Supplementary Information:

Reversible hydrogenation restores defected graphene to graphene

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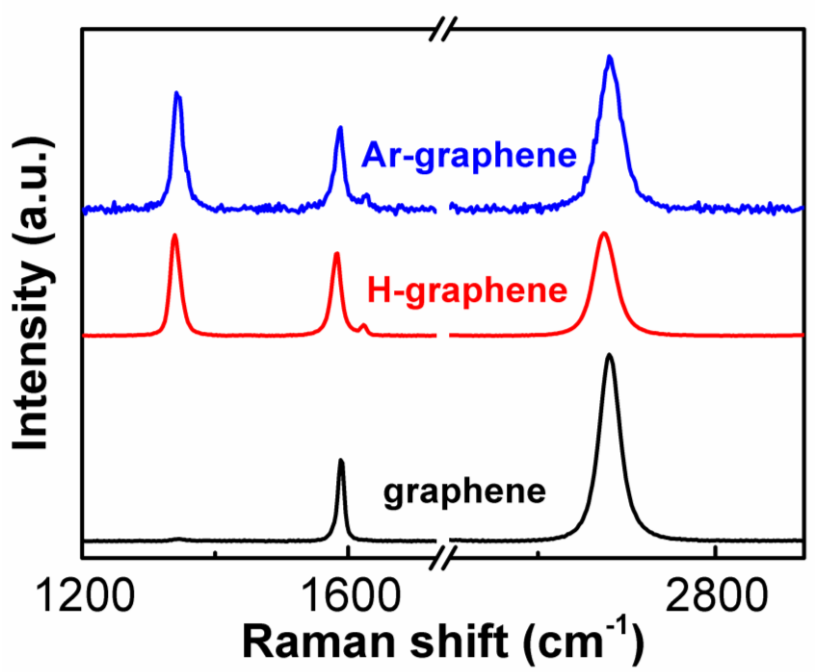


Figure S1. Raman spectra of graphene, H-graphene and Ar-graphene. Similar densities of hydrogenated and vacancy defects are confirmed by the peak intensity ratio between D (~1350 cm^{-1}) and G peaks (~1580 cm^{-1}). The spectra recorded using a 2.33 eV (532 nm) laser excitation are calibrated according to the G peak intensity.

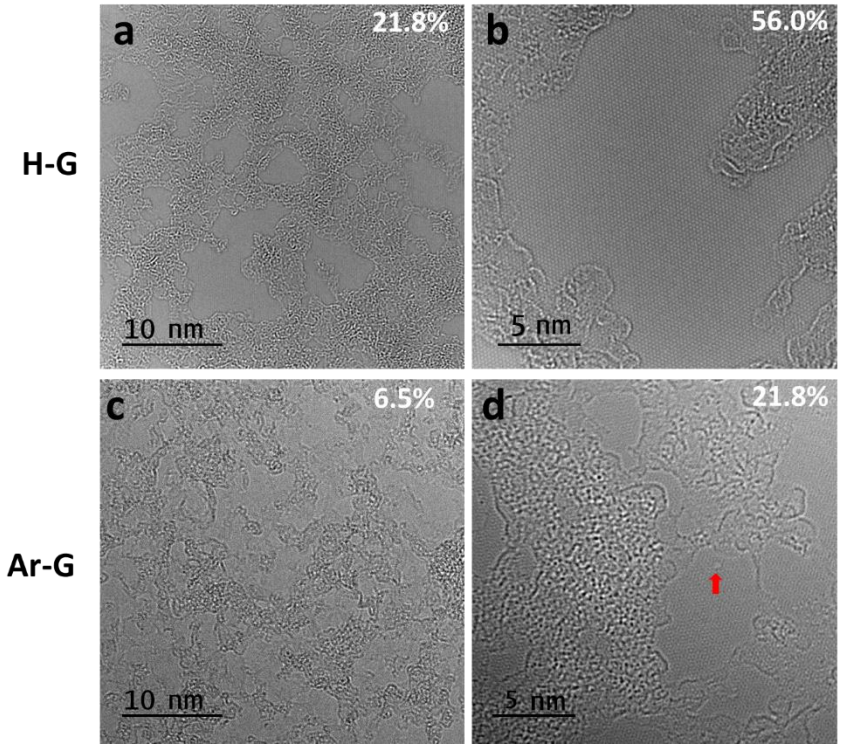


Figure S2. HRTEM images of H-G (a, b) and Ar-G (c, d). Ar-G: graphene treated by 13 s of argon plasma. A zoomed-in HRTEM image of the Ar-G in (d) with a void indicated by the red arrow, which could be induced by the energetic argon ion bombardment or electron irradiation during imaging.

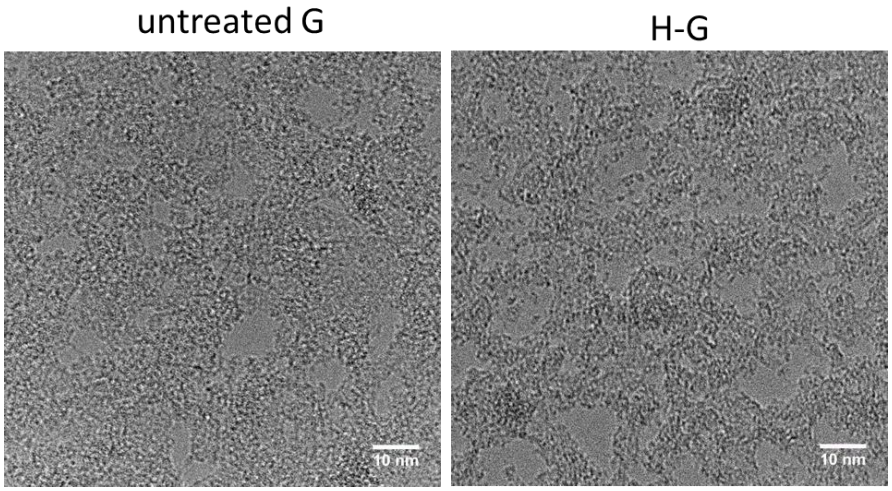


Figure S3. TEM images of untreated G vs H-G (60 s) after two week's exposure to ambient condition, both showing heavily contaminated surfaces.

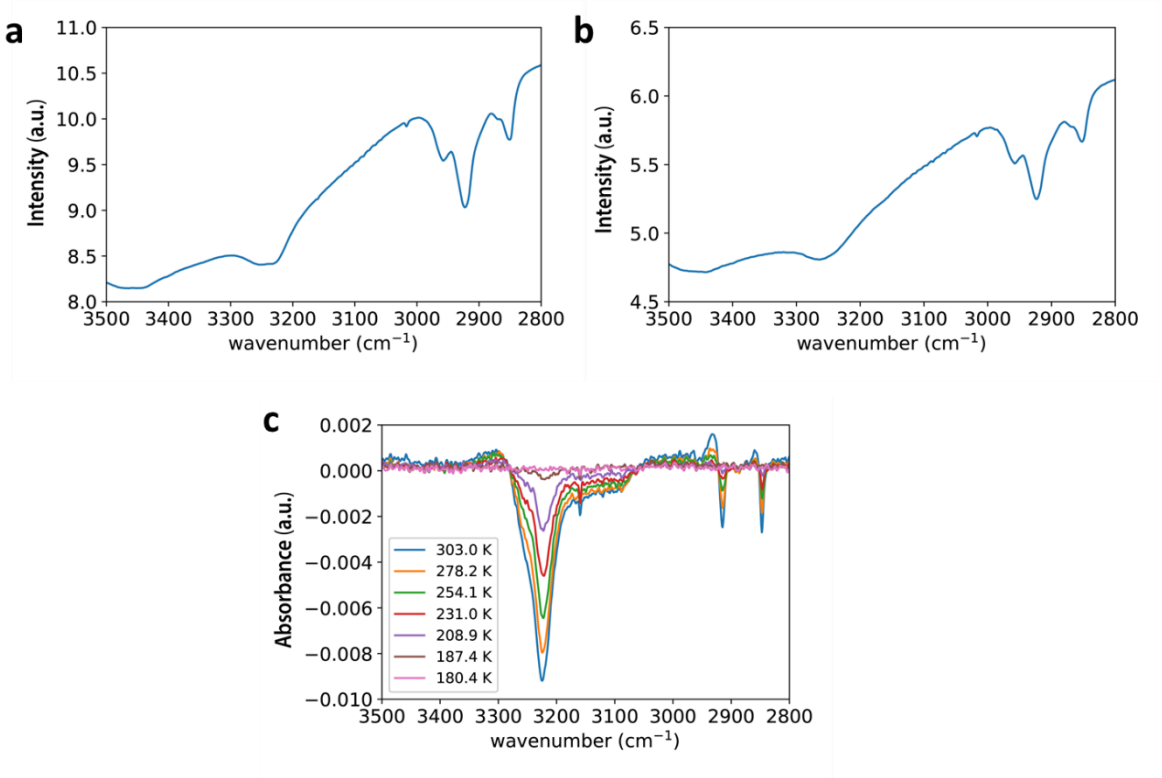


Figure S4. TPD-IR tests of water adsorption. (a) Background spectra for pristine graphene. (b) Background spectra for 60 s hydrogenated graphene. (c) IR spectra during cooling down from room temperature to 180 K. The peak around 3250 cm^{-1} indicates water desorbed from H-graphene surfaces.

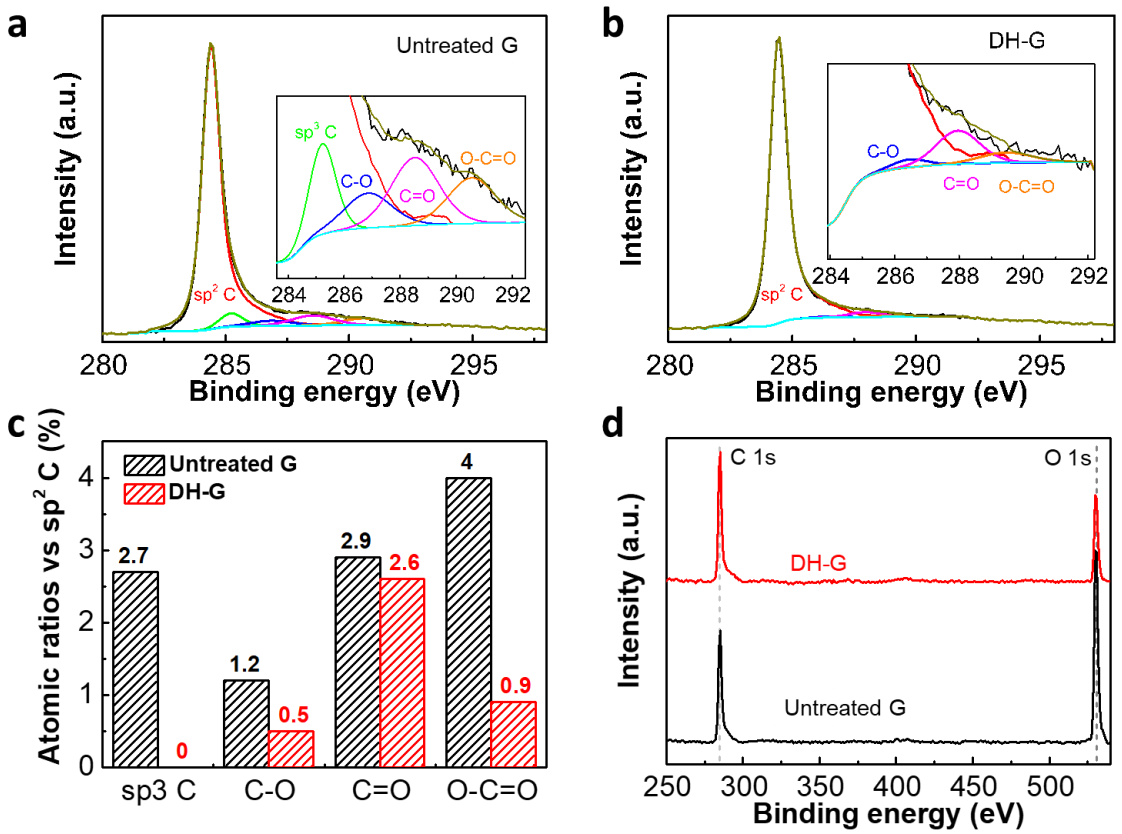


Figure S5. XPS characterization of dehydrogenated graphene. (a) C1s core spectra of G. (b) C1s core spectra of DH-G. (c) Atomic ratios of sp^3 C, C-O, C=O and O-C=O versus sp2 C for G and DH-G. (d) Survey spectra of C1s and O1s for G and DH-G.

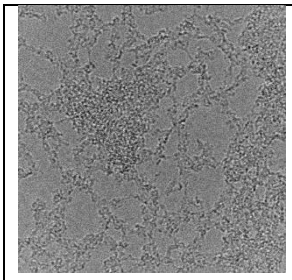
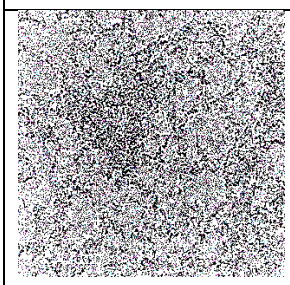
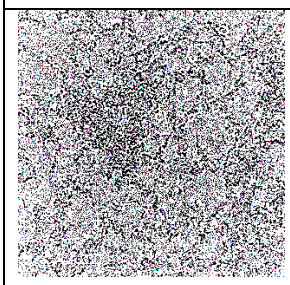
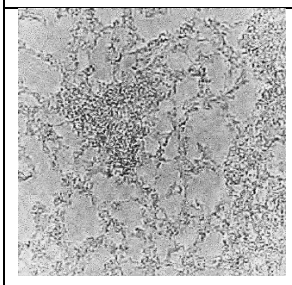
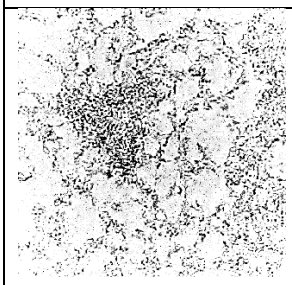
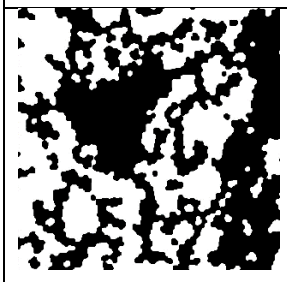
Supplementary Note 1:

MATLAB script to calculate the cleanness:

```
%% Initialization

N = 50; % number of the images to be analyzed.
level= 0.50; % bw conversion treshold value.
size = 10;

report = zeros (N , 3);
shape = 'disk';

for sequence = 1: N
   filename = num2str(sequence);
   im = imread([filename, '.tif']);

   image = rgb2gray(im); %rgb to gray conversion

   IniImage = image;

   %% improving the image

   image = adapthisteq(image); % increasing the image contrast

   image = uint8(255*mat2gray(image)); % normalization of the image


   %% Noise removal

   image = imclose(image,strel(shape,1));
   image = imopen(image,strel(shape,1));

   image = imgaussfilt(image, 2);

   %% conversion to a binary mage

   T = adaptthresh(image,level,'ForegroundPolarity','dark');
   image = imbinarize(image, T);


   %% segmentation

   image = imopen(image, strel(shape,size));
   image = imclose(image, strel(shape,size));

   %% finalizing
   clean = sum(sum(image == 1));
   total = numel(image);

   report(sequence, 1) = clean;
   report(sequence, 2) = total;
   report(sequence, 3) = clean*100/total;

   Figure., imshowpair(IniImage, image, 'montage'), title([filename, ': cleanless:',
num2str(report(sequence, 3)), '%,', ', size:', num2str(size), ', level: ', num2str(level)]);
   saveas(gcf,[filename, '_3rd_analyzed.jpg']);

   clear image
```

```
    clear im
    clear IniImage
    clear Image
end

Figure., bar(report(:, 3));
Figure., hist(report(:, 3));
Clenaliness = mode(report(:, 3));
display(Clenaliness);
```

Table S1. Process flow for analyzing the images

	**Step 1: Conversion into a grayscale image** The TEM image is converted into a grayscale image using the MATLAB built-in function rgb2gray().
	**Step 2: Adjusting of the contrast** The goal of this step is to increase the contrast of the image and improve the visibility of small contaminations. Built-in adapthisteq() function of the MATLAB performs this task by contrast-limited adaptive histogram equalization.
	**Step 3: Normalization of the image** The goal of this step is to eliminate the effect of any intensity variations from image to image. The intensity of each pixel of the image after this transformation (y) is calculated by: $y = 255 \times \frac{(x-min)}{(max-min)}$, where: x: the gray scale initial intensity of the pixel min: minimum gray scale intensity for the original image. max: maximum gray scale intensity for the original image.
	**Step 4: Denoising of the image** Several MATLAB built-in functions including imopen(), imclose(), imgaussfilt() are used to remove the noise. Opening and closing operations remove random flipped pixels. The imgaussfilt() smoothens the image by filtering with a 2-D Gaussian function.
	**Step 4: Conversion the *gray-scale* into the *binary* image** With this conversion, all the pixels with the intensity greater than a threshold, calculated by the adaptthresh() function will be replaced by 1 (white color, corresponding to the clean graphene) and the pixels with lower intensities will be replaced by 0 (black, corresponding to contaminations). This operation is performed using the MATLAB built-in function imbinarize().
	**Step 5: Segmentation** By using the built-in open()/close() functions (subsequent erosion and dilation operations) the extend of the contaminations is identified. A disk with the radius of 10 pixels is used as the structuring element to define the neighborhood. The cleanness of the image is eventually calculated as the sum of the number of the white pixels (clean graphene area) normalized by the total number of pixels.