

## Achieving detector-grade CdTe(Cl) single crystals through vapor-pressure-controlled vertical gradient freeze growth

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### Abstract

Cadmium telluride (CdTe), which has a high average atomic number and a unique band structure, is a leading material for room-temperature X/ $\gamma$ -ray detectors. Resistivity and mobility are the two most important properties of detector-grade CdTe single crystals. However, despite decades of research, the fabrication of high-resistivity and high-mobility CdTe single crystals faces persistent challenges, primarily because the stoichiometric composition cannot be well controlled owing to the high volatility of Cd under high-temperature conditions. This volatility introduces Te inclusions and cadmium vacancies (VCd) into the as-grown CdTe ingot, which significantly degrades the device performance. In this study, we successfully obtained detector-grade CdTe single crystals by simultaneously employing a Cd reservoir and chlorine (Cl) dopants via a vertical gradient freeze (VGF) method. By installing a Cd reservoir, we can maintain the Cd pressure under the crystal growth conditions, thereby preventing the accumulation of Te in the CdTe ingot. Additionally, the existence of the Cl dopant helps improve the CdTe resistivity by minimizing VCd density through the formation of an acceptor complex (ClTe-VCd)<sup>-1</sup>. The crystalline quality of the obtained CdTe(Cl) was evidenced by a reduction in large Te inclusions, high optical transmission (60%), and a sharp absorption edge (1.456 eV). The presence of substitutional Cl dopants, known as Cl<sup>+</sup>Te, simultaneously supports the record high resistivity of  $1.5 \times 10^{10} \Omega \cdot \text{cm}$  and remarkable electron mobility of  $1075 \pm 88 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  simultaneously, has been confirmed by photoluminescence spectroscopy. Moreover, using our crystals, we fabricated a detector with a decent radiation-detection feature. This study demonstrates that the vapor-pressure-controlled VGF method is a viable technical route for fabricating detector-grade CdTe crystals.

## Full Text

### Preamble

#### Achieving detector-grade CdTe(Cl) single crystals through vapor-pressure-controlled vertical gradient freeze growth

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Cadmium telluride (CdTe), which has a high average atomic number and a unique band structure, is a leading material for room-temperature X/ $\gamma$ -ray detectors. Resistivity and mobility are the two most important properties of detector-grade CdTe single crystals. However, despite decades of research, the fabrication of high-resistivity and high-mobility CdTe single crystals faces persistent challenges, primarily because the stoichiometric composition cannot be well controlled owing to the high volatility of Cd under high-temperature conditions. This volatility introduces Te inclusions and cadmium vacancies (VCd) into the as-grown CdTe ingot, which significantly degrades device performance. In this study, we successfully obtained detector-grade CdTe single crystals by simultaneously employing a Cd reservoir and chlorine (Cl) dopants via a vertical gradient freeze (VGF) method. By installing a Cd reservoir, we can maintain the Cd pressure under crystal growth conditions, thereby preventing the accumulation of Te in the CdTe ingot. Additionally, the existence of the Cl dopant helps improve the CdTe resistivity by minimizing VCd density through the formation of an acceptor complex (ClTe-VCd)<sup>-1</sup>. The crystalline quality of the obtained CdTe(Cl) was evidenced by a reduction in large Te inclusions, high optical transmission (60%), and a sharp absorption edge (1.456 eV). The presence of substitutional Cl dopants, known as Cl<sup>+</sup><sub>Te</sub>, simultaneously supports the record high resistivity of  $1.5 \times 10^{10} \Omega \cdot \text{cm}$  and remarkable electron mobility of  $1075 \pm 88 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ , as confirmed by photoluminescence spectroscopy. Moreover, using our crystals, we fabricated a planar detector with  $\tau_{\text{e}}$  of  $(1.11 \pm 0.04) \times 10^{-4} \text{ cm}^2/\text{V}$ , which exhibited decent radiation-detection performance. This study demonstrates that the vapor-pressure-controlled VGF method is a viable technical route for fabricating detector-grade CdTe crystals.

**Keywords:** CdTe, Semiconductor detector, Alpha-detector, Vertical gradient freeze method

## Introduction

Radiation detectors, which are often called special eyes for the visualization of high-energy rays (X/ $\gamma$ -rays), play critical roles in nuclear security and medical diagnosis [1-3]. At the core of these detectors are single crystals, which transform high-energy rays into photons or electron-hole pairs and substantially determine the performance of the detector devices [4-6]. Cadmium telluride (CdTe), a semiconductor with a large effective atomic mass and superior charge transport properties, is a leading material for room-temperature X-ray and gamma-ray detection via a direct detection mechanism [7-9]. The world's first commercial photon-counting computed tomography (CT) system, the NAEOTOM Alpha, was developed using CdTe radiation detectors by Siemens Healthineers. This system was authorized by the FDA (The United States Food and Drug Administration) as "the most significant technological advancement in the CT field in the past decade" [10].

For optimal CdTe crystal performance in detectors, both high resistivity ( $\geq 10^9 \Omega \cdot \text{cm}$ ) and high charge mobility ( $\geq 10^3 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ) are required [11, 12]. The high resistivity of these crystals is essential for minimizing the background signal from thermally generated carriers, enabling high-voltage operation at room temperature [11]. Similarly, a high charge mobility is crucial for supporting highly efficient charge collection and achieving decent energy resolution for detecting devices [12]. However, achieving detector-grade CdTe single crystals with both high resistivity and high mobility has been a challenge, primarily because of difficulties in controlling stoichiometry during the growth process.

The major challenge in controlling stoichiometry arises from the high volatility of Cd under high-temperature conditions. Specifically, the crystallization of stoichiometric CdTe occurs at 1092 °C, where the partial pressure of Cd is assumed to be 1.04 atm, whereas the partial pressure of Te is  $2.8 \times 10^{-3}$  atm, which is negligible compared with that of Cd [13]. Consequently, Cd makes a dominant contribution to the inner pressure of the crucible. To ensure pressure tolerance of the crucible and prevent breakage during the crystal growth process, CdTe materials are typically prepared from Te-rich starting materials [14]. Moreover, even with a stoichiometric CdTe starting material, Cd tends to evaporate into the vapor space in the ampoule, resulting in a Te-rich melt [15]. During crystallization, excess Te segregates at the solid-liquid interface, forming Te clusters that accumulate near the interface owing to the high melt viscosity [15]. This process leads to constitutional supercooling, disrupting the stability of the liquid-solid interface, and causing the incorporation of Te droplets (inclusions) into the growing crystals [15]. Furthermore, the loss of Cd can result in a deficiency of Cd at the solid-liquid interface, introducing vacancy defects into the crystal matrix. In the context of CdTe crystals used for detectors, Te inclusions introduce nonuniform internal electric fields and electron cloud trapping, deteriorating the charge transport properties [16]. In addition, a high concentration of Cd vacancies can act as acceptors and unfavorable charge-trapping

centers, compromising the resistive and charge-transport properties and limiting their application in detection.

Efforts to produce high-quality CdTe crystals with precise stoichiometry have led to various methods including the modified vertical Bridgman method (MVB), high-pressure Bridgman method (HPB), and traveling heater method (THM). Using the MVB technique for CdTe growth, Mochizuki et al. attempted to regulate the partial pressures by incorporating a Cd reservoir chamber [17]. Similarly, the HPB technique, which has been utilized for CdTe crystal growth, mitigates the loss of volatile Cd in CdTe by applying a high inert gas pressure during the growth process [18]. Additionally, THM, recognized as a Te solvent method and developed for the uniform growth of CdTe, offers advantages such as a low growth temperature and the purifying influence of the Te solvent zone. This method reduces the risk of ampoule breakage and facilitates the fabrication of high-purity CdTe with minimal Cd vacancies [19]. However, these methods typically involve relative mechanical motion between the heater and ampoule containing charged materials. Such mechanical motion can compromise the growth stability, leading to stoichiometry-correlated defects and a reduction in the yield of single crystals. By contrast, the vertical gradient freeze (VGF) method, which is a melt growth method, is modified using temperature-profile-programmed control of the furnace multizone instead of the mechanical movement of the ampoule or furnace [20]. VGF offers a higher growth rate than THM and ensures more stable conditions at the crystallization front by minimizing the thermal drift and temperature fluctuations [21]. Despite these advantages, Su et al. reported the growth of undoped CdTe using the VGF method, achieving a resistivity of  $1 \times 10^7 \Omega \cdot \text{cm}$  at room temperature, which falls short of radiation applications [22].

Given the pivotal role that doping plays in regulating the carrier concentration and band structure in semiconductors, chlorine dopants are often favored for their function as donors in CdTe crystals [8]. Cl dopants compensate for existing defects, reduce trap centers, enhance charge-collection efficiency, and increase resistivity. In this context, Bugár et al. grew CdTe crystals using the VGF method with Cl dopants, achieving high resistivity for the as-grown CdTe crystals [23]. However, the detector performance was suboptimal, mainly attributed to large Te inclusions, necessitating a sophisticated two-step post-growth annealing treatment. The treatment involved an initial step involving post-growth annealing in Cd overpressure to eliminate large-sized Te inclusions in the CdTe crystals, followed by a second annealing step under Te overpressure to restore high resistivity after Cd vapor annealing. Unfortunately, the detection capability of CdTe crystals treated in this manner was worse than that of the as-grown samples. Thus, attempts to obtain CdTe single crystals with both high resistivity and charge transport properties through direct VGF growth or post-treatment require further study. Conversely, the introduction of an additional Cd reservoir during the VGF growth of cadmium zinc telluride (CZT) is an effective approach for reducing large Te inclusions, resulting in decent spectral performance [24]. Therefore, controlling the Cd vapor pressure

during the VGF growth process of CdTe(Cl) could be the key to reducing large Te inclusions and enhancing detector performance.

In this study, we introduced an external Cd source to regulate the cadmium vapor pressure and incorporated Cl as a dopant during the VGF growth of CdTe to preserve the crystal stoichiometry and modulate its electrical properties. To evaluate the effectiveness of this approach, we conducted a comprehensive analysis of the properties of CdTe(Cl) crystals using various techniques, such as infrared transmittance spectroscopy, near-infrared transmission spectroscopy, and photoluminescence spectroscopy. Furthermore, we investigated the resistivity and charge transport properties of CdTe(Cl) crystals and successfully fabricated an alpha detector.

## Results and Discussion

Fig. 1a [Figure 1: see original paper] shows a schematic representation of the furnace/ampoule assembly, along with a typical gradient freeze temperature program. The synthesized polycrystalline CdTe was loaded into a homemade crucible with an extension to house a Cd reservoir. Recognizing that crystal quality can be improved by superheating [25], we performed a preheating process to increase the temperature of the ampoule to more than 20 °C above the melting point of CdTe before crystal growth. Consequently, the CdTe melt was heated to a superheating temperature to eliminate the need for seed crystals. The growth process involved the electronic translation of the temperature gradient through the melt. To optimize crystal growth, we employed low-temperature gradients ( $5\text{ °C cm}^{-1}$ ) and moderate growth rates ( $0.1\text{ cm h}^{-1}$ ) to reduce thermal stress and minimize dislocation density, which are critical factors for preventing constitutional supercooling effects.

In addition, to ensure accurate stoichiometry in both the CdTe melt and solid, we meticulously programmed the temperature range between 800 °C and 850 °C for the Cd reservoir during solidification and cooling. Fig. 1b shows a typical cross-sectional image of an as-grown CdTe(Cl) ingot with a diameter of 2.2 cm and length of 6 cm. This image shows that the ingot consists of several single-crystalline grains, including a large grain over  $0.5\text{ cm} \times 0.5\text{ cm}$  in size. Furthermore, the measured Cl concentration for CdTe(Cl) is  $1.79 \times 10^{18}\text{ cm}^{-3}$ . Te inclusions in the as-grown CdTe(Cl) crystals were assessed using IR transmission microscopy. As illustrated in Fig. 1c, the presence of Te inclusions in the sample decreased, and the average sizes of the Te inclusions were effectively reduced to below  $5\text{ }\mu\text{m}$ , with an average density of  $3.8 \times 10^6\text{ cm}^{-3}$  (Fig. 1d). These results indicate that the incorporation of large Te inclusions into the crystal was effectively suppressed by controlling the partial pressure of Cd in the growth ampoule.

We used optical methods to evaluate the obtained CdTe crystals. As illustrated in Fig. 2a [Figure 2: see original paper], a high transmittance value of nearly 60% can be observed over a large wavenumber range from  $4000\text{ cm}^{-1}$  to  $500$

$\text{cm}^{-1}$  at room temperature. Notably, the absorption behavior of a crystal plays a crucial role in determining its infrared transmittance characteristics, which are primarily driven by lattice absorption and free-carrier absorption [26]. The lattice absorption is primarily influenced by the presence of Te inclusions, which can lead to lattice distortion and disrupt the periodic structure of the lattice [27]. In our CdTe crystals, which are characterized by the reduction of large Te inclusions, the impact of lattice absorption was negligible. Consequently, free-carrier absorption became the dominant mechanism responsible for absorption. The absorption coefficient  $\alpha$  associated with the free-carrier absorption is directly proportional to the free-carrier density  $N$  [28]. Therefore, the significantly high transmittance observed for the CdTe(Cl) sample can be attributed to a reduction in free-carrier density.

A typical UV-Vis-NIR transmittance spectrum of CdTe(Cl) crystals is presented in Fig. 2b. Significantly, within the wavelength range of 820 nm to 950 nm, a distinct exponential change in transmittance was observed. This observation enabled the calculation of the absorption coefficient  $\alpha$ , thereby facilitating the determination of the bandgap ( $E_g$ ) of CdTe crystals near the absorption cutoff. The inset of Fig. 2b demonstrates the  $E_g$  calculations for direct transitions in CdTe by employing Equation [27]:  $(\alpha h\nu)^2 \propto (h\nu - E_g)$ , where  $h\nu$  is photon energy. Extrapolating these calculations revealed that CdTe(Cl) samples have  $E_g$  values close to 1.456 eV, with a relative error of less than 5%.

Photoluminescence (PL) spectroscopy was conducted at a temperature lower than that of liquid nitrogen ( $\text{N}_2$ ) to investigate the specific defects in CdTe(Cl). The typical PL spectra shown in Fig. 2c for the CdTe(Cl) samples reveal the presence of three distinct regions: (I) The first region, located around 1.58 eV, corresponds to excitonic recombination, specifically the near-band-edge shallow donor-bound exciton peak ( $\text{D}^0, \text{X}$ ) [29]. (II) The second region, situated around 1.54 eV, corresponds to the Donor-Acceptor Pair recombination (DAP) region [29]. (III) The third region, approximately at 1.46 eV, is broad and recognized as the defect-related deep emission peak known as the A-center [30]. Region (I) features a Cl-related shallow donor-bound exciton peak ( $\text{D}^0, \text{X}$ ) ( $h\nu = 1.582$  eV) and the free exciton line (FE:  $h\nu = 1.588$  eV) positioned at the high-energy side of ( $\text{D}^0, \text{X}$ ). The presence of free excitons indicates the high crystallinity of the CdTe(Cl) sample.

Region (II), centered at approximately 1.54 eV, was initially associated with an acceptor-like structural defect caused by Cd vacancies or complexes comprising a Cd vacancy and two residual donors (VCd-2D) [29]. However, this emission nearly disappeared upon Cl doping, mirroring the PL spectrum of CdTe(Cl) grown using THM [30]. This suggests consumption of Cd vacancies owing to the formation of Cl-related complexes. Simultaneously, the intensity of the A-center became more pronounced in the presence of Cl doping. According to experimental findings, this is primarily governed by the spontaneous formation of the acceptor complex  $(\text{ClTe-VCd})^{-1}$ . Consequently, a plausible defect reaction equation is formulated as follows:

$$\text{Te} + \text{V}^{-2}_{\text{Cd}} = (\text{ClTe} - \text{VCd})^{-1}$$

A Cl atom occupying a Te site ( $\text{Cl}^{+}_{\text{Te}}$ ) acts as a shallow donor and attracts charged Cd vacancies, forming an acceptor complex  $(\text{ClTe}-\text{VCd})^{-1}$ . This process is a form of self-purification.

The electrical properties were characterized using an I-V test. As demonstrated in the I-V curve in Fig. 3a [Figure 3: see original paper], the results align well with Ohmic law at a  $\pm 0.1$  V bias, and this data can be used to calculate the resistivity according to the equation: where  $\rho$  is the resistivity,  $s$  is the contact area  $0.6 \text{ cm} \times 0.6 \text{ cm}$ ,  $l$  is the sample thickness (0.2 cm), and  $R$  is the resistance.

Upon analyzing the typical I-V curve, a calculated resistivity of  $1.5 \times 10^{10} \Omega \cdot \text{cm}$  was obtained for a CdTe(Cl) wafer. Additionally, Fig. 3b presents a histogram showing the calculated resistivity values for five CdTe devices. This histogram exhibits a normal distribution with a peak around  $1.5 \times 10^{10} \Omega \cdot \text{cm}$ , and the resistivities of all the devices are above  $10^{10} \Omega \cdot \text{cm}$ , representing semi-insulating characteristics under Cl saturation conditions.

Additionally, electron mobility is a pivotal parameter that significantly influences the overall performance of the detector. To this end, a thorough investigation into the electron mobility of the CdTe(Cl) planar detector was conducted using the LBIC (laser beam-induced current) technique. As shown in Fig. 3c [Figure 3: see original paper], the typical time-of-flight waveform transients of electrons captured by the CdTe(Cl) planar detector under various bias voltages show an initial region where the signal experiences a rapid reduction, which is attributed to fast surface recombination processes [31]. Subsequently, the transient current waveforms exhibit a plateau, and the separation between this plateau and the trailing edge of the transient current is identified as the transit time  $\tau_r$  [31]. This critical point signifies the arrival of electrons at the anode, as indicated by the dashed line in Fig. 3c. The extended tail following the transit time is attributed to the gradual release of carriers from traps. By measuring the transit time and assuming a constant average electric field, the drift mobility can be calculated using [31]: where  $L$  represents the sample thickness (0.2 cm), and  $E$  denotes the applied electric field. Fig. 3d further illustrates the relationship between the electric field strength and electron drift velocity, displaying a linear correlation.

The slope of the electric field versus velocity graph signifies the electron mobility  $\mu_e$ , which was determined to be  $1075 \pm 88 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ .

After the above characterizations, we used the obtained CdTe crystals to fabricate a planar radiation detector. As shown in Fig. 4a [Figure 4: see original paper], the spectra of the  $\alpha$ -particles ( $^{241}\text{Am}@5.48 \text{ MeV}$ ) acquired using the Au/CdTe(Cl)/Au planar detector are shown at various bias voltages ranging from 60 V to 200 V. With an increase in bias voltage, the centroid of the full-energy peak shifts toward higher channel numbers, with a preference for higher amplitudes. Importantly, the spectra at a reverse bias voltage ( $V = 200 \text{ V}$ ) exhibit a symmetrical shape with prominent lines corresponding to the 5.48 MeV

energies of the  $^{241}\text{Am}$ - $\alpha$  sources. Concurrently, the full width at half maximum (FWHM) value narrowed to approximately 20 channels around the full-energy peak channel (840) at 200 V, resulting in an energy resolution of 2.38%. Considering the shallow penetration depth of  $\alpha$ -particles, which is approximately 14  $\mu\text{m}$  beneath the detector surface, the mobility-lifetime product  $\tau$  of the charge carriers can be estimated using the simplified Hecht equation for a single carrier [32]:

$$\text{CCE} = d^2 (1 - \exp(-$$

where CCE represents the charge collection efficiency,  $Q$  denotes the resulting peak centroid, and  $Q_0$  corresponds to the total number of carriers generated by an incident photon.

Thus, achieving a favorable mobility-lifetime ( $\tau$ ) product is a critical parameter to ensure superior charge collection efficiency for a planar detector with thickness ( $d$ ) under the applied voltage ( $U$ ). Moreover, because the cathode of the detector was irradiated with  $\alpha$ -particles, the experimental data could be fitted using Eq. (5), which allows the determination of the electron mobility lifetime product  $\tau_{-e}$  for CdTe(Cl), revealing a value of  $(1.11 \pm 0.04) \times 10^{-4} \text{ cm}^2/\text{V}$  (Fig. 4b).

Finally, we achieved high-resistivity CdTe single crystals with exceptionally high electron mobilities via Cl doping. Increased electrical resistivity can lower leakage currents and improve the signal-to-noise ratio, which is crucial for detecting weak X-ray signals with high precision. The enhanced charge transport properties can contribute to better charge collection efficiency, ultimately leading to improved energy resolution and sensitivity in X-ray detection applications. Typically, the resistivity of CdTe is approximately  $10^9 \Omega \cdot \text{cm}$ . For example, CdTe grown by THM through Acrorad Company, Ltd. exhibits a resistivity of approximately  $6 \times 10^9 \Omega \cdot \text{cm}$ , exceeding the intrinsic value of CdTe ( $4 \times 10^9 \Omega \cdot \text{cm}$ ) [33]. In our experiments, we obtained an even higher resistivity, primarily attributable to p-type conduction in high-resistivity CdTe crystals, which was heavily compensated by Cl. The observed resistivity surpassed the intrinsic value of CdTe, owing to the increased contribution of holes, which possess significantly lower mobility than electrons, to the conductivity of the material [33]. Furthermore, we recorded a significant improvement in electron mobility, which is the highest value ever reported for VGF-grown CdTe(Cl) crystals. This value is in agreement with the  $1100 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  achieved using THM-grown CdTe(Cl) crystals [34].

Achieving both high resistivity and high mobility in high-resistance CdTe devices is a considerable challenge. This necessitates the compensation of intrinsic defects in the crystal through doping to achieve high resistivity. However, both intrinsic defects and introduced impurities can adversely affect carrier transport via scattering and trapping. Additionally, there is an ongoing debate within the scientific community regarding the attainment of high resistivity in CdTe crystals. While the deep donor model attributes high resistivity to Fermi level pinning by a deep donor level induced by native defects or extrin-

sic deep donors, the compensation model in CdTe(Cl), as proposed by Maohua Du et al., suggests that self-compensation between the shallow donor  $\text{Cl}^+\{\text{Te}\}$  and its A-center  $(\text{ClTe-VCd})^{-1}$  maintains a fixed Fermi level, thus ensuring high resistance, particularly at high Cl concentrations up to its solubility limit [35]. The dominant A-center emission observed in the photoluminescence results of our high-resistivity CdTe(Cl) crystals supports the self-compensation model. This emission is associated with the recombination between the shallow donor  $\text{Cl}^+\{\text{Te}\}$  ( $E_c - 0.014$  eV) and the acceptor complex  $(\text{ClTe-VCd})^{-1}$  ( $E_v + 0.2$  eV). As the concentration of  $\text{Cl}^+\{\text{Te}\}$  increased, the concentration of the acceptor complex may surpass that of isolated  $V^{-2}\{\text{Cd}\}$  at higher levels of  $\text{Cl}^+\{\text{Te}\}$ . At this point, Fermi level pinning occurs and ceases to change owing to donor self-compensation. Notably, the A-center, known for having a charge transfer level of approximately 0.2 eV above the valence band [36], is not considered a trapping center based on the specific energy positions and cross-sections of defects [37]. Moreover, there is a reduction in Cd vacancy-related defects at energy levels 0.3 eV to 0.4 eV above the valence band edge in CdTe(Cl), which are expected to act as significant electron traps [36]. Reducing the size of Te inclusions in the CdTe crystals further enhances the charge transport properties by minimizing carrier trapping and scattering [16]. Consequently, the high electron mobility observed in CdTe(Cl) is likely owing to the formation of a complex between ClTe and VCd. This complex eliminates the intrinsic electron trap states of VCd, coupled with the reduction of large Te inclusions, contributing to improved charge transport characteristics.

## Conclusion

In summary, we employed a vapor-pressure-controlled vertical-gradient freeze-growth method to produce CdTe(Cl) crystals, resulting in the reduction of large Te inclusions. Additionally, we conducted a comprehensive investigation of the properties of CdTe(Cl) crystals. We observed significant improvements in both the optical and electrical properties following Cl doping. Remarkably, high IR transmission of up to 60% was observed for the CdTe(Cl) sample. UV-Vis-NIR analysis confirmed an energy bandgap value close to 1.456 eV for the CdTe(Cl) samples. Moreover, through PL spectroscopic analysis, we experimentally observed a dominant emission related to the A-center  $(\text{ClTe-VCd})^{-1}$  defect. This observation supports the notion of a self-compensation mechanism involving  $\text{Cl}^+\{\text{Te}\}$  and its A-center under Cl saturation conditions, which explains the high resistivity of CdTe(Cl). Consequently, the resistivity exhibits a remarkable increase to  $10^{10} \Omega \cdot \text{cm}$ . The TOF test demonstrated a high electron mobility of  $1075 \pm 88 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  for CdTe(Cl). Furthermore, alpha spectroscopic measurements revealed charge transport properties with an electron mobility lifetime product  $\tau_e$  of  $(1.11 \pm 0.04) \times 10^{-4} \text{ cm}^2/\text{V}$ . In conclusion, these findings collectively demonstrate the successful use of our vapor-pressure-controlled vertical-gradient freeze-growth method for fabricating high-quality detector-grade CdTe crystals.

## Experimental Methods

The CdTe compound was synthesized in a custom-designed rocking furnace. To ensure purity, we selected 6N-grade cadmium and tellurium as raw materials. Furthermore, we incorporated 4N-grade ultradry CdCl<sub>2</sub> salt as the dopant source. To grow the crystals, the initially synthesized CdTe was placed in a pyrolytic boron nitride (pBN) crucible, which was subsequently positioned inside a quartz ampoule. This quartz ampoule, featuring an extension containing a reservoir of Cd, was then evacuated to a pressure of  $1 \times 10^{-5}$  Pa and sealed under vacuum. The specific structures of the crystal growth ampoule and Cd reservoir are shown in Fig. 1a.

During the initial growth stages, the pBN crucible prevented the melt from falling into the Cd reservoir when the entire material was melted. Subsequently, a crystal growth process was conducted using a multizone vertical gradient freeze furnace equipped with Cd partial pressure control. We obtained a CdTe(Cl) ingot that was subsequently sliced into wafer forms using a diamond wire saw. We conducted glow discharge mass spectrometry (GDMS) to determine the Cl dopant concentration. In addition, an infrared transmission microscope was used to examine the Te inclusions in the CdTe crystals.

The CdTe samples were polished and etched with bromine-methanol solution. To measure IR transmittance in the wavenumber range  $4000 \text{ cm}^{-1}$  to  $5000 \text{ cm}^{-1}$ , we employed an infrared spectrometer (Nicolet Nexus 670 spectrometer, Thermo Fisher Scientific, Waltham, MA, USA).

UV-Vis-NIR transmission spectra were acquired using a UV-3150 ultraviolet-visible/near-infrared spectrometer (Shimadzu, Japan). To measure the photoluminescence spectrum at low temperatures (77 K), a 532 nm laser beam was used as the excitation source. The signals emitted from the samples were collected using a Horiba Scientific Labram HR Evolution Raman spectrometer (Horiba Scientific).

The planar Au/CdTe(Cl)/Au detector, with dimensions of  $0.7 \text{ cm} \times 0.7 \text{ cm} \times 0.2 \text{ cm}$ , was fabricated by thermal evaporation of Au on the crystal surface. Subsequently, it was prepared for I-V testing using a semiconductor parameter analyzer (Agilent 4155C, Agilent, USA). Electron mobility was investigated using LBIC (laser beam-induced current) measurements. A pulse laser with a wavelength of 527 nm, pulse width of 893 ps, and  $0.1 \mu\text{J}/\text{pulse}$  was directed onto the cathode to generate electron-hole pairs, resulting in approximately  $10^{12}$  photons per  $\text{mm}^2$  on the surface. A bias voltage was applied using a Keithley 6517 and the resulting current signals were recorded using a digital oscilloscope (LeCroy WaveRunner 610 Zi, 1 GHz, 20 GS/s) connected through a  $50 \Omega$  resistor.

To measure the response to  $\alpha$ -particles, the cathode was exposed to uncollimated <sup>241</sup>Am@5.49 MeV radioactive sources, while being biased using an ORTEC 710 bias supply.

The pulse shape information was amplified using an ORTEC 142 charge-sensitive preamplifier (CSP) and an ORTEC 570 shaping amplifier. Subsequently, the signals were analyzed using a standard Imdetek AMCA-01 multichannel analyzer (MCA) and processed using software to generate and display the response spectra. Measurements were conducted with a shaping time of 1  $\mu$ s and collection time of 300 s in a dark environment at room temperature, with the pressure maintained below 10 Pa.

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### Footnotes

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