Ultrasensitive multimodal physicochemical sensing for autonomous robotic decision-making has numerous applications in agriculture, security, environmental protection, and public health. Previously reported robotic sensing technologies have primarily focused on monitoring physical parameters such as pressure and temperature. Integrating chemical sensors for autonomous dry-phase analyte detection on a robotic platform is rather extremely challenging and substantially underdeveloped. Here, we introduce an artificial intelligence–powered multimodal robotic sensing system (M-Bot) with an all-printed mass-producible soft electronic skin–based human-machine interface. A scalable inkjet printing technology with custom-developed nanomaterial inks was used to manufacture flexible physicochemical sensor arrays for electrophysiology recording, tactile perception, and robotic sensing of a wide range of hazardous materials including nitroaromatic explosives, pesticides, nerve agents, and infectious pathogens such as SARS-CoV-2. The M-Bot decodes the surface electromyography signals collected from the human body through machine learning algorithms for remote robotic control and can perform in situ threat compound detection in extreme or contaminated environments with user-interactive tactile and threat alarm feedback. The printed electronic skin–based robotic sensing technology can be further generalized and applied to other remote sensing platforms. Such diversity was validated on an intelligent multimodal robotic boat platform that can efficiently track the source of trace amounts of hazardous compounds through autonomous and intelligent decision-making algorithms. This fully printed human-machine interactive multimodal sensing technology could play a crucial role in designing future intelligent robotic systems and can be easily reconfigured toward numerous practical wearable and robotic applications.

Because of its high flexibility and conformability, electronic skin (e-skin) presents itself as the ideal interface between electronics and the human/robot bodies. In literature, e-skin has demonstrated a wide range of physical and chemical sensing applications, ranging from consumer electronics, digital medicine, smart implants, to environmental surveillance (17–31). Despite such promise, several challenges exist for e-skin–based multifunctional robotic systems. Because most rapid detection approaches for hazardous compounds require manual solution-based sample preparation steps, integrating chemical sensors for autonomous remote dry-phase analyte detection onto an e-skin–based robotic sensing platform is extremely challenging and substantially underdeveloped, hindering e-skin’s capabilities for robotic interaction and cognition of the external world (7, 32). A robotic manipulator would require tactile, chemical, and...
temperature feedback to handle arbitrary objects, collect target samples, and carry out accurate chemical analysis in extreme environments (33). Another problem for e-skin interfaces is that preparing high-performance sensors generally requires manual drop-casting modifications of nanomaterials, which can lead to large sensor variations (34). Now, there is a lack of scalable low-cost manufacturing approaches to prepare thin, ultraflexible, multifunctional robotic physicochemical sensor patches. Despite these concerns, there is a strong need for an efficient human-machine interface that can reliably extract physiological features (35) as well as accurately control and receive real-time user-interactive feedback.

To address these challenges, we introduce here an artificial intelligence (AI)–powered human-machine interactive multimodal sensing robotic system (M-Bot) (Fig. 1A and Movie 1). The M-Bot is composed of two fully inkjet-printed stretchable e-skin patches, namely, e-skin-R and e-skin-H, which interface conformally with the robot and human skin, respectively. The e-skins with powerful physicochemical sensing capabilities are mass producible and reconfigurable and can be entirely prepared using a high-speed, low-cost, and scalable inkjet-printing technology with a series of custom-developed nanomaterial inks. Upon collecting physiological data, the machine learning model can decode the surface electromyography (sEMG) signals from muscular contractions (recorded by e-skin-H) for robotic hand control. Simultaneously, e-skin-R can perform proximity sensing and tactile and temperature perceptual mapping, alongside real-time hydrogel-assisted electrochemical on-site sampling and analysis of both solution-phase and dry-phase threat compounds including explosives [such as 2,4,6-trinitrotoluene (TNT)], pesticides (such as OPs), and biohazards (such as SARS-CoV-2 virus). Upon detection, real-time haptic and threat alarm feedback communications were achieved via
electrical stimulation of the human body with e-skin-H. The threat sensing capabilities of the M-Bot could pave the way for automated chemical sensing, facilitating machine-mediated decisions for a wide range of practical robotic assistance applications.

RESULTS
Design of the human-machine interactive e-skins
E-skin-R is composed of high-performance–printed nanoengineered multimodal physicochemical sensor arrays that can be placed on the palm and fingers of the robotic hand (Fig. 1, B and C). The entire sensor patch can be rapidly manufactured in a large-scale and low-cost method via a powerful drop-on-demand inkjet printing technology (Fig. 1D, fig. S1, and movie S1). On top of e-skin-R are engraved kirigami structures that provide high stretchability without conductivity changes under a 100% strain, which is crucial for any robotic hand with high degrees of freedom in movement. E-skin-H consists of four sEMG electrode arrays (channels), alongside a pair of electrical stimulation electrodes, which can be fabricated similarly with inkjet printing followed by transfer printing onto a stretchable polydimethylsiloxane (PDMS) substrate (Fig. 1E). With assistance from AI, multimodal physicochemical sensing, and electrical stimulation–based feedback control, e-skin-R and e-skin-H form a closed-loop human-machine interactive robotic sensing system (Fig. 1F).

Fabrication and characterization of the fully inkjet-printed multimodal sensor arrays
The multimodal physicochemical sensor arrays on e-skin-R were fabricated via serial printing of silver (interconnects and reference electrode), carbon (counter electrode and temperature sensor), polypyrrole (PPy) (encapsulation), and target-selective nanoengineered sensing films (tactile sensor and biochemical sensing electrodes) (Fig. 2A). Customized nanomaterial inks were developed to meet the viscosity, density, and surface tension requirement for inkjet printing and to achieve the desired analytical performance (Figs. 2 and 3 and table S1). The chemical sensors were coated with a soft gelatin hydrogel that was loaded with an electrolyte or redox probe to facilitate target analyte sampling and analysis in situ (Supplementary Methods and fig. S4). The inkjet-printed carbon electrodes (IPCEs) showed reproducible electroanalytical performance and rapid response for on-site detection of dry-phase analytes (redox probe Fe"+/Fe" was used in the hydrogel as an example) (fig. S5). The detection area or resolution can be enhanced by increasing either hydrogel size (fig. S6) or electrode density (fig. S7).

To effectively manipulate objects and to avoid harming either the e-skin or the object, real-time tactile feedback was enabled by incorporating a piezoresistive pressure sensor based on a printed silver nanowires (AgNWs)/nanotextured PDMS (N-PDMS) sensing film (Fig. 2, B and C). Such tactile sensation provides the robot with the haptic capability to grasp and handle samples. The geometry changes of the AgNWs/N-PDMS in response to a pressure load change the sensor’s conductance (Fig. 2D and fig. S8). The pressure sensor displayed stable performance under repetitive pressure loading (Fig. 2E and fig. S8).

To demonstrate the feasibility of using the printed biosensors for hazardous chemical detection, a standard chemical explosive (TNT), an OP nerve agent simulant (paraoxon-methyl), and a biohazard pathogenic protein from the SARS-CoV-2 virus were chosen. The detection of TNT was achieved using a Pt-nanoparticle–decorated graphene electrode, which was prepared by droplet inkjet printing of aqueous graphene oxide (GO), Pt ions, and propylene glycol and subsequently subjected to thermal reduction. The Pt-graphene showed superior electrocatalytic performance compared with classic carbon and graphene electrodes (Fig. 2, F to H, and fig. S9). The reduction of p-NO₂ to p-NH₂ catalyzed by the Pt-graphene can be detected via negative differential pulse voltammetry (nDPV) (9, 36). The obtained reduction peak amplitude in the nDPV voltammograms showed a linear relationship with the target TNT concentrations with a sensitivity of 0.95 μA cm⁻² ppm⁻¹ and a detection limit of 10.0 ppm (Fig. 2I). Note that a custom voltammogram analysis with an automatic peak extraction strategy was used by the robot to analyze the original nDPV curves as illustrated in fig. S10. When integrated onto a robotic hand, the hydrogel-coated Pt-graphene sensor could sample the dry-phase TNT efficiently and provide a stable current response within 3 min (Fig. 2J); the TNT sensor can be regenerated in situ through repetitive nDPV scans to deplete the sampled analyte molecules toward continuous robotic sensing (fig. S11). For OP analysis, Pt-graphene and carbon have low electrochemical activity, because Zr-based metal-organic framework (MOF-808) was reported to have strong interaction with OPs (37, 38). Thus, the printed MOF-808–modified gold nanoparticle electrode (MOF-808/Au) was selected to achieve efficient non-enzymatic OP reduction at a relatively low voltage (Fig. 2K to M, and fig. S12). In this way, the catalyzed reduction of paraoxon-methyl can be monitored via nDPV using the MOF-808/Au sensors with a sensitivity of 1.4 μA cm⁻² ppm⁻¹ and a detection limit of 4.9 ppm (Fig. 2N). In addition to high sensitivity, these printed sensors could also perform high-concentration threat analysis (fig. S13). Similar to TNT detection, a 3- to 4-min sampling time was found to be sufficient for stable robotic dry-phase OP analysis (Fig. 2O). The Pt-graphene TNT sensors and MOF-808/Au OP sensors showed high selectivity over other nitro compounds (figs. S14 and S15). Because of the excellent stability of the catalytic performance of Pt-graphene and MOF-808/Au, the printed sensors can perform continuous TNT and OP analysis (fig. S16).

Label-free SARS-CoV-2 virus detection was demonstrated from a printed multiwalled carbon nanotube (CNT) electrode that was functionalized with antibodies specific to SARS-CoV-2 spike 1 protein (S1) (Fig. 2, P and Q). The CNT layer had a high electroactive surface area for sensitive electrochemical sensing while providing rich carboxylic acid functional groups for amine-containing affinity probe immobilization to achieve versatile biohazard sensing (39–41). The successful surface modification of the S1 sensor was confirmed after each surface immobilization step (Fig. 2R and figs. S17 and S18). Parts-per-billion level S1 sensing was performed on the basis of the signal change of the electroactive redox probe (Fe"+/Fe"⁻) caused by the blockage of the electrode surface due to the S1 protein binding (Fig. 2S). The response variations of such S1 sensors can be further reduced in the future with an automatic surface modification process. The SARS-CoV-2 S1 sensor shows high selectivity over other viral proteins as illustrated in fig. S19. On-the-spot robotic S1 protein detection was successfully demonstrated using a collection and detection hydrogel containing the redox probe on the sensor that touched a surface containing a dry blot of the S1 protein (Fig. 2T). Although the nonspecific adsorption could potentially reduce the selectivity of the hydrogel detection process (fig. S20), the semiquantitative data conveniently and automatically
obtained on site by the sensor could still provide the users rapid, real-time feedback and alert on the presence of biohazard.

To ensure accurate hazard detection in extreme operational environments, a printed carbon-based temperature sensor was designed for on-site temperature sensing and chemical sensor calibration during operation (fig. S21). All printed sensors maintained similar performance under and after repetitive mechanical bending tests, indicating their high mechanical stability (fig. S22). The freshly prepared hydrogels can be stored at 4°C in a moist chamber for more than 1 week and maintain similar sensing performance (fig. S23). To minimize the influence of the shearing and normal forces on the sensor performance, the AgNWs/N-PDMS pressure sensor was designed to form a protection micowell for each hydrogel-coated biosensor and to facilitate reliable chemical analyte sample collection ( figs. S24 and S25); moreover, the tactile feedback from the AgNWs/N-PDMS pressure sensor could ensure stable electrochemical sensing performance (contact pressure was maintained between 0 and 500 Pa during operation).

Fig. 2. Characterization of the fully inkjet-printed multimodal sensor arrays on the e-skin-R. (A) Photograph of a multimodal flexible sensor array printed with custom nanomaterial inks that consists of a temperature sensor, a tactile sensor, and an electrochemical sensor coated with a soft analyze-sampling hydrogel film. Scale bar, 5 mm. (B and C) Schematic (B) and SEM image (C) of the printed AgNWs/N-PDMS tactile sensor. Scale bar, 1 μm. (D and E) Response of a tactile sensor under varied pressure loads (D) and repetitive pressure loading (E). (F and G) Schematic (F) and SEM (G) of the printed Pt-graphene electrode for TNT detection. Scale bar, 4 μm. (H) Cyclic voltammograms (CVs) of an IPCE and a printed Pt-graphene electrode in 0.5 M H₂SO₄ and in 5 mM K₃Fe(CN)₆ (inset), j, current density. (I) nDPV voltammograms and the calibration plots (inset) of TNT detection using a Pt-graphene electrode. (J) Dynamics of robotic fingertip detection of dry-phase TNT using an Pt-graphene sensor. (K and L) Schematic (K) and SEM image (L) of the printed MOF-808/Au electrode for OP detection. Scale bar, 100 nm. (M) CVs of an IPCE, a Au electrode, and a MOF-808/Au electrode in McIlvaine buffer and in 5 mM K₃Fe(CN)₆ (inset). (N) nDPV voltammograms of the OP detection. Inset: The calibration plots. (O) Robotic fingertip detection of dry-phase OP using a MOF-808/Au sensor. (P and Q) Schematic (P) and SEM image (Q) of the printed CNT electrode for SARS-CoV-2 detection. Scale bar, 250 nm. (R) DPV voltammograms of a printed CNT electrode in 5 mM K₃Fe(CN)₆ after each surface immobilization step. EDC, 1-ethyl-3-(3-dimethylaminopropyl)carbodiimide; AbC, capture antibody. (S) Calibration plots of the CNT-based sensor for S1 detection. Δj, percentage DPV peak current changes after target incubation. (T) Response of a CNT sensor in the presence and absence of dry-phase S1. All error bars represent the SD from three sensors.
Evaluation of e-skin-H for AI-assisted human-machine interaction

E-skin-H acts as a human-machine interface for autonomous robotic control and object manipulation (Fig. 3A). In particular, e-skin-H records neuromuscular activity, which provides an intuitive interface to perform hand gesture recognition, through its inkjet-printed, PDMS-encapsulated, four-channel, three-electrode sEMG arrays (Fig. 3, B and C, and fig. S26, A to C). Analyzing the interfacial contact with the skin, e-skin-H demonstrates high stretchability with good mechanical compliance during physical activities through its serpentine structure to provide reliable sEMG recordings (fig. S26, D to I).

Upon signal acquisition, various machine learning algorithms were evaluated for accurate gesture recognition including linear regression, random forest, artificial neural networks, support vector machines (kernels: radial, sigmoid, linear, and polynomial), and k-nearest neighbors (KNNs). Each algorithm was shown to extract motor intention from sEMG signals, acting as a bridge between conscious thought and prosthetic actuation. Of all the machine learning algorithms, the KNN model provided the highest prediction accuracy.
accuracy for all six hand gestures with an overall mean accuracy across 5000 randomly selected training data of 97.29 ± 1.11% based on real-time experimental results collected from a human participant (Fig. 3, D and E, and fig. S27). The next best model was the random forest classifier, which was found to have a similar average classification accuracy except with a higher variance. The KNN model was able to provide high-accuracy recognition of gestures with different angles when applying the e-skin-H to other body parts such as the neck, lower limb, and upper back—each time achieving an accuracy of greater than 90% (figs. S28 and S29).

For each gesture, five features were extracted from the associated peak in the root mean square (RMS)–filtered sEMG data (fig. S30): peak height (PH), peak standard deviation (STD), maximum slope (MS), peak average (PA), and peak energy (PE) (Supplementary Methods). The relevance of each feature and channel in the prediction method was further evaluated using Shapley additive explanation (SHAP) values (42). Through the SHAP values and the KNN accuracy, it was determined that PH was the most important feature for accurate gesture classification (Fig. 3F and fig. S31). When considered alongside PH, SD and PA both increased the classification accuracy, with SD being the most beneficial (figs. S31 and S32). In terms of channels, it was found that three EMG channels were sufficient to provide a high gesture accuracy of 96.31 ± 1.25%. Adding a fourth channel was beneficial but not statistically significant (tables S2 and S3).

With the KNN algorithm, the robot can imitate the user’s gesture in millisecond-level time for automatic object manipulation. The data acquisition and signal processing time delay to determine a gesture were around 200 ms, well below the required time for optimal real-time robotic control (43). This was achieved using a sampling rate of 534 Hz and analyzing the data in batches of 100 points. The M-Bot’s time delay was substantially reduced by training the KNN model on only half of any sEMG signal for gesture recognition. By reducing the required data needed to determine a gesture, the machine learning model was able to predict the movement almost immediately after the gesture was complete.

The AI-powered e-skin-H–enabled gesture recognition provides a framework for online multidirectional robotic control with high-accuracy remote object manipulation (as illustrated in Fig. 3, G to I, and movie S2). After object contact, recognition, and positive threat detection, tactile and alarm feedback can be activated to inform the user of any potential danger using a pulsed current load between the two stimulation electrodes (Fig. 3I). To facilitate safe robotic handling and to protect e-skin-R from uncontrolled collisions, a laser proximity sensor was integrated into the robotic hand to reduce the actuation speed as the hand approaches a barrier (<10 cm) (Fig. 3K and fig. S33).

Evaluation of the M-Bot in human-interactive robotic physicochemical sensing

With delicate and precise control, the human-machine interactive M-Bot was successfully evaluated for fingertip point-of-use robotic TNT detection (fig. S34 and movie S3). The multimodal sensor data could be captured in real time using a portable multichannel potentiostat, wirelessly transmitted to the mobile phone, and displayed on the cellphone app (fig. S34 and movie S3). The M-Bot was also able to perform object grasping and multispot tactile and chemical sensing (Fig. 4, A to D, and movie S4). Multiplexed physicochemical data were simultaneously recorded and automatically processed without signal interferences (fig. S35). In an example demonstration, seven AI-assisted gesture-controlled steps were used in sequence to control the robotic hand as it approached, grasped, and released a spherical object (Fig. 4E, fig. S36, and movie S4). In parallel, five sensor arrays were activated, displaying multiplexed tactile readings and surface TNT levels (Fig. 4, F and G).

The use of the M-Bot for multiplexed physicochemical robotic sensing was further evaluated on an OP–contaminated cylindrical surface (Fig. 4H). During the experiment, 14 sensor arrays on e-skin-R were activated. The tactile and OP sensor responses from each sensor, along with the corresponding color mapping of their distributions across the three-dimensional (3D) surface, are displayed in Fig. 4 (I and J, respectively) (detailed data are demonstrated in figs. S37 and S38). We anticipate that by further increasing the number and density of the multimodal sensor arrays, more accurate and informative data can be obtained from arbitrary objects and surfaces.

Evaluation of an e-skin–enabled M-Boat for autonomous source tracking

The multimodal robotic sensing platform was further generalized onto an autonomous robotic boat capable of tracking pollutants, explosives, chemical threats, and biological hazards for risk prevention and mitigation, which is an important topic in civil security (7, 44). In this regard, our printed multimodal e-skin–R technology was adapted onto a multimodal sensing robotic boat (M-Boat) for real-time hazard detection and to autonomously locate the source of water-based chemical leakages (Fig. 5A). 3D printed from simple computer-aided designs, the M-Boat contains an inkjet-printed multimodal sensor array with one temperature and three chemical sensors, two electrical motors (for boat propulsion and steering), and a printed circuit board (PCB) for data collection, signal processing, and motor control (Fig. 5, B to D, and fig. S39). The propulsion of the M-Boat can be precisely controlled by adjusting the individual duty cycle of pulsed voltages supplied to each motor (Fig. 5E, fig. S40, and movie S5). For source detection, an A* search algorithm (45) was implemented for autonomous decision-making while searching for the maximum concentration of the chemical leakage (Fig. 5, F and G, fig. S41, and Supplementary Methods). At each decision point, the sensors can detect small traces of the chemical leak in three equidistant locations around the boat. With this input, the algorithm calculates the optimal direction to travel using the gradient vector, indicating the direction of the highest concentration, and a heuristic estimate of the diffusion based on an interpolated map from previous points. By using the heuristic map in parallel with the gradient, the algorithm takes advantage of both the past and present results to precisely predict the spatial location of the source. The performance of the M-Boat was evaluated through simulations and experimentally in water tanks containing various chemical gradients induced by a low pH-corrosive fluid (Fig. 5, H and I, fig. S42, and movie S6) and OP leakage (Fig. 5, J and K). In the water tank, the M-Boat performed real-time detection of the surrounding analyte concentrations, automatically adjusting its trajectory based on the A* algorithm, to successfully identify the leakage source. The M-Boat was also able to perform continuous hazard analysis and autonomous leakage tracking in seawater (fig. S43 and movie S6). The surrounding pH and ionic strength of a real-world sample matrix (e.g., lake water or seawater) did not show substantial influence on the sensor performance (fig. S44). When necessary, more real-time calibration mechanisms for precise hazard analysis can be introduced by incorporating more related biosensors (e.g., pH and...
conductivity sensors) into the e-skin-R. With more advanced robotic control and sensing designs, the M-Boat platform could serve as an important basis for intelligent path planning and decision-making of autonomous vehicles.

DISCUSSION
Here, we have described a human-machine interactive e-skin-based robotic system (M-Boat) with multimodal physicochemical sensing capabilities. The mass-producible flexible sensor arrays allow for high-performance on-site monitoring of temperature, tactile pressure, and various hazardous chemicals (in both dry phase and liquid phase) such as explosives, OPs, and pathogenic proteins. The integration of such multimodal sensors onto a robotic e-skin platform provides autonomous systems with interactive cognitive capabilities and substantially broadens the range of tasks that robots can perform, such as combating infectious diseases like COVID-19.

Existing robotic sensing technologies are largely limited to monitoring physical parameters such as temperature and pressure. To achieve high-performance chemical sensing, nanomaterials are commonly used via manual drop-casting methods, which could lead to large sensor variations. Moreover, most electrochemical sensing strategies require detection in aqueous solutions, making them impractical for dry-phase robotic analysis. Now, there are no reported scalable low-cost manufacturing approaches to prepare robotic physicochemical sensors. In this work, we proposed a scale solution to fabricate flexible, multifunctional, and multimodal sensor arrays prepared entirely by high-speed inkjet printing. Custom-developed functional nanomaterial inks are designed and optimized to achieve highly sensitive and selective sensors for the specific hazardous target analytes. The hydrogel-coated printed nanobiosensors allow for efficient dry-phase chemical sampling and rapid on-site hazard analysis on a robotic platform.

Manufactured using the same approach, e-skin-H ensured stable contact with the soft human skin for reliable recording of neuromuscular activity to facilitate remote robotic sensing and control. To minimize the amount of data collected and analyzed for human-robotic interaction, AI and smart algorithms were applied to decode incoming information and efficiently predict and control robotic movement. An in-depth analysis into each sEMG channel’s individual

Fig. 4. Evaluation of the M-Boat in human-interactive robotic physicochemical sensing. (A to D) Time-lapse images of the human-interactive robotic control for object grasping and on-site TNT detection. Scale bars, 5 cm. (E to G) The sEMG data collected in real time, which allow the robotic hand to approach and grasp a spherical object (E) and the corresponding tactile (F) and TNT (G) sensor responses. Insets in (F) and (G) represent the colored mapping of pressure and TNT distributions on the object. (H to J) Photograph of the robotic OP sensing on a cylindrical object (H) and the corresponding responses of the tactile sensors (I) and OP sensors (J) on an e-skin-R. Insets in (I) and (J) represent the color mappings of pressure and OP distributions on the object. Scale bar, 5 cm.
contribution to the machine learning model was presented, allowing future researchers to optimize the number of electrodes needed for robotic control. Using the SHAP analysis, we further untangled the hidden overlapping information between each channel’s features and categorized which features present the most nonoverlapping information for gesture prediction. For the M-Boat, machine learning gesture prediction via e-skin-H was further coupled with user-interactive tactile and threat alarm feedback that allow seamless human-machine interaction for the remote deployment of robotic technology in extreme or contaminated environments. To obtain such real-time results, the robotic platform’s data acquisition, signal processing, feature extraction, and gesture prediction of the sEMG signals were performed with millisecond-level time after the gesture was complete. The M-Boat similarly used a smart A* algorithm for autonomous source tracking, minimizing the boat’s path, and subsequently, time and energy, in finding potentially hazardous chemical leaks. In these applications, the systems demonstrated real-time autonomous movement, all within a low-cost mass-producible system, lowering the barrier for real-time robotic perception.

This human-machine–interactive robotic sensing technology represents an attractive approach to develop advanced flexible and soft e-skins that can reliably collect vital data from the human body and the surrounding environments. Full system integration to achieve high-speed, wireless, and simultaneous multichannel physicochemical sensing is strongly desired for future field deployment and evaluation. Moreover, we envision that, by integrating a high density and new types of multimodal sensors, this technology could substantially enhance the perceptual capabilities of future intelligent robots and pave the way to numerous new practical wearable and robotic applications.

**MATERIALS AND METHODS**

**Materials**

Graphite flake was purchased from Alfa Aesar. Sodium nitrate, potassium permanganate, hydrogen peroxide, potassium hexacyanoferrate(III), citric acid, chloroplatinic acid, PDMS, zirconium(IV) chloride, aniline, gelatin, paraoxon-methyl, TNT solution, 4-nitrophenol, 2-nitrophenyl...
100 mg of NaNO₃ was added inside. Subsequently, 3 g of KMnO₄ was mixed with propylene glycol (80:20, v/v) to form the MOF-808 ink. A MOF-808 suspension in deionized (DI) water was prepared and naturally cooled to room temperature. The product was washed and kept at 120°C for 12 hours. After the reaction, the autoclave was opened and formic acid (1:0.56, v/v) solvent and sonicated for 20 min.

Preparation and characterizations of print inks
To prepare the Pt-graphene ink, GO was first prepared following a modified Hummer’s method (46). One gram of graphite flake was mixed with 23 ml of H₂SO₄ for more than 24 hours, and then 100 mg of NaNO₃ was added inside. Subsequently, 3 g of KMnO₄ was added below 5°C in an ice bath. After stirring at 40°C for another 30 min, 46 ml of H₂O was added while the solution temperature was slowly increased to 80°C. In the end, 140 ml of H₂O and 10 ml of HClO₃ were introduced into the mixture to complete the reaction. The GO was washed with 1 M HCl and filtered. After dried under vacuum at 60°C, a GO (2 mg ml⁻¹) suspension was prepared followed by sonication. Last, the suspension was mixed with 23 ml of H₂SO₄ for more than 24 hours, and then 10 ml of 1% BSA in PBS were dropped and incubated for 2 hours. Next, 10 μl of 1% BSA in PBS were dropped on the modified electrode and incubated for 1 hour to deactivate residual NHS esters. The modified sensors were stored in the refrigerator until use.

To assemble the robotic e-skin, the pins of the finger printed e-skin were connected with the bottom printed silver connections of palm part through a z axis conductive tape (3M), and then e-skin-R was set on a robotic hand printed with a 3D printer (Mars Pro, Elegoo Inc.).

Characterizations of the multimodal robotic sensing performance of e-skin-R
The printed biosensors were characterized with cyclic voltammetry (scan rate: 50 mV s⁻¹, unless otherwise noted), DPV, and amperometric current (i)-time (t) through an electrochemical workstation (CHI 660E). McIlvaine buffer solutions (pH 6.0) were used to prepare the analyte solutions. A commercial Ag/AgCl reference electrode (CHI111) was used for characterizing the printed sensing electrodes in the solution, whereas printed Ag solid-state electrodes were used for hydrogel-based sensor characterization (there was an ~0.1-V difference between these two types of reference electrodes in McIlvaine buffer). To quantify the electrochemical performance and the electrochemical surface areas, the print electrodes were tested in 5 mM K₃Fe(CN)₆ and 1 M KCl with scan rates of 5 mV s⁻¹ from −0.1 to 0.5 V.

For TNT and OP sensors, the conditions of nDPV measurements include a scan range of −0.15 to −0.5 V, an incremental potential of 0.004 V, a pulse amplitude of 0.05 V, a pulse width of 0.05 s, and a pulse period of 0.5 s. The reduction peaks of nDPV curves were extracted using a custom-developed iterative baseline correction.
algorithm. To prepare the electrolyte-loaded hydrogel for analyte sampling and sensing, 0.250 g of gelatin powder, 0.075 g of KCl, 0.071 g of citric acid, and 0.179 g of disodium phosphate were mixed in 10 ml of DI water and stirred at 80°C for 15 min. The hydrogel was stored and aged overnight. The gelatin electrolyte-loaded hydrogel was coated on the printed biosensors for dry blot detection.

For S1 protein detection, the modified electrode was incubated with 10 µl of S1 protein in PBS for 10 min, and the DPV measurements ranged from –0.1 to 0.5 V. The electrochemical signal of the sensor before and after antigen binding was measured in 5 mM K3Fe(CN)6. The difference between the peak current densities (ΔI) was obtained as sensor readout. A sampling hydrogel pad was prepared to demonstrate the feasibility for SAR-CoV-2 virus dry blot detection. To perform one-step detection, 10 µl of gelatin hydrogel [7.5 wt % gelatin, 10 mM K3Fe(CN)6, and 0.2 M phosphate buffer (pH 7.0)] was placed onto a dry S1 protein blot [from 10 µl of a SARSCoV-2 S1 protein droplet (1 µg ml–1)]. Such amount of S1 protein could potentially be found in the saliva droplet of a patient with COVID-19 (47). For dry-phase sensing selectivity study, the dry protein blots were created with the same amount of interference proteins (10 µl, 1 µg ml–1). The electrochemical signal of the gel was recorded immediately and 10 min after joining the biosensor with the gel.

The temperature sensor characterization was performed on a ceramic hot plate (Thermo Fisher Scientific), and an amperometric method (with an applied voltage of 2 V) was used to detect the temperature response. The piezoresistive tactile sensor characterization was also applied with a constant voltage of 2 V to record the current response under various pressure loads.

The scanning electron microscopy (SEM) images of the electrodes were obtained by a field-emission SEM (FEI Nova 600 NanoLab). Energy-dispersive spectroscopy (EDS) mapping were obtained by an EDS spectrometer (Bruker Quantax EDS).

Fabrication and assembly of e-skin-H
The fabrication process of e-skin-H was illustrated in fig. S26. A 2-min O2 plasma surface treatment was performed with Plasma Etch PE-25 to enhance the surface hydrophilicity of the PI substrate. Silver interconnects were printed with DMP-2850. The PI substrate without the printed patterns were removed with laser cut using a 50-W CO2 laser cutter (Universal Laser Systems). The optimized laser cutter parameters were power of 10%, speed of 80%, and pixels per inch (PPI) of 1000 in vector mode. After cleaned with ethanol and dried, the remaining patterns were transferred onto a 70-µm thick PDMS substrate and then encapsulated with another layer of PDMS film as well (with SEMG and electrical stimulation electrodes exposed). An adhesive electrode gel (Parker Laboratories Inc.) was spread onto the electrodes before placing on human participants.

Evaluation of the human-machine interactive multimodal sensing robot
To evaluate the performance of the M-Bot, the e-skin–R–interfaced 3D printed robotic hand was assembled onto a five-axis robotic arm (Innoffs Ltd.). The e-skin-H was then set around a human participant’s forearm after cleaning the skin with alcohol swabs. The SEMG data were acquired with four channels (three SEMG electrodes in each channel) through an open-source hardware shield (Olimex). The signals were sampled as integers between 0 and 1023 by a 10-bit analog-to-digital converter (ADC) and then processed through a serial (cluster communication) port. Each channel was then scaled back into voltages between 0 and 5 V. Although the robotic arm control was performed in real time, data processing was performed asynchronously to signal acquisition. During processing, the data were first put through a high-pass filter with a cutoff frequency of 100 Hz. The points were subsequently downsized using an RMS filter (batch size: 400 points and step size: 10 points). The peaks detected after processing were used as features for the machine learning model. Overlapping peaks from each channel (peaks within a half-peak width away) were categorized as a single group. If multiple peaks were detected in a single channel’s set, then the first peak was used. The KNN training model was built using 60 samples per each of the six gestures. The training and testing datasets were divided 2:1, respectively, and were randomly selected using an equal representation of each gesture. After the model was developed, it was further evaluated for accuracy using new data from each gesture.

The validation and evaluation of the M-Bot were performed using human participants in compliance with all the ethical regulations under protocols (ID 19-0895) that were approved by the Institutional Review Board at the California Institute of Technology. Three participants were recruited from the California Institute of Technology’s campus and the neighboring communities through advertisement. All participants gave written informed consent before study participation.

Machine learning data analysis
For each gesture, all five features were extracted from the associated peak in the RMS-filtered EMG data: height, average area, SD, average energy (intensity), and maximum slope. The features extracted were calculated in reference against their baselines, which were determined via a binary search of the previous data in 50-ms intervals.

After feature extraction, SHAP values were used to evaluate the performance enhancement of each feature extracted and EMG channel used. In addition to SHAP values, the average testing accuracy across 5000 training sessions was taken for each permutation of features and EMG channels, which supplemented the SHAP values in providing further insight into which channels and features contained nonoverlapping beneficial information for gesture determination. For each of the 5000 trials, the testing points represented 33% of the dataset, with each gesture in the test set being proportionally represented in the full dataset. For the arm EMG dataset, this amounted to 387 movements split across six gestures; of those points, the KNN model was fit using 257 training points and scored on the remaining 128 testing points (testing and training were proportionally stratified across all six gestures).

Evaluation of the M-Boat
To evaluate the performance of the M-Boat, the e-skin–R was assembled onto a 3D printed boat with a four-layer PCB, as shown in Fig. 5 (B and C). On the PCB, a Bluetooth low-energy (BLE) module (CYBLE-222014-01, Cypress Semiconductor) was used for controlling the electrochemical front end through a serial peripheral interface (SPI). This module was also used to control the motor...
driver through general purpose input/output (GPIO) pins and pulse width modulation (PWM) and to transmit data over BLE. An electrochemical front end (ADS941, Analog Devices) was set up via SPI to perform multiplexed electrochemical measurements with the sensor arrays and to send the acquired data to the BLE module for signal processing and BLE transmission. A BLE dongle (CY5677, Cypress Semiconductor) was used to establish a BLE connection with the M-Boat and to securely receive the sensor data via BLE indications. An A* algorithm was used to analyze the sensor data and compute the next M-Boat’s movement path with the optimal motor speed. The calculated motor speed information was sent back to the BLE module in real time for the pulse width–modulated control of two motors (QSL2BQ280001) through a dual dc motor driver (TB6612FNG, Toshiba). The entire system was powered by a 3.7-V Li-ion battery (40 mAh).

For the OP chemical threat tracking experiment, a natural diffusion gradient was created by 10 droppings of 20 μL of 0.1 M OP into a 0.1 M NaCl solution tank. The seawater samples were collected from the Pacific Ocean in Los Angeles. The M-Boat was set into the tank after 30 min. For the corrosive acidic threat tracking experiment, pH sensors were modified on e-skin-R instead. Briefly, a polyaniline pH-sensitive film was electropolymerized on the IPCE in a solution containing 0.1 M aniline and 0.1 M HCl using a CV from −0.2 to 1 V for 25 cycles at a scan rate of 50 mV s⁻¹. Then, 100 μL of H₂SO₄ (2 M) as the leakage source was dropped into the middle of the water tank. Last, the M-Boat was set after 45/30 min with/without barriers in the tank, respectively.

### Statistical analysis

All quantitative values were presented as means ± SD of the mean. For all sensor evaluation plots, the error bars were calculated on the basis of the SD from three sensors. For the hydrogel stability study, the error bars were calculated on the basis of the SD from three independent measurements. For the machine learning analysis of the sEMG data, the model was trained on the same data across 5000 trials of randomly splitting the points between training and testing data. The accuracy profile of this training was then fit to a skewed normal distribution, where the mean was extracted.

**SUPPLEMENTARY MATERIALS**

www.science.org/doi/10.1126/scirobotics.abm0495

Methods

- Figs. S1 to S44
- Tables S1 to S3
- Movies S1 to S6

View/request a protocol for this paper from Bio-protocol.

### REFERENCES AND NOTES


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Data and materials availability: All data needed to support the conclusions of this manuscript are included in the main text or Supplementary Materials. The code for this study is available at https://github.com/Samwich1998/Boat-Search-Algorithm (M-Boat search algorithm).

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All-printed soft human-machine interface for robotic physicochemical sensing
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