accounted to the chemical reduction of gold ions to Au^0 by photoreduction or the functional groups, and precipitation of gold ions with the adsorbents.

These gold adsorbents exhibit a high gold extraction capacity at a gold concentration from 500 ppm-3000 ppm[12–22], but this capacity decreases to less than 250 mg/g at a gold concentration range relevant to e-waste recycling, specifically, from ppb level to tens of ppm[4,12,19]. Furthermore, as the e-waste contains complex co-existing metal elements, the practical gold extraction process often requires good extraction selectivity to gold, so that separation of co-existing elements from the extracted gold can be avoided, achieving a high resource- and energy- efficiency for gold recycling. The existing novel gold adsorbents have demonstrated good selectivity to gold, but their adsorption to the co-existing metal elements is still unneglectable[4,12,15,16,19]. Therefore, the development of materials with high gold extraction capacity to trace the amount of gold, precise gold selectivity and economic viability remains lacking.

Here we report an exceptionally high gold extraction capacity of chemically reduced graphene oxide (rGO), reaching 1850 mg/g and 9059 mg/g when extracting gold from its 10 ppm solution at 25 °C and 60 °C, respectively, combined with an ability to extract gold at minute concentrations, down to parts per trillion and high selectivity. During extraction, rGO reduces >95% gold ions to metallic gold, avoiding elution and precipitation necessary in post-adsorption processing. Moreover, gold extraction can be done selectively, without adsorption of the other 14 elements normally present in e-waste. This, in turn, enables the recycling of copper, the second most valuable metal in e-waste. Finally, we demonstrate a highly efficient flow-through process for gold extraction using rGO membranes. Our findings show a promising venue for addressing global e-waste challenges and gold scarcity.

Results and Discussion

Highly efficient gold extraction by rGO

The rGO was obtained by chemical reduction of commercial, mass-produced graphene oxide (GO) nanosheets using ascorbic acid as a reductant (Methods). The resulting rGO nanosheets were repeatedly washed with deionized water, until a stable colloid was formed, and then the rGO suspension was added directly into solutions containing gold ions (Fig. 1a). Considering the hydrochemical process is a flexible, low-cost, and sustainable method for e-waste recycling[23,24], and $[AuCl_4]^-$ is a common gold complex seen in a hydrochemical process[4,7,12], we have chosen $KAuCl_4$ solutions prepared in different concentrations (C) to examine the gold extraction capacity of rGO (Fig. 1a, b); Similar to the behaviour observed for other adsorbents[4,13,14], the extraction capacity increased with increasing C and reached a plateau above 10 ppm (Fig. 1b). Specifically, 1 g of rGO can extract ~ 690, 1180 and 1850 mg of gold from 0.1, 1 and 10 ppm gold solutions, respectively. This extraction capacity outperformed those of novel nanoporous adsorbents at the same concentrations and, importantly, extends to solutions containing only 1 ppb of gold (Supplementary Note 1; Supplementary Fig. 1 and Supplementary Table 1). Although we could not quantify the extraction capacity for even lower C (our quantitative methods reached their detection limit), we still observed significant gold extraction down to as little as 10 ppt (Supplementary Fig. 1). This makes the reported rGO technique particularly interesting because wastewater and freshwater often contain gold with C below 10 ppb, and its efficient reclaim is challenging[12].

When measuring the extraction capacity of rGO to different pH from 2 to 11, we found the extraction capacity varied with the pH, which is not unusual and similar to many gold adsorbents[4,13–16,19–21]. As shown in Supplementary Fig. 2, the maximum gold uptake was found at pH ≈ 4. This is attributed to the fact that, at lower pH, the rGO colloid loses its stability[25,26] whereas, at basic pH, rGO undergoes deprotonation, becomes negatively charged[4] and starts repelling negatively charged $[AuCl_4]^-$. Both effects decrease the extraction capacity. It is important to note that the extraction capacities at the acidic and basic solution are still higher than the previously reported gold extraction capacities at the same C (Supplementary Figs. 1 and 2), and by increasing the amount of rGO, ~99% gold ions from its 10 ppm solution with the pH of 2–11 can be extracted (Methods and Fig. 1b). We also investigated the gold extraction as a function of time (Fig. 1c and Supplementary Fig. 3). Within 10 min, 1 g of rGO provided extraction of ~1010 and 325 mg of gold from 10 ppm and 100 ppb solutions, respectively (Fig. 1c), suggesting not only efficient but also rapid extraction. Interestingly, after extraction to 1 ppm gold, rGO changed its colour from black to gold within hours, suggesting almost instantaneous Au recovery (Supplementary Movie 1).

Mechanism study of rGO's gold extraction behavior

To understand the ultrahigh gold extraction behavior of rGO, we started with the characterization of rGO after the gold extraction. We found nanoparticles on top of rGO nanosheets after 24 h of its exposure to gold solution (Fig. 2a). X-ray diffraction (XRD) showed this particulate to be metallic gold, suggesting that $[AuCl_4]^-$ was reduced to Au^0 (Supplementary Fig. 4). This was confirmed by X-ray photoelectron spectroscopy (XPS) analysis (Fig. 2b) which showed that >95% of the particulate was metallic Au^0 rather than $[AuCl_4]^-$. Thermal analysis (Supplementary Note 2; Supplementary Fig. 4) provided good agreement with the XPS results, as no endothermic peak for the

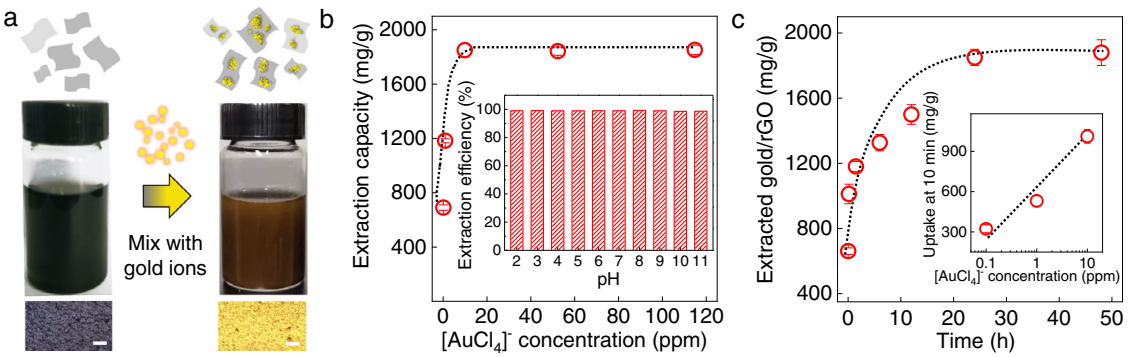

Fig. 1 | High-efficiency extraction of trace amounts of gold. a Schematic of the extraction process using rGO. After mixed with gold ion (10 ppm) for 12 h, rGO suspension gradually changed its color from black to brown. The bottom panels are the optical images of rGO films deposited from rGO nanosheets before (black) and after gold extraction (gold). Scale bar: 20 μm. **b** Extraction capacity as a function of gold concentration after 24 h. The inset plots the extraction efficiency measured for 10 ppm solutions at different pH. **c** Extraction capacity as a function of time for a 10 ppm gold solution. Inset: Extraction from different solutions after 10 min. Dashed lines, guides to the eye. All the experiments were performed at 25 °C. All the error bars in this figure represent the standard deviation.