1. **The convergence test of MCA.** The convergence of magnetocrystalline anisotropy (MCA) and magnetic anisotropic energy (MAE) is tested with different $k$-meshes, shown in Figure S4. Here, we show the changes of MAE with $k$-meshes of $26\times26\times1$, $30\times30\times1$, $36\times36\times1$, $40\times40\times1$, $50\times50\times1$, $56\times56\times1$, $60\times60\times1$, and $66\times66\times1$. The parameter $ISYM = -1$ is set in the VASP, and the corresponding IBZKPT includes 676, 900, 1296, 1600, 2500, 3136, 3600, and 4356 $k$-points in the BZ, respectively. The MCA difference between $k$-mesh of $60\times60\times1$ and $66\times66\times1$ is only 0.012 erg/cm$^2$. Therefore, we adopt the $66\times66\times1$ to calculate MCA.
Figure S1. The evolution of MAE change with the $k$-mesh of 26×26×1, 30×30×1, 36×36×1, 40×40×1, 50×50×1, 56×56×1, 60×60×1, and 66×66×1, respectively.

2. **The geometry of T-VSeTe.** The geometry and electronic properties of T-VSeTe are also investigated. T-VSeTe presents $C_{3v}$ point group. The optimized lattice of T-VSeTe is 3.559 Å, and the vertical distance between V, and Se, Te atoms are 0.905, 1.878 Å, respectively. The ferromagnetic (FM) and antiferromagnetic (AFM) orders are investigated, and the corresponding spin densities are shown in Figure S1. The T-VSeTe shows AFM ground state, and the energy difference between FM and AFM orders is 56.6 meV. The band structures and density of the states (DOS) are also calculated with PBE+U method. The T-VSeTe is spin-polarized metal.
Figure S2. (a-b) The optimized geometry of the T-VSeTe. (a) The top and (b) side view of the T-VSeTe. The spin density of (c) FM and (d) AFM order of VSeTe, the isovalue is 0.77 e/Å³. (e) The spin-polarized band structure and (f) density of the states of the VSeTe. The red and blue lines present spin-α and spin-β electrons, respectively. In the DOS, the black, blue and red lines present PDOS of V, Te and Se atoms, respectively.
3. Band structures with HSE06. The band structure and PDOS of Janus 2H-VSeTe are also calculated with HSE06. And Janus VSeTe is still a bipolar magnetic semiconductor, confirming the result of PBE+U. VSeTe is still a spin-polarized semiconductor with indirect gap of 0.450 eV. Both VBM and CBM mainly come from the contribution of V atom.

![Figure S3. (a) band structure and (b) PDOS of 2H-VSeTe monolayer (ML) are calculated with HSE06. The red and blue lines present spin-α and spin-β electron channels, respectively.](image)

4. PDOS of ML Janus 2H-VSeTe. The partial density of the states (PDOS) of the monolayer (ML) Janus VSeTe with PBE+U. From Figure S2, we can find that the VBM is mainly consisted of $d_{xz}$ and $d_{yz}$ orbital of V atoms, while the CBM is consisted of $d_{z^2}$ orbital of V atoms, respectively.
Figure S4. The PDOS of the ML Janus VSeTe with PBE+U. The states near the fermi-level are mainly contributed by the V atoms. And the red, green, blue, cyan and pink lines with dots represent the contribution of $d_{xy}$, $d_{yz}$, $d_{xz}$, $d_{x^2-z^2}$ and $d_{x^2-y^2}$ of V atoms, respectively.

5. Band structure with SOC. The band structure with SOC along [001] direction is also calculated. Two-dimensional (2D) plots of valance band (VB) and conductance band (CB) (projected to the $k_xk_y$-plane). The values at K and K’ points are the local maximum of the VB, shown in Figure S5 (a). The value at K point is obvious smaller than K’ point (the value difference corresponds to the valley splitting). While the values at K and K’ points are the global minimum (correspond to the dark blue) of the CB, shown in Figure S5 (b).

Figure S5. 2D plots of (a) VB and (b) CB (are projected to the $k_xk_y$-plane in the Brillouin
The different colors show different isovalue. The blue and red correspond to the smallest and largest values, respectively.

6. **Band structure with SOC.** The spin-resolved band structures obtained with SOC along [001] and [100] directions for the Janus VSeTe are calculated, and the easy axis of magnetization is along [100] direction. We can find the states at K and K' points of the Brillouin zone are mainly contributed by vanadium $d_{xy}$ and $d_{x^2-y^2}$ orbitals, above the Fermi-level. While the VBM is mainly contributed by the vanadium $d_{z^2}$ orbital. There is obvious difference in band structure between the along [001] and [100] directions, as there is a large polarized valley in the band structure along [001] direction.

7. **Phonon spectrum of 2H-VSe$_2$ and 2H-VTe$_2$.** The phonon spectrum and phonon density of the monolayer VSe$_2$ and VTe$_2$ are also evaluated. The phonon spectrum, phonon density of states are calculated using finite displacement method, in which we use a supercell with dimension of $6\times6\times1$ for VSe$_2$ and VTe$_2$. In the calculation of phonon spectra, geometries are
fully relaxed until energy and force is converged to $10^{-8}$ eV and $10^{-3}$ eV/Å, respectively.

![Figure S7](image.png)

Figure S7. (a-d) Phonon spectra, phonon density of states (PHDOS) of ML (a, b) of VSe$_2$, and (c, d) of VTe$_2$, respectively.

8. **Synthetization of Janus VSeTe.** 2D Janus TMDs, such as MoSSe is synthesized by selenizing the MoS$_2$ monolayer or substituting MoSe$_2$ monolayer with sulfurization. The Janus VSeTe is expected to be synthesized by replacing the top-layer Se atoms with Te atoms through VSe$_2$ monolayer. Replacing the top-layer Te atoms with Se atom through VTe$_2$ monolayer could also get Janus VSeTe in chemical vapor deposition.
9. **Parameters in CALYPSO.**

In the PSO search, the population size is set to be 30, and the number of generations is maintained at 30. The various supercell sizes are considered with the total number of atoms less than 18.