Telluriding monolayer MoS$_2$ and WS$_2$ via alkali metal scooter

The conversion of chalcogen atoms to other types in transition metal dichalcogenides has significant advantages for tuning bandgaps and constructing in-plane heterojunctions; however, difficulty arises from the conversion of sulfur or selenium to tellurium atoms owing to the low decomposition temperature of tellurides. Here, we propose the use of sodium for converting monolayer molybdenum disulfide (MoS$_2$) to molybdenum ditelluride (MoTe$_2$) under Te-rich vapors. Sodium easily anchors tellurium and reduces the exchange barrier energy by scooting the tellurium to replace sulfur. The conversion was initiated at the edges and grain boundaries of MoS$_2$, followed by complete conversion in the entire region. By controlling sodium concentration and reaction temperature of monolayer MoS$_2$, we tailored various phases such as semiconducting 2H-MoTe$_2$, metallic 1T'-MoTe$_2$, and 2H-MoS$_2$$_{2-x}$Te$_x$ alloys. This concept was further extended to WS$_2$. A high valley polarization of ~37% in circularly polarized photoluminescence was obtained in the monolayer WS$_2$$_{2-x}$Te$_x$ alloy at room temperature.
wo-dimensional monolayer transition metal dichalcogenides (TMDs) have unique growth behaviors that are different from bulk. While chemical vapor deposition (CVD) is commonly introduced to grow various types of TMDs\(^1\)–\(^3\), the conversion process is also available by the substitutional reaction of chalcogen atoms\(^4\)–\(^6\). For example, monolayer molybdenum disulfide (MoS\(_2\)) is converted to molybdenum diselenide (MoSe\(_2\)) by a heat treatment under Se-rich conditions or vice versa\(^7\). This method is useful for doping, alloying, spin-orbit coupling (SOC) engineering\(^8\), and generating heterointerfaces by selective patterning\(^9\).

Difficulty exists, however, in the Te-conversion case due to the low decomposition temperature of tellurides and the lower activity of Te atoms with transition metals than that of sulfur or selenium atoms. Recently, molybdenum ditelluride (MoTe\(_2\)) and tungsten ditelluride (WTe\(_2\)) have been intensively investigated due to their unique phase engineering\(^10\)–\(^11\) and novel physical nature of Weyl semimetals and topological insulators. Moreover, MoS\(_2\)–Te\(_x\) alloy, 2H, and 1T–MoTe\(_2\) phase by adjusting tellurization parameters and construct a phase modulation diagram with Na-scooter concentration and telluriding temperature. The bandgap and SOC can be engineered by Te composition. The partially tellurized WS\(_2\) sample reveals high valley polarization up to 37% at room temperature.

**Results**

**Telluriding molybdenum disulfide by sodium-assisted tellurization process.** Figure 1a is a schematic of the sodium-assisted tellurization process for MoS\(_2\). A sodium hydroxide (NaOH)-coated substrate was brought into a chamber with a CVD-grown monolayer MoS\(_2\) sample and the two were placed facing each other in the tellurization zone (see Methods section and Supplementary Figure 1). The Na\(_2\)Te, which is most probable Na–Te compound, converted from NaOH while supplying a Te vapor (Supplementary Figure 2 and Supplementary Note 1) is scooted to the MoS\(_2\) substrate to exchange Te atoms with S atoms (Fig. 1b). Other compounds such as NaTe and NaTe\(_3\) were converted to Na\(_2\)Te under Te-rich conditions.

The Na\(_2\)Te, which is regarded as the most probable Na–Te compound from sodium hydroxide, plays a role as the Te carrier and catalyst for exchanging with S atoms. After exchanging S atoms with Te atoms, Na\(_2\)S is converted to Na\(_2\)Te under Te-rich conditions. Gibbs free energy changes for conversion without/with Na-scooter. ΔG is reduced to around 70 kJ mol\(^{-1}\).

**Fig. 1** Alkali-metal-assisted conversion from MoS\(_2\) to MoTe\(_2\). **a** Schematic of the Na-assisted tellurization process. MoS\(_2\) was flipped over to face the NaOH-coated substrate. **b** Illustration for the conversion step from Mo–S to Mo–Te via a Na-scooter. Na\(_2\)Te, which is regarded as the most probable Na–Te compound from sodium hydroxide, plays a role as the Te carrier and catalyst for exchanging with S atoms. After exchanging S atoms with Te atoms, Na\(_2\)S is converted to Na\(_2\)Te under Te-rich conditions. **c** Gibbs free energy changes for conversion without/with Na-scooter. ΔG is reduced to around 70 kJ mol\(^{-1}\). **d** Raman spectra (d) and XPS taken for Mo, S, and Te (e) of partially tellurized MoS\(_2\)–Te\(_x\) without/with Na-scooter at 600 °C for 30 min.
negligibly formed during tellurization of NaOH (Supplementary Figure 2c). In fact, Na₂Te is a well-established compound as an efficient tellurizing reagent. Due to its higher reactivity of Na₂Te than pure Te, Na₂Te is widely used for synthesizing various telluride compounds.

The reduction in the barrier height for the proposed conversion reaction via the Na-scooter (Na₂Te) is understood by calculating the Gibbs free energy (Fig. 1c and Supplementary Table 1). In the presence of a Na-scooter (or efficient carrier), the conversion of MoS₂ to MoTe₂ requires around 70 kJ mol⁻¹ less Gibbs free energy (equivalent to 300 °C or 0.73 eV by the Readhead equation) than a conversion without Na. It can also be assumed that the reaction temperature at 900 °C without a Na-scooter is reduced to 500 °C in the presence of a Na-scooter (black line). This temperature reduction for tellurization is crucial to stabilize the MoTe₂, since the decomposition temperature of MoTe₂ is around 700 °C (Supplementary Table 2). In this sense, Na-scooter plays as both an efficient Te carrier and catalyst. Although the Gibbs free energy is still positive, such a reaction could occur under Te-rich conditions at a reasonable temperature. The formed Na₂S could be converted back to Na₂Te under Te-rich conditions (bottom-orange curve), guaranteeing continuous tellurization. There is still a trace of conversion to MoTe₂ even without a Na-scooter over 650 °C; however, the converted MoTe₂ would be etched and dissociated (Supplementary Figure 3).

The proposed mechanism is confirmed by Raman spectroscopy and X-ray photoemission spectroscopy (XPS) for the tellurized MoS₂ prepared with/without NaOH. The A₁g and E₁2g modes of the 2H-MoS₂ phase observed near 402 and 383 cm⁻¹, respectively, in the absence of a Na-scooter, are identical to those of the pristine monolayer MoS₂. Meanwhile, the E₁2g (240 cm⁻¹) mode of 2H-MoTe₂ and some unknown peak (near 350 cm⁻¹ in the red-dotted circle) clearly appeared with the Na-scooter (bottom). The remaining MoS₂ peak is red-shifted by 9 cm⁻¹ for E₁2g, indicating a compressive strain in the MoS₂ (see supplementary Figure 6 for more information). The peak shift is negligible for A₁g, indicating no appreciable charge.

Fig. 2 Time evolution of conversion from MoS₂ to MoTe₂. a Optical images and the corresponding schematics of the initial growth stages: 5, 10, and 15 min. The dark color indicates the fully converted MoTe₂ region from MoS₂. Conversion is favored at the edges and grain boundaries; scale bars, 50 μm. b Amplified optical image of the sample grown for 15 min. The MoS₂ flake is completely converted to MoTe₂ in the yellow-dotted circle; scale bars, 50 μm. c, d XPS mapping image of tellurized MoS₂ flake for S 2p3/2 (c) and Te 3d5/2 (d) in the white-dotted square in b; scale bars, 30 μm. A high Te content was detected in the dark region (no sulfur content was detected), while the S content (~20%) is seen in the bright region. e Raman spectra of bright and dark regions. The dark region shows the Raman spectrum exclusively from MoTe₂ (1T’—2H mixed phase). Some unknown peaks (349 and 435 cm⁻¹ in red circles) in the bright region, which are far from MoS₂ and MoTe₂ Raman peaks are observed.
transfer. This implies that the obtained sample is in a form of the MoS$_2$−$x$Te$_x$ alloy.

More distinct features of the conversion were demonstrated from XPS analysis. While Te peaks were absent from the sample without a Na-scooter, clear Te peaks were identified with red-shifted (0.4 eV) Mo 3d peaks in the Na-assisted tellurized sample (Fig. 1e). The lowered Fermi level indicates electron withdrawal.

Tellurium oxide peaks were also visible, indicating that the sample is easily oxidized under ambient conditions.

Time evolution of tellurization process. Figure 2a displays the optical images and corresponding schematics for the time evolution of tellurization for MoS$_2$. No change in the optical contrast is seen in the sample tellurized for 5 min compared with pristine MoS$_2$. When the tellurization time is prolonged to 10 min, dark-colored regions start emerging mostly at the edges and grain boundaries. The exposed dangling bonds at the edge and some defect sites such as grain boundaries and S vacancies are known to have higher reactivity than the basal surface of MoS$_2$. In this sense, tellurization takes place preferentially at the edge and grain boundaries in MoS$_2$. The area of the dark regions is diffused and widened from the edge to the entire area of the flakes at 15 min of tellurization (yellow-dashed circle in Fig. 2b). If the flake size is large, the longer reaction time is needed for full conversion (white-dashed box in Fig. 2b).

To analyze the chemical composition in conjunction with the optical contrast, we conducted XPS mapping for a sample tellurized for 15 min (white-dashed square, Fig. 2b). The outer region shows a relatively dark optical contrast compared to the inner region. Figure 2c and d shows XPS mapping images for S 2p$_3/2$ and Te 3d$_5/2$ (Supplementary Figure 4). S 2p$_3/2$ peaks were detected only in the bright region, while Te 3d$_5/2$ peaks were rich in the dark region. The calculated chemical compositions of Te/(S + Te) are 0.8 and 1 for the bright region and dark region,
respectively (Supplementary Figure 5). The negligible sulfur content in the dark region indicates that MoS2 is fully converted to MoTe2, while the bright region still has remaining sulfur atoms.

A similar trend is also observed in the Raman spectra (Fig. 2e). The bright region reveals the MoS$_2$–$x$Te$_x$ alloy peaks with a MoTe$_2$-like peak near 227 cm$^{-1}$, a MoS$_2$-like peak near 395 cm$^{-1}$ and unknown peaks in red circles. To clarify the peak near 227 cm$^{-1}$, the peak was deconvoluted to LA(M) mode for 2H-MoS$_2$ and E$_{2g}$ mode for 2H-MoTe$_2$ (more details are discussed in Fig. 3g and Supplementary Figure 6). Meanwhile, the dark region shows a uniquely MoTe$_2$ feature.

**Tellurization of molybdenum disulfide in atomic scale.**

To study the tellurization of MoS$_2$ at the atomic scale, we conducted transmission electron microscopy (TEM). Figure 3a is the low-magnification TEM image of a tellurized MoS$_2$ flake grown at 625 °C and NaOH concentration of 1 μmol cm$^{-2}$ for 30 min. Electron diffraction patterns of the hexagon (Fig. 3b) are confirmed for each region indicated in Fig. 3a. Two features are noted here: (i) at low Te loading content, MoS$_2$–$x$Te$_x$ alloy still maintains single crystallinity within the inner region of triangular flakes (marked by number 1, 2, and 3). (ii) Additional hexagon emergent in region 3 (expanded pattern in Fig. 3c) with a lattice constant of 0.34 nm for a lattice, equivalent to a MoTe$_2$ phase in addition to the MoS$_2$ phase ($a = 0.306$ nm), while maintaining the same orientation as MoS$_2$. When more Te content is converted (region 4) (i.e., on both sides), MoTe$_2$ may suffer from local strain, deviating the orientation of the hexagon.

Three different phases of MoS$_2$, MoS$_2$–$x$Te$_x$, and MoTe$_2$ are schematically drawn in the top panel (Fig. 3d). The annular dark field scanning TEM (ADF-STEM) image (middle panel) reveals three such types of intensity distributions; the brightest intensity spot in the chalcogen site corresponds to Te–Te (red circle), the middle intensity corresponds to Te–S (yellow circle), and the dark spot corresponds to S–S species (white circle). Such species were identified by comparing each intensity to the Mo intensity (bottom panel and
2H-MoTe₂. The threshold voltage in the MoS₂-Te₂ alloy (red) is shifted positively from pristine MoS₂ (green). The p-type behavior is shown for 2H-MoTe₂ (blue).

 Supplementary Figure 7). The false-colored fast-Fourier transform (FFT) filtered image (Fig. 3e) of region 1 clearly demonstrates a few spots of Te–S and one Te–Te spot. Meanwhile, at spot 3, more Te–Te species were emergent with a large portion of Te–S species. In this case, the intensity of Mo atoms adjacent to Te atoms is exaggerated by artificial filtering (Supplementary Figure 8).

It is not clear from STEM analysis if Te atoms in MoSₓTe₁ are located on the top or bottom of Mo layer. There are two possible arrangements of Te–S species in MoSₓTe₁ structure during conversion from MoS₂ to MoTe₂ (Fig. 3f). One is called Janus phase, where S atoms at the top layer is replaced by Te atoms while retaining the bottom S layer, which is consistent with recently reported phase of MoSₓTe₁. In addition, there is another possibility of combination, called randomly distributed phase. The total Te coverage is still half but part of Te atoms are located on top and bottom layer, distinguished from Janus phase. The corresponding Raman spectrum for MoSₓTe₁ alloy is provided with calculated vibrational modes of Janus phase and randomly distributed phase in Fig. 3g. Although the theoretical peaks calculated from density functional theory (DFT) represent experimental peaks to a some degree, the precise assignment of the spectra to either Janus or random phase is premature at the moment (more detail in Supplementary Figure 6). This ambiguity is partly ascribed to insufficient coverage of MoSₓTe₁, as shown in Fig. 3e. The intermediate phase could be an interesting phase that reveals piezoelectric properties and requires further studies. The bandgap tuning window by the tellurization of MoS₂ from 2.14 to 1.1 eV (see Supplementary Figures 9–11) is nearly twice that of MoS₂-xSeₓ alloys or Mo₁-xWₓS₂ alloys.

Temperature-dependent tellurization behavior. Conversion temperature is another sensitive variable for conversion kinetics. As the tellurization temperature increases, Te content increases gradually and reaches maximum (MoTe₂) at 650 °C (Supplementary Figure 12). More interestingly, tellurization occurs more dominantly from the edge (and grain boundaries) at relatively low temperature, while this occurs on the entire surface of MoS₂ flakes at high temperature to form MoSₓTe₁ alloys, as shown in the schematic of Fig. 4a. Figure 4b illustrates optical images of tellurized monolayer MoS₂ flakes at two representative temperatures (550 °C and 625 °C). There are two distinct regions in terms of the optical contrast (bright and dark) in the tellurized MoS₂ samples. The corresponding Raman spectra are provided in Fig. 4c. At 550 °C, the bright regions show unaltered 2H-MoS₂ peaks, whereas the dark regions reveal semiconducting 2H-MoTe₂ peaks. At 625 °C, the bright regions reveal MoSₓTe₁ alloy peaks, whereas the dark regions exhibit mixed semiconducting 2H phase and metallic 1T′ phase. This indicates that semiconducting 2H-MoTe₂ formed near the edge is favored at low temperature and metallic 1T′-MoTe₂ is favored at high temperature, reflecting the bulk phase stability. Photo-luminescence (PL) is conducted for further characterization (Fig. 4d). The PL spectrum of the bright regions at 550 °C sample exhibits emission from pristine MoS₂ (650 nm), while the bright regions at 625 °C reveal an alloy peak at 1.72 eV (720 nm).
We also note that a sharp interface at the edge distinguished by optical contrast is formed at low temperature, while at high temperature, a mixed alloy is preferably formed. At low temperature, tellurization is preferentially initiated at the reactive sites such as the edge and grain boundaries. In that regime, more dominant enthalpic contribution than entropic contribution to Gibbs free energy results in phase segregation to minimize its interfacial energy. Meanwhile, at high temperature, the entropic contribution is dominant and Te atoms have sufficient thermal energy to overcome tellurization energy barrier randomly in the entire MoS\textsubscript{2} basal surface. This temperature-dependent behavior is also similar to the previous report on the substitution of Mo atoms in WS\textsubscript{2}.\textsuperscript{23}

**Fig. 6** Formation of MoS\textsubscript{2}/MoTe\textsubscript{2} lateral junction and WS\textsubscript{2}−xTe\textsubscript{x} alloy. a Schematic of 2H-MoS\textsubscript{2}/2H-MoTe\textsubscript{2} in-plane heterojunction. b-e Optical micrograph (b); scale bar, 20 μm, confocal Raman mapping of E\textsubscript{1g} 2H-MoTe\textsubscript{2} (c), and 2H-MoS\textsubscript{2} (d) for MoTe\textsubscript{2}/MoS\textsubscript{2} junction; scale bars, 50 μm and the corresponding diode I-V curve (e). A clear diode behavior with the band diagram of the MoTe\textsubscript{2}/MoS\textsubscript{2} heterostructure reveals a type-II junction. For electrical measurements, source (MoTe\textsubscript{2}) and drain (MoS\textsubscript{2}) electrodes were fabricated by depositing 5/50 nm Cr/Au. f Optical micrograph of tellurized monolayer WS\textsubscript{2} flakes. WTe\textsubscript{2}/WS\textsubscript{2}−xTe\textsubscript{x} junction; scale bar, 20 μm. g Raman spectra from each region of a and b in f. The Raman spectrum of 2H-WS\textsubscript{2} is also provided for comparison. h Circularly polarized PL spectra of 2H-WS\textsubscript{2} and the WS\textsubscript{2}−xTe\textsubscript{x} alloy (at 300 K) excited by σ\textsuperscript{+} polarized light at energy of 2.33 eV. The PL is red-shifted to ~0.1 eV and valley polarization is enhanced up to 37% in the WS\textsubscript{2}−xTe\textsubscript{x} alloy. Valley polarization (P\textsubscript{c}) is defined as P\textsubscript{c} = \frac{I(\sigma\textsuperscript{+}) - I(\sigma\textsuperscript{−})}{I(\sigma\textsuperscript{+}) + I(\sigma\textsuperscript{−})}.

**Phase modulation with NaOH concentration and temperature.** To study the effect of tellurization temperature and NaOH concentration systematically, a series of experiments were done and summarized in Figs. 5a and b. Figure 5a displays the representative Raman spectra of the tellurized MoS\textsubscript{2} samples. Four representative types of phases were identified. In the top panel, a 2H-MoS\textsubscript{2}-like feature is shown with two distinct A\textsubscript{1g} and E\textsubscript{1g} peaks of MoS\textsubscript{2} and no additional peaks, although the precise positions of such peaks vary slightly with composition. The second panel shows the Raman peaks of the MoS\textsubscript{2}−xTe\textsubscript{x} alloy. The third and bottom panels represent 2H-MoTe\textsubscript{2} and 2H–1T′-mixed MoTe\textsubscript{2} phases, respectively, identified by the E\textsubscript{1g} peaks of 2H-MoTe\textsubscript{2} and the B\textsubscript{g} peaks for 1T′-MoTe\textsubscript{2} (black-dotted line). Note that the 2H semiconducting phase is retained after the full conversion.
Figure 5b shows the phase modulation of MoS2 with temperature and Na content determined by Raman spectra and optical images (Supplementary Figure 13). The tellurization rate increases in proportion to NaOH concentration and tellurization temperature. The conversion ratio is estimated by the area of dark region where MoS2 is fully converted to MoTe2 in optical images (Supplementary Figure 13a), which is expressed in the phase modulation by the gray color scale. In the temperature range from 525 °C to 575 °C, two phases of 2H-MoS2 and 2H-MoTe2 coexist due to the edge-selective tellurization. From 600 °C to 650 °C, both edge and surface are tellurized, resulting in the formation of MoS2−xTex alloy and fully converted 1T−2H MoTe2.

Figure 5c−d are confocal Raman mapping images of the intensity and full width at half maximum (FWHM) for E12g mode from fully converted samples of 2H-MoTe2 (yellow-dotted circle in Fig. 5b). The uniform contrasts of both images indicate reasonable sample uniformity while retaining a monolayer morphology without fragmentation in μm scale. The FWHM value for E12g mode is around 6.6 cm−1 (Supplementary Figure 14), indicating that the converted MoTe2 has reasonable crystallinity compared to exfoliated one (5.0 cm−1).44 Tellurization was done with good uniformity in mm. The uniformity of MoS2−xTex alloy and 1T−2H mixed phase MoTe2 flakes are characterized by Raman mapping and the uniformity dependence on positions are also characterized by Raman spectroscopy (Supplementary Figures 15 and 16).

One interesting feature is that the proposed conversion provokes not only an alloying effect but also a carrier-type conversion. Figure 5e shows the transfer characteristics of MoS2, MoS2−xTex, and 2H-MoTe2, and the pristine MoS2 clearly shows an n-type behavior. With the MoS2−xTex alloy, an n-type behavior is still retained, but the threshold voltage is upshifted significantly, indicating a p-type doping effect. Completely converted MoTe2 clearly demonstrates a p-type behavior; in other words, the carrier type is converted from electron to hole. It is worth mentioning that Na contamination gives n-type doping effect in MoS2 FET (Supplementary Figure S17). In our case, Na compounds are washed out during transfer. Therefore, the p-type doping effect in tellurized MoS2 originates exclusively from Te substitution, not from Na contamination.

Lateral heterojunction and tellurization of WS2. We further provide a few more examples to demonstrate the strength of our efficient Na-assisted tellurization. We generate 2H-MoTe2/2H-MoS2 in-plane heterojunction by telluridizing MoS2 at 550 °C (Fig. 6a). The optical micrograph reveals a clear contrast between the edge and inner regions (Fig. 6b). The confocal Raman mapping of the E12g peak intensity of MoTe2 was identified at the edge (Fig. 6c), whereas that of E12g of MoS2 was identified at the inner region (Fig. 6d). The sharp interface between MoTe2 and MoS2 is attributed to energy minimization by reducing interfacial energy, which is confirmed by the PL measurement (Supplementary Figure 18). The diode was fabricated across the interface via electron beam lithography. A clear rectifying behavior is demonstrated due to a type-II junction formation (Fig. 6e) (band diagram shown in inset).

Na-assisted tellurization is also possible for tungsten disulfide. The similar optical contrast at different regions was observed in the tellurized WS2 sample (Fig. 6f). Raman spectra for regions (a) and (b) are presented with pristine WS2 for comparison (Fig. 6g). While the inner region (a) reveals an alloy phase of WS2−xTe, the outer region (b) clearly shows Raman modes for Td′-TeW2.25 Due to the semi-metallic nature of Td′-TeW2,26 no PL was observed in the region (b). On the other hand, the alloy phase of WS2−xTe2 shows a PL at 1.93 eV (2.03 eV for pristine WS2) (Fig. 6h).

It has been reported that valley polarization can be tuned by doping27, defect28, and alloying engineering.3 Especially, alloying can modify the valley polarization by engineered SOC. Therefore, it is intriguing to observe valley polarization of tellurized WS2 by circularly polarized PL. We define the degree of valley polarization, Pσ, as follows:

\[
P_{\sigma} = \frac{I(\sigma+) - I(\sigma-)}{I(\sigma+) + I(\sigma-)},
\]

where \(I(\sigma\pm)\) is the intensity of the left (right) circularly polarized PL. No appreciable polarization was observed from pure WS2 at room temperature with a 2.33 eV excitation as seen in another report29 (top panel). On the other hand, a large valley polarization of ~37% is observed in the alloy which can be further enhanced by modifying the resonance radiation.30-32 The enhancement of valley polarization can be attributed to two reasons: (i) enhanced SOC strength and (ii) inversion symmetry breaking by Te–S species. High SOC strength of Te atom can extend the spin-orbit splitting (\(\Delta_{SO}\) (TeW2): 484 meV, \(\Delta_{SO}\) (WS2): 412 meV).33 We identified Te–S species in WS2−xTe alloy by STEM and simulation (Supplementary Figure 19). The Te–S species affect the out-of-plane \(d_{xz}\) orbitals and induce Rashba spin splitting. This can be another reason for enhanced valley polarization value20.

Discussion

In conclusion, we have realized a facile route for tellurization via a Na-scooter. The activation barrier height for conversion of WS2 to MoTe2 (M = Mo, W) is dramatically decreased by Na-scooter, resulting in tellurization happening at relatively low temperatures where tellurides are stable. Due to emulating its host material’s nature in the conversion process, we synthesized monolayer 2H-MoTe2 for the first time. Furthermore, the bandgap tuning window has enlarged to 1.0 eV (2.14−1.1 eV). Finally, edge-selective tellurization and modified SOC by Te content can realize two-dimensional heterojunctions and enhance the degree of valley polarization. Our approach will open up new avenues to explore intriguing physics phenomena of tellurides.

Methods

Synthesis of monolayer MoS2 and WS2 by CVD. Monolayer MoS2 and WS2 were grown on SiO2/Si wafers by atmospheric pressure CVD. A water-soluble metal precursor was coated on the SiO2/Si substrate first. The precursor solution was prepared by mixing three types of chemical solutions (defined as A, B, and C).

A (Metal precursor): 0.1 g of ammonium heptamolybdate (Sigma-Aldrich, 431346) and 0.2 g of ammonium metatungstate (Sigma-Aldrich, 463922), were dissolved in 10 mL of deionized (DI) water for Mo and W precursors, respectively.

B (Promoter): 0.3 g of sodium chloride hydrate dissolved in DI water (Sigma-Aldrich, C6445) was introduced for promoting monolayer TMDCs.

C (Medium solution): An OptiPrep density gradient medium (Sigma-Aldrich, D1556, 60% (w v−1) solution of iodixanol in water) was adopted as a medium. It does not affect the growth of TMDCs but allows for a better spin-casting process due to its high viscosity.

The precursor solution in which A, B, and C were mixed with the ratios of 2, 6, and 1, respectively, was coated onto the SiO2/Si wafer by spin casting at 3000 r.p.m. for 1 min. The precursor-coated substrate and 0.2 g of sulfur (Sigma, 344621) were separately introduced to a two-zone furnace. The sulfur zone was heated up to 210 °C at a rate of 50 °C/min. The substrate zone was set to 800 °C. Temperature profiles in both cases (MoS2 and WS2 growth) were the same, but 5 sccm of hydrogen gas was introduced only for the WS2 case with 500 sccm of nitrogen as a carrier gas.

Sodium-assisted conversion process. NaOH of 0.1 g was dissolved in 60 mL of DI water. This NaOH solution of 25−100 μL was dropped on a 1 × 1 cm2 sapphire substrate and dried in an oven (NaOH concentrations were varied from 1.0−4.0 μm2 cm−2). The NaOH-coated substrate and MoS2 on the SiO2/Si wafer were placed on the ceramic crucible (1.5 × 5 W × 5 H 1.5 cm−3). The distance between the MoS2 and NaOH-coated substrate was fixed to 2 mm by placing the
NaOH substrate 1 cm above the supporter, in which the MoS2 substrate faced the NaOH substrate directly. A 1-inch two-zone CVD was introduced for controlling the temperature difference between the Te supply zone and a monitoring zone, respectively. The loading mass of Te is fixed to 1 g and temperature of the Te supply zone is fixed to 600 °C during the whole experiments, giving rise to a Te vapor pressure of ~5.91 Torr. The tellurization process was conducted at atmospheric pressure with a flow rate of 300 sccm for N2 gas and 25 sccm for H2 gas. It should be noted that the H2 gas was introduced to suppress oxidation by oxygen leakage and enhance the detachment of sulfur efficiently by the hydrodesulfurization process.

**Data availability** The data that support the findings of this study are available from the corresponding author upon request.

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Author contributions
S.J.Y. designed and developed the work. J.Z. performed the TEM measurement. B.G.S. performed the STS measurement. D.D.L. calculated the density of states of the monolayer TMdCs. G.H.H. and H.K. assisted with the growth of materials. Q.A.V. performed electrical transport measurements. J.L. contributed the circularly polarized PL measurement. S.M.L. calculated the vibrational modes of MoS1Te1 structures. Y.H.L. guided and analyzed the work. S.J.Y. and Y.H.L. wrote the manuscript. All authors participated in the manuscript review.

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