
<tr>
<td>Ag/LDH</td>
<td>Acetonitrile</td>
<td>8</td>
<td>80.8</td>
<td>91.1</td>
<td>[52]</td>
</tr>
<tr>
<td>Ag/4A Zeolite</td>
<td>Acetonitrile</td>
<td>48</td>
<td>80.8</td>
<td>89.2</td>
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<td>TiO₂-Ag</td>
<td>Toluene</td>
<td>14</td>
<td>83.9</td>
<td>66.8</td>
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<td>TiO₂-GO</td>
<td>Acetonitrile</td>
<td>12</td>
<td>93.3</td>
<td>85.9</td>
<td>[55]</td>
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<tr>
<td>(Ag₀.₂₇Ni₀.₇₃)₃₇RGO₆₃</td>
<td>Acetonitrile</td>
<td>10</td>
<td>95</td>
<td>87</td>
<td>Present work</td>
</tr>
</tbody>
</table>

Click Reaction
Table S10. Comparison of the catalytic efficiency of \((\text{Ag}_{0.27}\text{Ni}_{0.73})_{37}\text{RGO}_{63}\) with different reported catalysts in the preparation of 2-phenyl-2-(4-phenyl-1H-1,2,3-triazole-1-yl) ethanol via Click reaction.

<table>
<thead>
<tr>
<th>Catalyst</th>
<th>Solvent</th>
<th>Time</th>
<th>% of Yield</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu(^{II})-hydrotalcite</td>
<td>Water</td>
<td>5 h</td>
<td>91</td>
<td>[56]</td>
</tr>
<tr>
<td>Cu(^{II})-PhTPY</td>
<td>water</td>
<td>1 h</td>
<td>95</td>
<td>[57]</td>
</tr>
<tr>
<td>copper(I)@phosphorated SiO(_2)</td>
<td>water</td>
<td>1 h</td>
<td>94</td>
<td>[58]</td>
</tr>
<tr>
<td>Copper ferrite nanoparticles</td>
<td>water</td>
<td>6 h</td>
<td>87</td>
<td>[59]</td>
</tr>
<tr>
<td>CuI</td>
<td>PEG-400</td>
<td>16 h</td>
<td>83</td>
<td>[60]</td>
</tr>
<tr>
<td>CuSO(_4).5H(_2)O/Sodiumascorbate</td>
<td>water</td>
<td>4 h</td>
<td>92</td>
<td>[61]</td>
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<tr>
<td>GO@PTA-Cu</td>
<td>water</td>
<td>35 min</td>
<td>94</td>
<td>[62]</td>
</tr>
<tr>
<td>Cu@SBA-15@CF</td>
<td>water</td>
<td>2 h</td>
<td>90</td>
<td>[63]</td>
</tr>
<tr>
<td>CuO@mTiO(_2@CF)</td>
<td>water</td>
<td>6 h</td>
<td>89</td>
<td>[64]</td>
</tr>
<tr>
<td>98BiFeO(_3)-2RGO</td>
<td>water</td>
<td>5 h</td>
<td>91</td>
<td>[65]</td>
</tr>
<tr>
<td>((\text{Ag}<em>{0.27}\text{Ni}</em>{0.73})<em>{37}\text{RGO}</em>{63})</td>
<td>water</td>
<td>12 h</td>
<td>88</td>
<td>Present work</td>
</tr>
</tbody>
</table>

Spectral Data
Product of A3 coupling reaction (Table 2)

**Entry-1: N,N-diethyl-3-phenylprop-2-yn-1-amine**

$^1$H NMR (CDCl$_3$) $\delta$ = 7.45-7.43 (m, 2H), 7.32-7.30 (m, 3H), 3.66 (s, 2H), 2.67-2.62 (q, 4H), 1.15-1.12 (t, 6H) (Ref. 43), IR (Liquid film cm$^{-1}$): $\nu$ = 2974, 2815, 1600, 1494, 1380, 1318, 1195, 1089, 1054, 983, 754, 701, 613. LC-MS calculated. (C$_{13}$H$_{17}$N) (M$^+$) : 187.14 found : 188.00.

**Entry-2: N,N-diethyl-1,3-diphenylprop-2-yn-1-amine**

$^1$H NMR (CDCl$_3$) $\delta$ = 7.54-7.38 (m, 10H), 1.70 (s, 3H), 1.69 (m, 2H), 0.90 (m, 3H), 0.83 (m, 4H) (Ref. 43, 66), IR (Liquid film cm$^{-1}$): $\nu$ = 2983, 2806, 2306, 1990, 1931, 1852, 1615, 1547, 1485, 1370, 1197, 1067, 981, 750, 657. LC-MS calculated. (M$^+$) : 263.17 (C$_{19}$H$_{21}$N) found : 262.70.

**Entry-3: 1-(1,3-diphenylprop-2-yn-1-yl)piperidine**

$^1$H NMR (CDCl$_3$) $\delta$ = 7.67-7.66 (m, 2H), 7.56-7.54 (m, 2H), 7.41-7.32 (m, 6H), 4.85 (s, 1H), 2.60 (m, 4H), 1.65-1.61 (m, 4H), 1.48-1.46 (m, 2H) (Ref. 43), IR (Liquid film cm$^{-1}$): $\nu$ = 3286, 3062, 2935, 2779, 2723, 1694, 153, 1491, 1442, 1315, 1206, 1164, 1071, 999, 916, 829, 756, 684, 643. LC-MS calculated. (M$^+$) : 275.17 (C$_{20}$H$_{21}$N) found : 276.00.

**Entry-4: 1-(3-phenyl-1-(p-tolyl)prop-2-yn-1-yl)piperidine**

$^1$H NMR (CDCl$_3$) $\delta$ = 7.54-7.52 (m, 4H), 7.35-7.34 (m, 3H), 7.20-7.18 (m, 2H), 4.80 (s, 1H), 2.59 (s, 4H), 2.38 (s, 3H), 1.64-1.61 (m, 4H), 1.47-1.46 (m, 2H) (Ref. 67), IR (Liquid film cm$^{-1}$): $\nu$ =
3026, 2886, 2731, 1931, 1548, 1480, 1305, 1164, 1106, 984, 909, 815, 747, 689, 542. LC-MS calculated. $(M^+): 289.18(C_{21}H_{23}N)$ found : 290.00.

**Entry-5: 1-(1-(4-chlorophenyl)-3-phenylprop-2-yn-1-yl)piperidine**

$^1$H NMR (CDCl$_3$) $\delta$ = 7.61 - 7.59 (m, 2H), 7.54 - 7.53 (m, 2H), 7.36 - 7.33 (m, 5H), 4.80 (s, 1H), 2.57 (s, 4H), 1.64 - 1.60 (m, 4H), 1.48 - 1.47 (m, 2H) (Ref. 42), IR (Liquid film cm$^{-1}$): $\nu = 3285, 3057, 2840, 2716, 1924, 1699, 1574, 1481, 1378, 1285, 1202, 1089, 1006, 820, 748, 686, 549$. LC-MS calculated. $(M^+): 309.83(C_{20}H_{20}NCl)$ found : 310.00 42.

**Entry-6: 1-(1-(4-bromophenyl)-3-phenylprop-2-yn-1-yl)piperidine**

$^1$H NMR (CDCl$_3$) $\delta$ = 7.78 - 7.70 (m, 2H), 7.56 - 7.48 (m, 4H) 7.36 - 7.35 (m, 3H), 4.79 (s, 1H) 2.57 (s, 4H), 1.63 - 1.59 (m, 4H), 1.48 - 1.46 (m, 2H), IR (Liquid film cm$^{-1}$): $\nu = 2932, 2850, 2798, 1671, 1599, 1485, 1444, 1392, 1320, 1279, 1165, 1072, 1000, 814, 752, 690, 534$. LC-MS calculated. $(M^+): 354.28 (C_{20}H_{20}NBr)$ found : 355.80 42.

**Entry-7: 1-(3-phenylprop-2-yn-1-yl)piperidine**

$^1$H NMR (CDCl$_3$) $\delta$ = 7.46 - 7.45 (m, 2H), 7.32 - 7.30 (m, 3H), 3.50 (s, 2H), 2.59 (s, 4H), 1.69 - 1.65 (m, 4H), 1.47 (s, 2H) (Ref. 43), IR (Liquid film cm$^{-1}$): $\nu = 3286, 3062, 2935, 2799, 2723, 1694, 1593, 1491, 1442, 1315, 1206, 1164, 1071, 999, 916, 829, 756, 684, 643 66. LC-MS calculated. $(M^+): 199.29(C_{14}H_{17}N)$ found : 199.70 66.

**Entry-8: N-(1,3-diphenylprop-2-yn-1-yl)aniline**
\textbf{Entry-9:} N-(3-phenyl-1-(p-tolyl)prop-2-yn-1-yl)aniline

\textbf{Entry-10:} N-(1-(4-methoxyphenyl)-3-phenylprop-2-yn-1-yl)aniline

\textbf{Entry-11:} N-(1-(4-nitrophenyl)-3-phenylprop-2-yn-1-yl)aniline

\textbf{Product of ‘Click reaction’}

2-phenyl-2-(4-phenyl-1\textit{H}-1,2,3-triazole-1-yl)ethanol
Solid, m.p. 126-128°C (reported 125-127 °C (Ref.62)). IR (KBr): 3427, 3123, 3089, 3029, 2939, 1493, 1455, 1431, 1223, 1084, 1057, 755, 693 cm⁻¹. LC-MS (EI) m/z calculated for C₁₆H₁₅N₃O 265.12, observed 266.00 (reported 265.12 (Ref. 62)).

2-(4-Phenyl-1H-1,2,3-triazol-1-yl)cyclohexanol

Solid, m.p. 170-172 °C (reported 167.8-171.8 °C (Ref.62)). IR (KBr): 3298, 3119, 2938, 2858, 1447, 1232, 1054, 763, 696 cm⁻¹. LC-MS (EI) m/z calculated for C₁₄H₁₇N₃O 243.13, observed 244.00 (HRMS reported for C₁₄H₁₈N₃O 244.14 (Ref. 62)).

1,3-Bis(4-phenyl-1H-1,2,3-triazol-1-yl)propan-2-ol

Solid, m.p. 235-237 °C (reported 233-236°C (Ref. 62)), IR (KBr): 2959, 2938, 2858, 1447, 1232, 1054, 763 cm⁻¹. LC-MS (EI) m/z calculated for C₁₉H₁₉N₆O 346.13, observed 347.00 (HRMS reported for C₁₉H₁₉N₆O 346.20 (Ref. 62)).
**Fig. S25b.** LC-MS full scan of N,N-diethyl-1,3-diphenylprop-2-yn-1-amine.

**Fig. S25c.** LC-MS full scan of 1-(1,3-diphenylprop-2-yn-1-yl)piperidine.
Fig. S25d. LC-MS full scan of 1-(3-phenyl-1-(p-tolyl)prop-2-yn-1-yl)piperidine.

Fig. S25e. LC-MS full scan of 1-(1-(4-chlorophenyl)-3-phenylprop-2-yn-1-yl)piperidine.
**Fig. S25f.** LC-MS full scan of 1-(1-(4-bromophenyl)-3-phenylprop-2-yn-1-yl)piperidine.

**Fig. S25g.** LC-MS full scan of 1-(3-phenylprop-2-yn-1-yl)piperidine.
**Fig. S25h.** LC-MS full scan of N-(1,3-diphenylprop-2-yn-1-yl)aniline.

**Fig. S25i.** LC-MS full scan of N-(3-phenyl-1-(p-tolyl)prop-2-yn-1-yl)aniline.
Fig. S25j. LC-MS full scan of N-(1-(4-methoxyphenyl)-3-phenylprop-2-yn-1-yl)aniline.

Fig. S25k. LC-MS full scan of N-(1-(4-nitrophenyl)-3-phenylprop-2-yn-1-yl)aniline.
Fig. S25l. LC-MS full scan of 2-phenyl-2-(4-phenyl-1H-1,2,3-triazole-1-yl)ethanol.

Fig. S25m. LC-MS full scan of 2-(4-phenyl-1H-1,2,3-triazole-1-yl)cyclohexanol.
**Fig. S25n.** LC-MS full scan of 1,3-Bis(4-phenyl-1H-1,2,3-triazole-1-yl)propan-2-ol.

**Fig. S26a.** $^1$H NMR spectrum of N,N-diethyl-3-phenylprop-2-yn-1-amine synthesized via A3 coupling reaction.
Fig. S26b. $^1$H NMR spectrum of N,N-diethyl-1,3-diphenylprop-2-yn-1-amine synthesized via A3 coupling reaction.

Fig. S26c. $^1$H NMR spectrum of 1-(1,3-diphenylprop-2-yn-1-yl)piperidine synthesized via A3 coupling reaction.
**Fig. S26d.** $^1$H NMR spectrum of 1-(3-phenyl-1-(p-tolyl)prop-2-yn-1-yl)piperidine synthesized via A3 coupling reaction.

**Fig. S26e.** $^1$H NMR spectrum of 1-(1-(4-chlorophenyl)-3-phenylprop-2-yn-1-yl)piperidine synthesized via A3 coupling reaction.
**Fig. S26f.** $^1$H NMR spectrum of 1-(1-(4-bromophenyl)-3-phenylprop-2-yn-1-yl)piperidine synthesized via A3 coupling reaction.

**Fig. S26g.** $^1$H NMR spectrum of 1-(3-phenylprop-2-yn-1-yl)piperidine synthesized via A3 coupling reaction.
**Fig. S26h.** $^1$H NMR spectrum of N-(1,3-diphenylprop-2-yn-1-yl)aniline synthesized via A3 coupling reaction.

**Fig. S26i.** $^1$H NMR spectrum of N-(3-phenyl-1-(p-tolyl)prop-2-yn-1-yl)aniline synthesized via A3 coupling reaction.
Fig. S26j. $^1$H NMR spectrum of 1-(3-phenylprop-2-yn-1-yl)aniline synthesized via A3 coupling reaction.

Fig. S26k. $^1$H NMR spectrum of N-(1-(4-nitrophenyl)-3-phenylprop-2-yn-1-yl)aniline synthesized via A3 coupling reaction.
**Fig. S27.** FT-IR spectrum of (a) N,N-diethyl-3-phenylprop-2-yn-1-amine, (b) N,N-diethyl-1,3-diphenylprop-2-yn-1-amine, (c) 1-(1,3-diphenylprop-2-yn-1-yl)piperidine, (d) 1-(3-phenyl-1-(p-tolyl)prop-2-yn-1-yl)piperidine, (e) 1-(1-(4-chlorophenyl)-3-phenylprop-2-yn-1-yl)piperidine, (f) 1-(1-(4-bromophenyl)-3-phenylprop-2-yn-1-yl)piperidine, (g) 1-(3-phenylprop-2-yn-1-yl)piperidine, (h) N-(1,3-diphenylprop-2-yn-1-yl)aniline, (i) N-(3-phenyl-1-(p-tolyl)prop-2-yn-1-yl)aniline, (j) N-(1-(4-methoxyphenyl)-3-phenylprop-2-yn-1-yl)aniline, (k) N-(1-(4-nitrophenyl)-3-phenylprop-2-yn-1-yl)aniline.

**Fig. S28.** FT-IR spectrum of (a) 2-phenyl-2-(4-phenyl-1H-1,2,3-triazole-1-yl) ethanol (b) 2-(4-Phenyl-1H-1,2,3-triazol-1-yl) cyclohexanol (c) 1,3-Bis(4-phenyl-1H-1,2,3-triazol-1-yl)propan-2-ol.
Fig. S29. DSC of (a) 2-phenyl-2-(4-phenyl-1H-1,2,3-triazole-1-yl) ethanol and (b) 2-(4-Phenyl-1H-1,2,3-triazol-1-yl)cyclohexanol (c) 1,3-Bis(4-phenyl-1H-1,2,3-triazol-1-yl)propan-2-ol.
**Fig. S30.** Room temperature magnetic hysteresis loop of Pure Ni, Ni$_{30}$RGO$_{70}$, (Ag$_{0.15}$Ni$_{0.85}$)$_{33.5}$RGO$_{66.5}$, (Ag$_{0.27}$Ni$_{0.73}$)$_{37}$RGO$_{63}$, (Ag$_{0.37}$Ni$_{0.63}$)$_{40.5}$RGO$_{59.5}$, and (Ag$_{0.45}$Ni$_{0.55}$)$_{44}$RGO$_{56}$.

**Fig. S31.** Magnetic separation of the catalyst by applying a magnet externally after completion of (a) reduction of 4-NP in presence of NaBH$_4$, (b) A3 coupling reaction, (c) epoxidation of styrene, and (d) Click reaction.
Fig. S32. (A) XRD and (B) FESEM image of the recycled $(\text{Ag}_{0.27}\text{Ni}_{0.73})_{37}\text{RGO}_{63}$ catalyst.

References:

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